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Atomic Layer Semiconductors as Biological Analog Photocells for Intrinsically Regulated Light Harvesting

Nathaniel Gabor
REGENTS OF THE UNIVERSITY OF CALIFORNIA AT RIVERSIDE

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Atomic Layer Semiconductors as Biological Analog Photocells for Intrinsically Regulated Light Harvesting

Annual (Final) Performance Report August 2019 (Year 3)

In the following, I highlight the progress of AFOSR Biosystems Award number FA9550-16-1-0216. I first include a brief summary of the proposal, and in **Section A**, a summary of the research objectives of the original proposal. **Section B** describes the progress of the proposed research for 2018-2019, Year 3 of the proposal. In **Section C**, I record all publications from Year 1-3 and include a year-to-date list. I also include a list of awards and merits for the PI and student working on the proposed work.

Proposal Summary: Nature realizes a vast array of complex structures composed of molecular building blocks, the electronic structure of which can be well described by quantum mechanics. In photosynthetic light harvesting, quantum behavior has generated tremendous recent interest, yet the role of quantum coherence, a property of wave-like electronic states, remains controversial. Quantum coherence has been observed in various light harvesting pigment-protein complexes, however there exists little evidence that plants take advantage of these properties. We have established a new paradigm, based on internal thermodynamic fluctuations, which attempts to describe biological light harvesting without the need for quantum coherence. By understanding the connection between electronic structure and energy fluctuations, we predict the existence of an intrinsic regulation mechanism that emerges from quantum or molecular structure alone, which we discovered through the analysis of a two-channel quantum heat engine photocell.

To test our predictions, which we expect to be valid for a large class of nanoscale devices, we propose optoelectronic measurements that explore the fundamental photoresponse of intrinsic regulation in atomic layer semiconductor devices. These novel photocell devices, which mimic chlorophyll hetero-dimers of green plants, are composed of transition metal dichalcogenide heterostructures, and will allow the first demonstration of the key signatures of intrinsic regulation: control and enhancement of power conversion efficiency using multi-channel input optical power, suppression of internal power fluctuations, and the reduction of electronic and thermal energy dissipation. In developing the proposed photocells, we aim to establish that power conversion can be self-regulated by choice of quantum structure. In addition to informing new design strategies for ultra-efficient biophotonic and optoelectronic devices, the intrinsic regulation process may underscore the critical role played by molecular or quantum structure in the regulatory function of photosynthetic light harvesting photocells.

A. Summary of Objectives

Hypothesis. In both photovoltaics and photosynthesis, the power produced by a solar photocell changes dramatically in response to unpredictable light conditions, yet storing solar energy requires a steady, regulated flow of energy. We predict the existence of an intrinsic regulation mechanism that emerges from quantum or molecular structure alone, which we discovered through the analysis of a two-channel quantum heat engine photocell. Remarkably, we find - using the measured terrestrial solar spectrum - that this novel regulation process reproduces the absorption spectrum and molecular structure of photosynthetic organelle in green plants, suggesting a profound and so far un-established connection between fluctuating quantum systems and photosynthetic light harvesting.

Objectives. The objectives of this proposal are two-fold: First, we propose to fabricate and characterize atomically thin heterostructure optoelectronic devices that serve as biological analogs of the reaction centers of photosynthesis, in that they absorb and emit through very narrow energy bands and can be coupled together by direct charge transfer to form complex electronic systems. These transition metal dichalcogenide (TMD) heterostructures have recently been proposed to be novel solar energy harvesting materials due to high absorption efficiency, efficient charge transfer, and short interfacial channel lengths. Second, we propose near-infrared ultrafast excitation-emission and photoresponse measurements of these devices that aim to be the first demonstration of the key signatures of intrinsic regulation: (***I***) the control and enhancement of power conversion efficiency using fluctuating multi-channel input optical power. (***II***) the suppression of internal power fluctuations by two-channel absorption, and (***III***) the reduction of electronic and thermal energy dissipation through efficient charge carrier energy relaxation.

B. Research Progress

1. Continued Infrastructure development: Nanofabrication and bacterial growth facilities for the direct demonstration of intrinsically regulated light harvesting.

To explore the physics of intrinsic regulation and test the key signatures (***I***) – (***III***) outlined in **Section A**, it is necessary to produce ultra-high quality atomically thin devices. To advance this goal, **the PI has lead and continued the development of the NanoDevices Lab at UCR** to support the AFOSR projects proposed here (Figure 1). As the leader of this effort at UCR, I proposed that a shared-cost and shared-responsibility laboratory space should be maintained and supported by the institution to further support the efforts of faculty research efforts, particularly the efforts of those within the highly vulnerable early career phase. Five cooperating faculty, with resources and organizational support provided by the institution, would manage such a space. Future hires in related fields could utilize the space, as well as contribute through small contributions from initial lab funding.

Five experimental research groups (PIs: Nathan Gabor, Joshua Lui, Peng Wei, Yongtao Cui, Jing Shi) in the Department of Physics and Astronomy are building a shared nanodevice facility. The facility will consist of commonly used equipment for fabrication and characterization of nanodevices primarily based on two-dimensional materials and thin films. *This direction is a common interest for all five groups and also strength of many UCR groups across different departments.* This facility fills the gap of the general-purpose fabrication equipment available in the UCR cleanroom and building up state-of-art capabilities for 2D materials research. The facility will be primarily used and maintained by the five groups, and, once established, also open to other research groups.

The PI has established partial funds for several key components of the NanoDevice Lab and has been given a 900 sq.ft. space in the Materials Science and Engineering building to develop the

Figure 1: The NanoDevices Laboratory being developed by the PI as a shared technology facility at UCR.

shared laboratory space (Fig. 1). Gabor and Cui have already implemented safety protocols and begun to populate the lab space. While several of the newest pieces of equipment have been fully funded by combined research expenditures of all faculty, the future investments may require additional support. As part of this work, we have developed a strategic plan for priority equipment that is needed to fully encompass all aspects of NanoDevice fabrication.

A key advancement of Year 3 was the development of several nanofabrication techniques for the synthesis of biological analog photocells. The lab efforts have grown substantially and as of Year 3, several major projects across many disciplines have been advanced by the infrastructure of this facility. *This could be viewed as a major outcome of this award.* Based on the above advanced device demonstrations and characterization techniques, we are moving steadily toward our goals and maintaining a strong focus on the objectives. We are also exploring additional avenues and unexpected outcomes that support the proposed research.

In addition to device physics infrastructure, we have now begun work with close collaborators using an anaerobic chamber to culture rhodobacter spheroides, a purple bacteria that has been the subject of decades of photosynthesis research. This new infrastructure will follow the model developed for our device labs and will be the next generation of infrastructure development at UCR. Below, we discuss some of the recent outcomes of this development, and show how it will be used to continue exploring intrinsically regulated networks in biosystems.

2. Quieting a Noisy Antenna reproduces photosynthetic absorption spectra

The following section briefly describes the scientific content within a manuscript currently in revision at Nature, entitled, “Quieting a noisy antenna reproduces photosynthetic absorption spectra”.

Photosynthesis is remarkable, achieving near unity light harvesting quantum efficiency in spite of dynamic light conditions, rapidly fluctuating molecular structure, and highly intricate energy transfer pathways. The delicate interplay of quantum effects with molecular mechanisms of energy management have been explored across highly diverse phototrophs, giving unique insight for bio-inspired technologies. However, it remains unknown whether there exists a fundamental organizing principle that gives rise to robust photosynthetic light harvesting. In this recent work, we present a classical noise-canceling network model that relates noisy physiological conditions, power conversion efficiency, and the resulting absorption spectrum of photosynthetic organisms. Taking external light conditions in three distinct niches - full solar exposure, light filtered by oxygenic phototrophs, and under sea water - we derive optimal absorption characteristics for efficient solar power conversion. We show how light harvesting antennae can be finely tuned to maximize power conversion efficiency by passively minimizing excitation noise, thus providing a unified theoretical basis for the experimentally observed wavelength dependence of light absorption in green plants, purple bacteria, and green sulphur bacteria. Our noise-canceling antenna model, which establishes the elementary connection between highly efficient light energy harvesting and energetic fluctuations, provides an underlying design principle for future ultra-efficient bio-inspired network technologies.

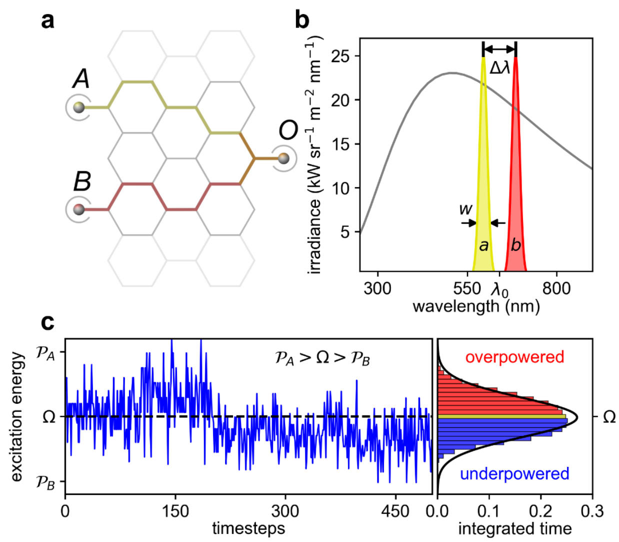

Figure 2. Light harvesting noisy antenna. Schematic of a photosynthetic antenna reduced into a network with two input nodes.

Transforming noisy inputs into quiet outputs represents a key design challenge in network architectures including multi-national energy grids,[1-5] auditory and visual neural networks,[6-9] and nanoscale photocells for next generation optoelectronics[10]. While network inputs exhibit statistical fluctuations (e.g., rapid changes of sunlight absorbed by a leaf or solar panel), network outputs may demand a steady rate of information or energy for optimal performance (e.g., constant power from the grid to maintain indoor lighting). Statistical fluctuations - arising from environmental variations and internal processes - fundamentally limit the throughput efficiency of any network. If the flow of energy (power) into a network is significantly larger or smaller than the flow out of the network required to optimally match the output demand, the network must adapt or be structured in such a way as to reduce the sudden over- or under-flow of energy. When the network fails to manage these sudden fluctuations, the results may be remarkable (e.g., photo-oxidative stress in photosynthetic light harvesting or explosive damage to transformers due to fluctuations in the grid).

In photosynthesis, light energy harvesting begins with the absorption of sunlight. Photoexcitation energy is rapidly transferred through an antenna network before reaching the reaction center, where charge transfer converts excitation energy into an electrochemical potential gradient across the photosynthetic membrane. Figure 2 illustrates our model, which employs generalizations of networks to extract the essential aspects of photosynthetic light harvesting. We begin by constructing a simple network of nodes connected by links, shown schematically in Fig. 2a. The nodes (points at which lines intercept) and links (connecting lines) represent physical objects: excitation energy levels and intermolecular transfer events within the antenna system, respectively. By analyzing the stochastic flow of excitation energy, we can characterize the antenna network by statistical averages (power throughput) and fluctuations in the rate of energy flow, which we will call noise. The power throughput of the antenna system is determined by external light conditions, the absorption characteristics of the absorbing pigment molecules (Figure 2b), or input nodes, and the molecular dynamics of the network. The antenna inputs are described in the usual way: light absorption by the pigment molecules is characterized by peak widths w, separation $\Delta\lambda = |\lambda_B - \lambda_A|$, and the center wavelength (or average distance) between the peaks λ_0. The solar spectral irradiance (grey line figure 2b) - which varies as light propagates through air, the canopy, or sea water - gives the *average* power available within a given range of wavelengths. Choosing the wavelength of an absorption peak simultaneously specifies both the excitation energy and power entering the noisy antenna. While the excitation energy is inversely proportional to wavelength, the absorbed power $\mathcal{P}_A$ or $\mathcal{P}_B$ entering the network is the integrated product of the spectral irradiance and the absorption characteristics of the light harvesting antenna.

Tuning only the absorption characteristics, our goal is to find a finely tuned network that spends the least amount of time in a state for which the input power is too large or too small compared to the output of the network, thus maximizing the power conversion efficiency (Figure 1c). Figure 3, our main result, shows three prototypical photosynthetic antennae - the light harvesting complex (LHC2) of green plants (Fig. 3a), the light harvesting complex (LH2) of purple bacteria (Fig. 3b), and the chlorosome of green sulphur bacteria (Fig. 3c) - and compares their absorption spectrum (Figs. 3d-f) to that predicted by our model (Figs. 3g-i). To obtain the results of Figure 3, our model takes as input the local irradiance spectrum, shown as grey lines in Figures 3d-i. Details of internal protein dynamics and the numerous potential electronic pathways through the network are embedded in rates p_A and p_B that couple the inputs of the network $\mathcal{P}_A$ and $\mathcal{P}_B$ to the output Ω: $p_A\mathcal{P}_A + p_B\mathcal{P}_B = \Omega$. Minimizing the variance (noise) of the average distribution $p_A\mathcal{P}_A + p_B\mathcal{P}_B$ then yields the optimal absorption characteristics for noise-cancellation.

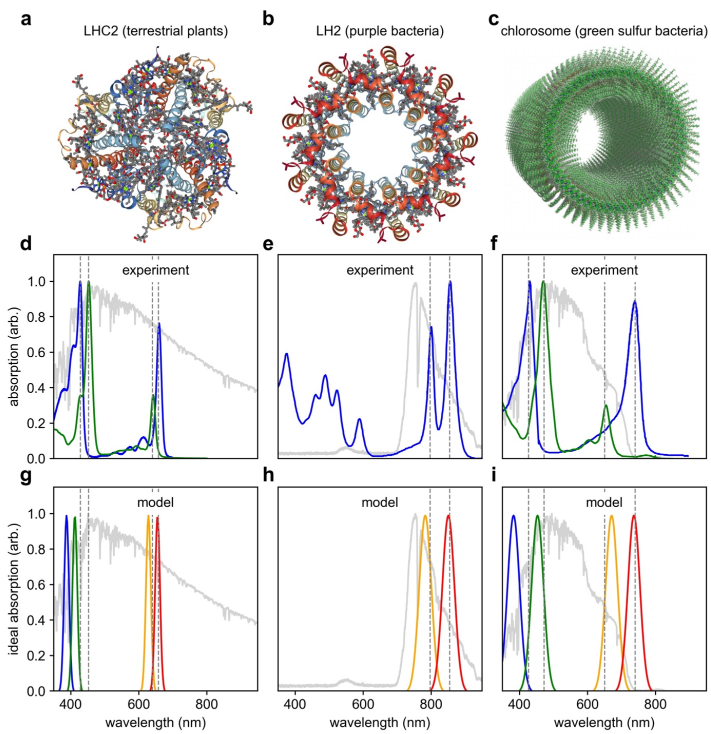

Figure 3. The noisy antenna model reproduces photosynthetic absorption spectra in three distinct niches. **Top,** Molecular structure of the light harvesting antenna LHC2 of green plants, the LH2 of purple bacteria, and the chlorosome of green sulphur bacteria, respectively.

It is remarkable that this simple model can describe a broad class of photosynthetic organisms, and we will continue to pursue new methodologies of measuring noise directly in real biological systems. Over the next year, the PI plans to spend several weeks at the University of Glasgow to work with collaborators there and bring new techniques into the lab.

3. Moving toward highly parallelized data-intensive measurements of photosynthetic doubling in rhodobacter sphaeroides

This section very briefly describes our most recent experimental results in culturing photosynthetic bacteria for future work on directed evolution and massively parallelized analysis of photosynthetic efficiencies.

Photosynthesis, an organic process developed in the nascent stages of life on earth, converts electromagnetic radiation into chemical energy. Photosynthesis is generally understood to be a process endemic to plants; however, this process is found also to occur in numerous species of bacteria. Our research therefore investigates the photosynthetic nature of a small but important class of photosynthetic bacteria, *Rhodobacter Sphaeroides*. The bacteria we are using for this research are non-sulfur purple bacteria. These metabolically versatile bacteria and their photosynthetic systems are well understood; thus, they serve as an excellent biosystem to explore the physics of photosynthesis. The strain used in our recent work is the non-mutated Wild type *2.4.1*. Establishing growth of this species is relatively easy and the bacteria is well understood, both at a macroscopic and microscopic level.

Stage 1. Growth under aerobic conditions. We began by using a typical medium for aerobic growth: 3g beef extract and 5g peptone diluted to 1L of ultrapure water. This medium is commonly used to grow *R.sphaeroides* aerobically. The bacteria were then inoculated into 3ml of this medium in a test tube. The chosen volume of the media is arbitrary; however, there needs to be enough medium volume to make proper measurements. The test tube is then placed into a 30℃ incubator with limited access to light. These conditions are recommended for optimal aerobic growth of the R.*sphaeroides*, and we have successfully grown the bacterial culture under these conditions.

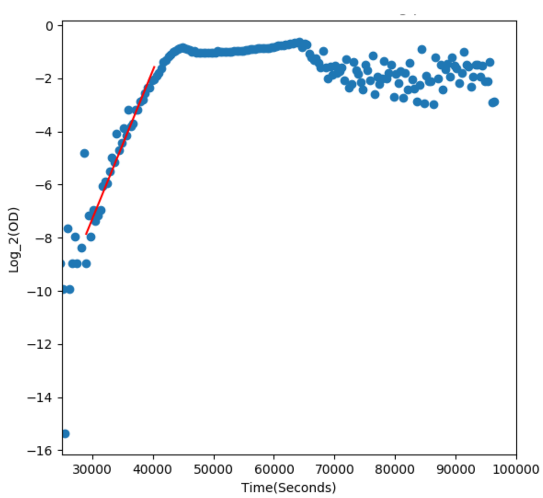

Figure 4. Absorption vs. time for Rhodobacter Spheroides measured at 600nm. From the exponential increase in optical density, the doubling time is found to be 30 minutes.

Stage 2. Growth under anaerobic conditions. For anaerobic growth, we again use standard procedures. The medium used is Sistrom's Minimal Medium (SIS), a commonly referenced growth medium for *R.sphaeroides*. To allow the bacteria to grow photosynthetically, the bacteria need an anaerobic environment. To provide this environment we used a BACTRON 900 anaerobic chamber. The chamber contains two incubators which have temperature control. The anaerobic mixed gas (AMG) used was: 5% H_2, 5% CO_2 and 90% N_2. The hydrogen, combined with O_2 in the environment and palladium pellets in the O_2 scrubber causes a catalytic reaction which produces water condensation as an output. The CO_2 is necessary for fixation in the light dependent Calvin cycle of the bacteria. Since the *R.sphaeroides* are metabolically diverse, we can also do fixation with H_2. To further ensure our anaerobic environment, we degas and spurge the medium in which the bacteria will be inoculated. We use a desiccator, magnetic stirrer, pump, and fish tank sparger to ensure anaerobic conditions.

Using a spectrophotometer, A 6-well plate was used for measurement of the optical density while growing the bacterial culture. 2 ml of the aerobic media was placed in five of the wells. One well was left empty and designated as a control. *R.sphaeroides* were inoculated directly from the freezer culture into each of the five wells. Figure 4 shows the results: a semi log plot of the optical density vs time at a selected wavelength using the plate reader shows a clear growth of the culture. The plate reader recorded a total of 250 cycles over a time span of 27 hours. A line of regression was placed to show the period of growth of the bacteria. The inverse slope corresponds to the doubling time of the bacteria. For 600nm that doubling time was found to be 30 minutes. All of the results are consistent with the literature values.

Based on these recent results, we are now poised to begin a new class of biophysical measurements to understand photosynthesis from the perspective of noise. Our future work will focus heavily in this direction and will combine the tools of bacterial genetics with those of nanoscale optics and optoelectronics to explore this exciting frontier.

C. Summary of Publications and Merits

1. Publications resulting from this award (to date)

In Year 3, **three** major publications under AFOSR support were published, and **one** manuscript is under review in a high impact journal:

8. “Quieting a Noisy antenna reproduces photosynthetic absorption spectra,” Jed Kistner-Morris, Trevor B. Arp, Vivek Aji, Richard Cogdell, Rienk van Grondelle, Nathaniel M. Gabor*; *in revision* ***Nature*** (2019).

7. “Multiple parameter dynamic photoresponse microscopy for data-intensive optoelectronic measurements of van der Waals heterostructures,” Trevor B. Arp, Nathaniel M. Gabor*; ***Review of Scientific Instruments*** *90,* 023702 (2019). ArXiv:1812.03232v1.

6. “Electron-hole liquid in a van der Waals heterostructure photocell at room temperature,” Trevor B. Arp, Dennis Pleskot, Vivek Aji, Nathaniel M. Gabor*; ***Nature Photonics*** (2019). ArXiv:1711.06917v1.

5. “Giant intrinsic photoresponse in pristine graphene,” Qiong Ma, Chun Hung Lui, Justin C.W. Song, Jian Feng Kong, Yuan Cao, Nityan L. Nair, Wenjing Fang, Kenji Watanabe, Takashi Taniguchi, Su-Yang Xu, Jing Kong, Nuh Gedik, Nathaniel M. Gabor*, Pablo Jarillo-Herrero; ***Nature Nanotechnology*** (2018).

In Year 2, **two** manuscripts under AFOSR support were published:

4. "Electron quantum metamaterials in van der Waals heterostructures," Justin C. W. Song, Nathaniel M. Gabor, ***Nature Nanotechnology*** 13, 986 (2018).

3. "Hot Carrier-Enhanced interlayer electron-hole pair multiplication in 2D semiconductor heterostructure photocells," Fatemeh Barati, Max Grossnickle, Shanshan Su, Roger K. Lake, Vivek Aji, Nathaniel M. Gabor; ***Nature Nanotechnology*** 12, 1134 (2017).

In Year 1, **two** manuscripts under AFOSR support were published:

2. "Natural regulation of energy flow in a green quantum photocell," Trevor B. Arp, Yafis Barlas, Vivek Aji, Nathaniel M. Gabor; ***Nano Letters***, DOI: 10.1021/acs.nanolett.6b03162 (2016).

1. "Nd:AlN polycrystalline ceramics: A candidate media for tunable, high energy, near IR lasers," Andrew Wieg, Max Grossnickle, Yasuhiro Kodera, Nathaniel M. Gabor, Javier Garay; ***Applied Physics Letters*** **109**, 121901 (2016).

2. Merits, promotions, awards, and invited seminars (Year 3)

The PI was reviewed and awarded a merit promotion to tenured professor in 2018 by the College of Natural and Agricultural Sciences at the University of California Riverside. The PI now holds the rank of **JET Distinguished Associate Professor with tenure**. In addition to receiving the NSF Career Award, the AFOSR YIP Award, and the Cottrell scholar award in 2017, the PI was awarded with several important young faculty awards during the past 2 years. These are listed below:

Presidential Early Career Award for Scientists and Engineers 2019 – 'The PECASE award is the highest honor bestowed by the United States government to outstanding scientists and engineers.'

Department of the Navy HBCU/MI (Historically Black Colleges and Universities and Minority Serving Institutions) Award 2019 – 'This highly competitive award supports research efforts that will contribute to the science and technology mission and vision of the U.S. Navy and U.S. Marine Corps while increasing the engagement of students, including underrepresented minorities, in STEM fields.

National Academy of Sciences Kavli Frontiers Fellow 2019 – 'The Kavli Frontiers of Science symposium is the Academy's premier activity for distinguished young scientists.'

Research Corporation for Science Advancement Scialog Fellow 2018 – 'Bringing together promising early career investigators and distinguished scientific leaders for the search and discovery of truly transformative ideas.'

CIFAR Global Scholar Award 2018 Bio-Inspired Solar energy – 'The Canadian Institute for Advanced Research (CIFAR) invites outstanding early career investigators from across the globe into research programs addressing some of the most complex challenges facing the world today.'

The PI has given numerous invited talks in 2017-2019 related to the work proposed and discussed here:

1. "From heat engines to green leaves: a physicist's perspective on photosynthesis," *University of California Davis Chemistry Colloquium*, Davis, CA (2019). *Penn State Physics Colloquium*, State College, PA (2018). *LASSP Seminar Cornell University*, Ithaca, NY (2018)*CEPCEB Noel T. Keen Symposium Plant Biology and Physiology*, Riverside, CA (2018). *CIFAR Bio-Inspired Solar Energy,* Toronto, Ontario, Canada (2018).

2. "Nanoscience in the Age of Big Data,"
RCSA Cottrell Conference, Tucson, AZ (2017).

3. "Room temperature 2D condensate of electrons and holes in ultrathin MoTe2 photocells," *National Academy of Sciences Kavli Frontiers,* Irvine, CA (2019). *Stanford AMO Seminar*, Palo Alto, CA (2019), *Caltech Applied Physics and Materials Science Seminar*, Pasadena, CA (2018). *N2D Spain, Workshop on Nanophotonics in 2D Materials*, San Sebastian, Spain (2017). *QCM Workshop Enduring Problems in Quantum Condensed Matter, A Symposium Honoring Chandra Varma,* Riverside, CA (2017)

The graduate student researcher within the PIs lab and working on this project, Fatemeh Barati, successfully defended her graduate thesis and was also awarded with several important awards and fellowships. These are listed below:

New York University Postdoctoral Fellowship in the Center for Quantum Phenomena 2019 (Fatemeh Barati), 'Dr. Barati will take her expertise in nanoscale device fabrication and advanced optical measurements to the laboratory of Prof. Shabani, working to develop new qubits for applications in quantum computation.'

American Physical Society Ken Hass Award 2018 (Fatemeh Barati), **'**The purpose of this award is to recognize the best student paper addressing the subject of industrial applications of physics. The Ken Hass Outstanding Student Paper Award is named in recognition of the many contributions of Ken Hass to the industrial applications of physics (especially automotive applications of theoretical solid-state physics) and of his service to the APS in the FIAP Chair-line from 2001 to 2004. This is the highest student award granted at the annual APS March Meeting.'

Dr. Janet M. Boyce Memorial Award 2017 (Fatemeh Barati), 'Outstanding women in the science based upon strong academic research and impressive letters of recommendation, recognized by Dr. Isgouhi Kaloshian, chair of the CNAS committee on honors &

scholarships, and Dr. Michael McKibben, CNAS divisional dean of student academic affairs'

Benjamin C. Shen Award 2016 (Fatemeh Barati) – Outstanding 3rd year grad student. 'For exceptional contributions to the study of electron transport in low-dimensional materials and their heterostructures.'

Outstanding Teaching (OT) Award 2016 (Fatemeh Barati) - Acknowledging extraordinary efforts toward fulfilling the goals and missions of education at the University of California. This is among the highest honors of Teaching Assistants.

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