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Artificially Engineered Exction Quantum Dot Arrays for Quantum Information Science
Applications

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14. ABSTRACT
This proeject focused on the following research activities:
(a) Exploring MBE/CVD growth of monolayer TMDs and investigate the interlayer exciton diffusion in TMDs heterostructures
(b) STM/S investigations of honeycomb BeO on epitaxial Ag films
(c) Optical spectroscopy investigation of the exciton binding energy of TMDs in different dielectric environments.
(d) Momentum space investigations of a wafer scale monolayer TMD semiconductors and heterostructures using ARPES
(e) Development of time-resolved ARPES to investigate excited state properties of quantum materials including monolayer TMD
(f) Optical spectroscopy investigation of TMD bilayer systems to determine interlayer coupling energy at different critical points in the Brillouin zone.
(g) Optical spectroscopy studies of MoSe2/MoS2 hetero-bilayers with twist angle dependent intra-layer excitons and inter-layer excitons

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AFOSR Deliverable Reporting Format:
Final and Interim Reports

This document has been developed to provide Principal Investigators (PIs), co-PIs, and research organizations with:

- A listing of the questions that will be asked in the new AFOSR project reporting format;
- Assistance in planning for the submission of the report

Overview: There are two main sections of the AFOSR Deliverable Report. Section 1 is filled out in Qualtrics, and Section 2 is uploaded by PDF.

- Section 1: Structured Survey Questions in Qualtrics
  - This section captures information in a structured survey format as required by the Research Performance Progress Report Format (RPPR) guidance. Information in this section will include publications, participants, and other intellectual property questions. All questions in this section will be asked within Qualtrics.

- Section 2: Technical Report PDF
  - This section captures unstructured technical information not captured in the above section. PI's will upload PDF reports that contain information on awards, changes to scope, and other technical updates. This PDF upload will be very similar to previous AFOSR report uploads. Please contact your individual program officer if you have further questions about what should be contained in this report.

***Note: The information being asked in this deliverable report is explicitly defined by the official RPPR guidance which can be found here: http://www.nsf.gov/bfa/dias/policy/rppr/index.jsp. We have automated several questions to make the reporting less burdensome on the principal investigator. Your report link, found in the deliverable reminder email, contains individualized information that is specific to your deliverable report. You may edit this information if anything appears to be incorrect.

Section 1: Structured Survey Questions

Award Information

- Award Number (Federal Award Identification Number FA2386-18-1-4097
- Report Type: Final report
- Principal Investigator: Chih-Kang Shih
- Principal Investigator Email: shih@physics.utecas.edu
- Principal Investigator Phone: 512-471-6603
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- Recipient Organization: The University of Texas at Austin
- Business Office Email:
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- Report Period Start Date: September 1, 2018
- Report Period End Date: August 31, 2021
- Current Program Officer: Jeremy Knopp
- Please list any other Co-Program Officers (if applicable):

Publications

[1] Lee, W., Lin, Y., Lu, L. S., Chueh, W. C., Liu, M., Li, X., ... & Shih, C. K. (2021). Time-resolved ARPES Determination of a Quasi-Particle Band Gap and Hot Electron Dynamics in Monolayer MoS2. Nano Letters, 21(17), 7363-7370. *DOI: 10.1021/acs.nanolett.1c02674*

[2] Lee, W., Lu, L. S., Chang, W. H., & Shih, C. K. (2021). Momentum-Resolved Electronic Structures of a Monolayer-MoS2/Multilayer-MoSe2 Heterostructure. The Journal of Physical Chemistry C, 125(30), 16591-16597. *DOI: 10.1021/acs.jpcc.1c04219*

[3] Zhang, H., Holbrook, M., Cheng, F., Nam, H., Liu, M., Pan, C. R., ... & Shih, C. K. (2021). Epitaxial growth of two-dimensional insulator monolayer honeycomb BeO. ACS nano, 15(2), 2497-2505. *DOI: 10.1021/acsnano.0c06596*

[4] Wang, C. Y., Sang, Y., Yang, X., Raja, S. S., Cheng, C. W., Li, H., ... & Shi, J. (2020). Engineering giant Rabi splitting via strong coupling between localized and propagating plasmon modes on metal surface lattices: observation of $\sqrt{N}$ scaling rule. Nano letters, 21(1), 605-611. *DOI: 10.1021/acs.nanolett.0c04099*

[5] Lee, W., Pan, C. R., Nam, H., Chou, M. Y., & Shih, C. K. (2020). Critical role of parallel momentum in quantum well state couplings in multi-stacked nanofilms: An angle resolved photoemission study. AIP Advances, 10(12), 125211. *DOI: 10.1063/5.0022706*

[6] Hsu, W. T., Lin, B. H., Lu, L. S., Lee, M. H., Chu, M. W., Li, L. J., ... & Shih, C. K. (2019). Tailoring excitonic states of van der Waals bilayers through stacking configuration, band alignment, and valley spin. Science advances, 5(12), eaax7407. *DOI: 10.1126/sciadv.aax7407*

[7] Hsu, W. T., Quan, J., Wang, C. Y., Lu, L. S., Campbell, M., Chang, W. H., ... & Shih, C. K. (2019). Dielectric impact on exciton binding energy and quasiparticle bandgap in monolayer WS2 and WSe2. 2D Materials, 6(2), 025028. *DOI: 10.1088/2053-1583/ab072a*

[8] Choi, J., Hsu, W. T., Lu, L. S., Sun, L., Cheng, H. Y., Lee, M. H., ... & Chang, W. H. (2020). Moiré potential impedes interlayer exciton diffusion in van der Waals heterostructures. Science advances, 6(39), eaba8866. *DOI: 10.1126/sciadv.aba8866*

Participants

Chih-Kang Shih
Email: shih@physics.utexas.edu
Organization: The University of Texas at Austin
Most Senior Project Role: PD/PI
Nearest Person Month Worked: 1
Contribution to the Project: PI plans, coordinates and supervises graduate students and postdocs working on different aspects of this project. He also forges the collaboration relationship within the institution and outside the institution
Funding Support: AFOSR
International Collaboration: Yes, Taiwan
International Travel: Yes, Taiwan - 0 years, 2 months, 7 days

Wei-Ting Hsu
Email: wthsu53@gmail.com
Organization: The University of Texas at Austin
Most Senior Project Role: Postdoctoral (scholar, fellow or other postdoctoral position)
Nearest Person Month Worked: 12
Contribution to the Project: Dr. Hsu's role in this project focuses primarily on Laser spectroscopy investigations of the optical properties of TMD heterostructures.
Funding Support: Dr. Hsu receives partial support from this grant. His primary financial support comes from a fellowship program of the ministry of science and technology, Taiwan.
International Collaboration: Yes, Taiwan
International Travel: No

Chun-Yuan Wang
Email: roberts900821@gmail.com
Organization: The University of Texas at Austin
Most Senior Project Role: Postdoctoral (scholar, fellow or other postdoctoral position)
Nearest Person Month Worked: 3
Contribution to the Project: Dr. Wang role in this project entails two aspects (a) fabrication of TMD heterostructures using exfoliation and stacking, and other nanofabrication techniques such as FIB; (b) Laser spectroscopy investigations of the optical properties of TMD heterostructures
Funding Support: This grant, NSF grand and a DOD grant
International Collaboration: No
International Travel: No

Madisen Holbrook
Email: madisen.holbrook@gmail.com

Organization: The University of Texas at Austin
Most Senior Project Role: Graduate Student (research assistant)
Nearest Person Month Worked: 3
Contribution to the Project: Ms. Holbrook works on MBE growth and UHV STM characterization of 2D electronic materials. In addition, she also study ex-situ grown 2D electronic materials by cleaning them in ultra-high vacuum. She carries out experiment, analyzing data, and prepare the manuscript under the guidance of Professor Shih. In addition, she collaborate with other members (postdoc and other graduate student) in the lab, as well as external collaborators.
Funding Support: This grant, NSF grant, and partial support from the UT MRSEC program
International Collaboration: No
International Travel: No

Woojoo Lee
Email: fstbwj@gmail.com
Organization: The University of Texas at Austin
Most Senior Project Role: Graduate Student (research assistant)
Nearest Person Month Worked: 6
Contribution to the Project: Mr. Lee works on MBE growth and Angle Resolved Photoemission (ARPES) investigations of 2D electronic materials. In addition, he also studies ex-situ grown 2D electronic materials using CVD (by our collaborator) when such samples can be cleaned in ultra-high vacuum. He develops time-resolved ARPES (outside the scope of this project) supported by the MRSEC program at UT. He carries out experiment, analyzing data, and prepare the manuscript under the guidance of Professor Shih. In addition, he collaborates with other members in the lab, as well as external collaborators.
Funding Support: In addition to this grant, Mr. Lee is also supported by the MRSEC program at the University of Texas and NSF grant
International Collaboration: Yes, Taiwan
International Travel: No

Mengke Liu
Email: mkliu@utexas.edu
Organization: The University of Texas at Austin
Most Senior Project Role: Graduate Student (research assistant)
Nearest Person Month Worked: 6
Contribution to the Project: Ms. Liu's role in this project primarily focuses on low temperature (down to 2 Kelvin) STM investigations of 2D materials. She is in charge of the high resolution STM/S instrumentation, primarily dedicated for investigation of superconducting materials. Very low temperature high resolution STM/S sometimes becomes necessary for us to understand the electronic

structure better. Although she plays more of a supporting role for this project, her contribution is quite important for some of our recent investigations.

Funding Support: In addition to this grant, she is supported by another NSF grant and a grant from the Welch foundation.
International Collaboration: No
International Travel: No

Other Partners or Collaborators

Institute of Atomic and Molecular Sciences, Academia Sinica
Organization Type: Academic Institution
Organization Location: Taipei, Taiwan
Partner's Contribution to the Project: Collaborative Research
More Detail on Partner and Contribution: We have been collaborating with Professor Mei-Yin Chou of the IAMS, Academia Sinica on 2D electronic materials for many years, starting from her tenure at Georgia Tech and continuing until now. Her expertise is first principle electronic structure calculation using density functional theory.

National Yang Ming Chiao-Tung University
Organization Type: Academic Institution
Organization Location: Hsin-Chu, Taiwan
Partner's Contribution to the Project: Collaborative Research
More Detail on Partner and Contribution: We are collaborating with Professor Wen-Hao Chang and Professor Hyeyoung Ahn's research groups on TMD heterostructure projects. The collaboration is extensive. They provide direct CVD growth of vertically stacked heterostructures and characterizations of the twist angles. Such twist angles are re-examined at Shih group at UT in addition to detailed optical spectroscopy investigations. Shih's group also carry out STM investigations of some of the samples grown at NYCU.

University of Hong Kong
Organization Type: Academic Institution
Organization Location: Hong Kong, China
Partner's Contribution to the Project: Collaborative Research
More Detail on Partner and Contribution: We collaborate with Professor Wang Yao and his students at the University of Hong Kong. Professor Yao's expertise is theory of optical properties of semiconductor nanostructures.

Taiwan Semiconductor Manufacturing Company (TSMC)
Organization Type: Industry Company
Organization Location: Hsinchu, Taiwan
Partner's Contribution to the Project: Collaborative Research
More Detail on Partner and Contribution: We collaborate with Dr. Lain-Jong Li on the 2D TMD sample preparation.

Inventions, patent applications, licenses, and technologies/techniques

Identify inventions, patent applications with date, and/or licenses that have resulted from the research. Submission of this information as part of an interim or final research performance progress report is not a substitute for any other invention reporting required under the terms and conditions of an award. You will still have to separately file the appropriate form 882 for your invention/patent.

- Patents
  Nothing to report
- Inventions
  Nothing to report
- Licenses
  Nothing to report
- Technologies or Techniques
  Nothing to report
- Other Products
  Nothing to report

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Accomplishments

The information provided in this section allows AFOSR to assess whether satisfactory progress has been made during this reporting period. The PI is reminded that the recipient organization is required to obtain prior written approval from the awarding agency grants official whenever there are significant changes in the project or its direction. See agency specific instructions for submission of these requests.

Research Activities:

We have been focusing on the following research activities
(a) Exploring MBE/CVD growth of monolayer TMDs and investigate the interlayer exciton diffusion in TMDs heterostructures
(b) STM/S investigations of honeycomb BeO on epitaxial Ag films
(c) Optical spectroscopy investigation of the exciton binding energy of TMDs in different dielectric environments.
(d) Momentum space investigations of a wafer scale monolayer TMD semiconductors and heterostructures using ARPES
(e) Development of time-resolved ARPES to investigate excited state properties of quantum materials including monolayer TMD
(f) Optical spectroscopy investigation of TMD bilayer systems to determine interlayer coupling energy at different critical points in the Brillouin zone.
(g) Optical spectroscopy studies of MoSe2/MoS2 hetero-bilayers with twist angle dependent intra-layer excitons and inter-layer excitons.

Research Objectives:

There are several objectives for our research activities.
(a) First, we are aiming at determination of interlayer coupling in vdW bilayers as a function of interlayer distance and in-plane atomic alignments. Accurate determination of

these quantities will provide researchers in the field with powerful designing parameters for tailoring the functionality of vdW heterobilayers.

(b) Second, we are exploring the exciton binding energy under different dielectric environment. Which may play an important role for the design of van der Waals heterostructures and TMD-based optoelectronic devices.

(c) Third, exciton diffusion represents an important channel of energy transport in transition metal dichalcogenides (TMDs). We experimentally demonstrate the rich phenomena of interlayer exciton diffusion in TMDs heterostructures by tuning the twist angle of the heterostructure. provide critical guidance for intensive efforts on searching for IX BEC or quantum emitters in TMD heterostructures.

(d) Fourth, we are exploring epitaxial growth of new phases of 2D monolayers that do not exist in the bulk form. In the monolayer limit, the interaction with the substrate may stabilize certain phase by lowering the free energy in the single layer or a few-layer limit. The new phases will undoubtedly lead to new electronic structures which will expand the tool set for designing new functionalities for 2D electronic materials

(e) Finally, we are exploring a novel approach to create wafer scale monolayer TMDs with well aligned in-plane orientation. Such a platform will allow use to apply advanced characterization techniques such as ARPES and time-resolved ARPES to investigate momentum resolved ground/excited state electronic structures. Moreover it can provide a scalable technological platform.

Research Accomplishments

(a) We have used optical spectroscopy to determine the interlayer coupling in vdW hetero-bilayers (MoS2/WS2; MoSe2/WSe2) and homobilayers (MoS2).

In the case of heterobilayer systems, we have focused primarily on the interlayer coupling at K-valley as that is currently a topic received a lot attention, especially in the subject of moire excitons. We have unveiled the role of stacking configuration, band alignment, and valley spin in the determination of K-valley coupling strength. In addition, we demonstrate the ability to create a new class of excitons in hetero- and homobilayers that combines advantages of monolayer and interlayer excitons, i.e., featuring both large optical and electric dipoles. These excitons consist of an electron confined in an individual layer, and a hole extended in both layers, where the carrier-species-dependent layer hybridization can be controlled through rotational, translational, band offset, and valley-spin degrees of freedom. We observe different species of layer-hybridized valley excitons, which can be used for realizing strongly interacting polaritonic gases and optical quantum controls of bidirectional interlayer carrier transfer. The result has been disseminated through a publication in Science Advances in Dec 2019.

More recently, we have extended this study into a regime where we can control the interlayer spacing using a diamond cell. The diamond cell allows use to apply hydostatic pressure upto ~ 20 GPa while allowing optical spectroscopy to be performed under pressure. The interlayer spacing is pressure dependent which had been calibrated previously by our collaborator. Using this pressure cell, we can vary the interlayer

spacing by about 7%. We show that the interlayer coupling strength is an exponential function of the interlayer spacing and determine the decay length at different critical points in the Brillouin zone. More significantly, by reducing the layer spacing by 7%, we are able to enhance the K-valley coupling strength upto 300%. This demonstration illustrates the possibility to tailor the moire potential in the current topic of moire superlattice to an energy scale exceeding room temperature thermal energy.
In addition to determination of interlayer coupling at K-point, we have extended the investigations to unveil the interlayer coupling at Gamma point an Q point as a function of the interlayer spacing.
We have recently submitted a paper entitled "Quantitative determination of interlayer electronic coupling at various critical points in bilayer MoS2" to Physical Review Letters.

(b) We investigated monolayer WS2 and WSe2 by exciton Rydberg spectroscopy. Excitons, bound electron–hole pairs in a 2D plane, dominate the optical properties of monolayer transition metal dichalcogenides (TMDs). A large exciton binding energy on the order of 0.5 eV was theoretically predicted and experimentally determined recently. These ultrastable excitons thus open an avenue to explore the exciton physics such as Bose–Einstein condensation and superfluidity at room temperature. In order to utilize 'dielectric tuning' of the exciton binding energy and quasiparticle band gaps, one must use insulating dielectrics. We investigated the impact of insulating dielectric environments on the exciton binding energy of monolayer WS2 and WSe2 by exciton Rydberg spectroscopy, in which the dielectric environment is systematically varied from $\kappa = 1.49$ to 3.82. We found that, with increasing κ value, the exciton binding energy and quasiparticle bandgap exhibit significant reductions. Quantitatively, our result follows the prediction of nonlocally-screened Keldysh potential very well. The fitted 2D polarizability, χ_{2D}, agrees rather well with previous density function theory calculations. Their close agreement validates the nonlocally screened Keldysh model which can be used to quantitatively predict the exciton binding energy for monolayer TMDs (and possibly other 2D materials) in different dielectric environments. Such a predictive model will play an important role for the design of van der Waals heterostructures and TMD-based optoelectronic devices. This work was published on 2D Material in March 2019.

(c) We experimentally demonstrate the rich phenomena of interlayer exciton diffusion in WSe2/MoSe2 heterostructures by comparing several samples prepared with chemical vapor deposition and mechanical stacking with accurately controlled twist angles.
The properties of van der Waals heterostructures are drastically altered by a tunable moiré superlattice arising from periodically varying atomic alignment between the layers. Exciton diffusion represents an important channel of energy transport in transition metal dichalcogenides (TMDs). While early studies performed on TMD heterobilayers suggested that carriers and excitons exhibit long diffusion, a rich variety of scenarios can exist. In a moiré crystal with a large supercell and deep potential, interlayer excitons may be completely localized. As the moiré period reduces at a larger twist angle, excitons can

tunnel between supercells and diffuse over a longer lifetime. The diffusion should be the longest in commensurate heterostructures where the moiré superlattice is completely absent.
Moiré potential has been demonstrated to have a profound influence on the electronic and optical properties of vdW heterostructures. However, its role in exciton diffusion has been largely ignored in previous studies on hBLs consisting of TMD monolayers with rather different lattice constants or with larger twist angles (16, 17, 20). Our experiments present a complementary view of IX diffusion from previous studies and highlight the influence of the moiré potential on exciton diffusion in hBLs. These studies provide critical guidance for intensive efforts on searching for IX BEC or quantum emitters in TMD heterostructures. This work has been published on Science Advance in Sep 2020.

(d). In terms of epitaxial growth of new phases of 2D monolayers, two systems are particularly noteworthy. The first one is the discovery of honeycomb BeO on epitaxial Ag. BeO is a polar wurtzite structure material with remarkable physical and mechanical properties, such as a high thermal conductivity, hardness, and a large insulating bandgap of 10.7 eV. These attractive properties have inspired significant theoretical research interest concerning the existence of other possible structural phases of BeO. In particular, as a member of the isoelectronic series of first row elements like graphene and hBN, studies predicted BeO might also exist in a sp2 hybridized atomic layer with a honeycomb structure (h-BeO). Later, monolayer graphitic h-BeO was predicted to be energetically stable in free space, and that it could be synthesized. Though h-BeO is predicted to exist at the ultra-thin limit, synthesis of this intriguing material has remained elusive. In this work, we report the realization of the epitaxial growth of a single atomic sheet of h-BeO grown on Ag(111) substrates using molecular beam epitaxy (MBE). We have used a set of characterization techniques to determine the atomic structure, long range crystallinity, and the bandgap of this novel honeycomb BeO. This work has been published in ACS Nano Feb, 2021.

(e) Recently, CVD growth of TMD materials has experienced significant progress in producing s shown to be able to grow in-plane aligned TMD monolayers on sapphires. Taking advantage of recent advancements, here we create a wafer-size MoS2 monolayer on graphite, with well-aligned lateral orientation for advanced electron spectroscopy studies. Low energy electron diffraction and scanning tunneling microscopy (STM) demonstrate atomically clean surfaces with in-plane crystalline orientation. The ground state and excited state electronic structures are probed using scanning tunneling spectroscopy (STS), angle-resolved photoemission (ARPES) and time-resolved (tr-) ARPES. In addition to mapping out the momentum-space quasiparticle band structure in the valence and conduction bands, we unveil ultrafast excited state dynamics, including inter- and intra-valley carrier scattering and a rapid downward energy shift by ~ 0.2eV lower than the initial free carrier state at sigma point. The work has been published in Nano Letter, August 2021.
In addition, we have also successfully transfer monolayer MoS2 onto cleaved MoSe2 and

created a ML-MoS2/multi-layer MoSe2 heterostructure. The electronic structure of this heterostructure has been probed using ARPES which reveal k-dependent interlayer coupling at different regions of Brillouin zone. The paper has been published in The Journal of Physical Chemistry, July 2021.

Key outcomes or Other achievements:

During the report period, we have 8 papers published. We have successfully developed time-resolved angle-resolved photoemission to probe the excited states of quantum materials.

- Dissemination of Research Results:
  We have disseminated our result primarily through (a) publications in refereed journals; (b) posting manuscripts on arxiv; (c) giving presentation in national and international conferences; (d) senior personnel giving lectures in major research universities; and (e) internal UT presentation in journal clubs and seminars.

Impacts

This component is used to describe ways in which the work, findings, and specific products of the project have had an impact during this reporting period. Describe distinctive contributions, major accomplishments, innovations, successes, or any change in practice or behavior that has come about as a result of the project. You can report on the following impact categories, but you are not required to report on all categories. Please only report in the categories that are relevant to your project:

Development of the principal discipline(s) of the project

Accomplishing our research goal on the determination of how interlayer electronic coupling depends on the interlayer atomic alignment in vdW bilayers have important impacts on the general field of condensed matter physics as well as the general research field of 2D electronic materials.

Other disciplines:

Interlayer coupling is the most important designing parameter to design novel electronic structures of vdW heterostructures. Our quantitative determination of Interlayer coupling as a function of interlayer atomic alignment and interlayer spacing allow the researchers in the field of 2D electronic materials powerful designing tool to tailor novel electronic structures as well as novel 2D electronic devices.

Describe the impact in this reporting period on the development of human resources

For example, how has the project provided opportunities for research and teaching in the relevant fields; improved the performance, skills, or attitudes of members of underrepresented groups that will improve their access to or retention in research, teaching, or other related professions; provided scholarships; provided exposure to science and technology for practitioners, teachers, young people, or other members of the public?

Students and postdocs working under this project receive broad training in materials growth and materials characterizations. They also have excellent opportunity to work with materials theorists to gain valuable experiences in interdisciplinary research.

Describe the impact on teaching and educational experiences

Nothing to report

Describe the impact in this reporting period on physical, institutional, and information resources that form infrastructure.

Our development of molecular beam epitaxy for 2D electronic materials impact the UT campus by providing an infrastructure for other researchers on UT campus.
In addition to the development of MBE systems for 2D electronic materials, the PI has also developed a new capability on UT campus – time resolved ARPES. The development of this facility is in collaboration with other MRSEC faculty members with the PI as the lead in this project. This facility impacts not only the MRSEC as a center but also impacts the campus wide effort in quantum materials research.

Impact on society beyond science and technology

The PI has been very active in recruiting and mentoring female graduate students in the Physics. For example, in the last 10 years, 50% of his Ph.D. mentees are female graduate students. Currently he is supervising 6 graduate students and four them are female. His effort has made important impacts in broadening participation of under-representative groups. In the past the PI also participate in educational outreach to high school students. But in 2020 and 2021 summer, this was suspended due to covid-19.

Changes

Changes in approach
Nothing to report

Problems or delays
Nothing to report

Expenditure Impacts
Nothing to report
Significant changes in the use or care of human subjects, vertebrate animals and/or biohazards
Nothing to report

Changes to the primary place of performance from that originally proposed
Nothing to report

Technical Updates

Nothing to report

Thank you for your support in helping AFOSR discover, shape and champion basic science research that profoundly impacts that future of the Air and Space Forces!