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Integrated 2D Nonlinear Nanophotonics

Arka Majumdar
UNIVERSITY OF WASHINGTON

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Dr. Arka Majumdar

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14. ABSTRACT
Under this proposal, we explored several novel nonlinear optical effect in 2D materials, including the nonlinar effect originating from the exciton and trions as well as the auger recombination in this material system. Additionally, we developed integrated photonic technologies to have a robust transfer process. Finally, we demonstrate fabricating a whole device by patterning 2D materials.

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Arka Majumdar

University of Washington, Seattle

Email: arka@uw.edu

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1. Encapsulated silicon nitride nanobeam cavity for nanophotonics using layered materials, Taylor K. Fryett, Yueyang Chen, James Whitehead, Zane Matthew Peycke, Xiaodong Xu, Arka Majumdar, ACS Photonics, 5 (6), pp 2176–2181, 2018.
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3. Deterministic positioning of colloidal quantum dots on silicon nitride nanobeam cavities, Yueyang Chen, Albert Ryou, Max Friedfeld, Taylor Fryett, James Whitehead, Brandi Cossairt, Arka Majumdar, Nano Letters, 18 (10), pp 6404–6410, 2018.
4. Ultrathin van der Waals metalenses, Chang-Hua Liu, Jiajiu Zheng, Shane Colburn, Taylor K. Fryett, Yueyang Chen, Xiaodong Xu, Arka Majumdar, Nano Letters, 18 (11), pp 6961–6966, (2018).
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Submitted/ In Preparation:

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7. Auger Photocurrent Spectroscopy of Excitons in Monolayer Semiconductors, Colin M. Chow, Hongyi Yu, John R. Schaibley, Pasqual Rivera, Joseph Finney, Jiaqiang Yan, David G. Mandrus, Takashi Taniguchi, Kenji Watanabe, Wang Yao, David H. Cobden, Xiaodong Xu, in preparation for Nature Nanotechnology.
8. Nonlinear Optical Spectroscopy of Interacting Valley Exciton and Trions in 2D Semiconductors, John R. Schaibley, Hongyi Yu, Todd Karin, Pasqual Rivera, Marie Scott, Jiaqiang Yan, David G. Mandrus, Takashi Taniguchi, Kenji Watanabe, Wang Yao, Kai-Mei Fu and Xiaodong Xu, in preparation for Nature Physics.
9. Chiral plasmons in quantum anomalous Hall insulators, Furu Zhang, Jianhui Zhou, Di Xiao, and Yugui Yao, in preparation for Phys. Rev. Letters.

Encapsulated SiN cavity development and demonstration of 2D material coupling:

Most existing implementations of silicon nitride photonic crystal cavities rely on suspended membranes due to its low refractive index. Such floating membranes are not mechanically robust, making them suboptimal for developing a hybrid optoelectronic platform where new materials, such as layered 2D materials, are transferred onto pre-fabricated optical cavities. To address this issue, we design and fabricate a silicon nitride nanobeam resonator where the silicon nitride membrane is encapsulated by material with a refractive index of ~1.5, such as silicon dioxide or PMMA [1]. The theoretically calculated quality factor of the cavities can be as large as 10^5, with a mode-volume of $\sim 2.5\left(\frac{\lambda}{n}\right)^3$. We fabricated the cavity and measured the transmission spectrum with the highest quality factor reaching 7,000. We also successfully transferred monolayer tungsten diselenide on the encapsulated silicon nitride nanobeam and demonstrated coupling of the cavity with both the monolayer exciton and the defect emissions.

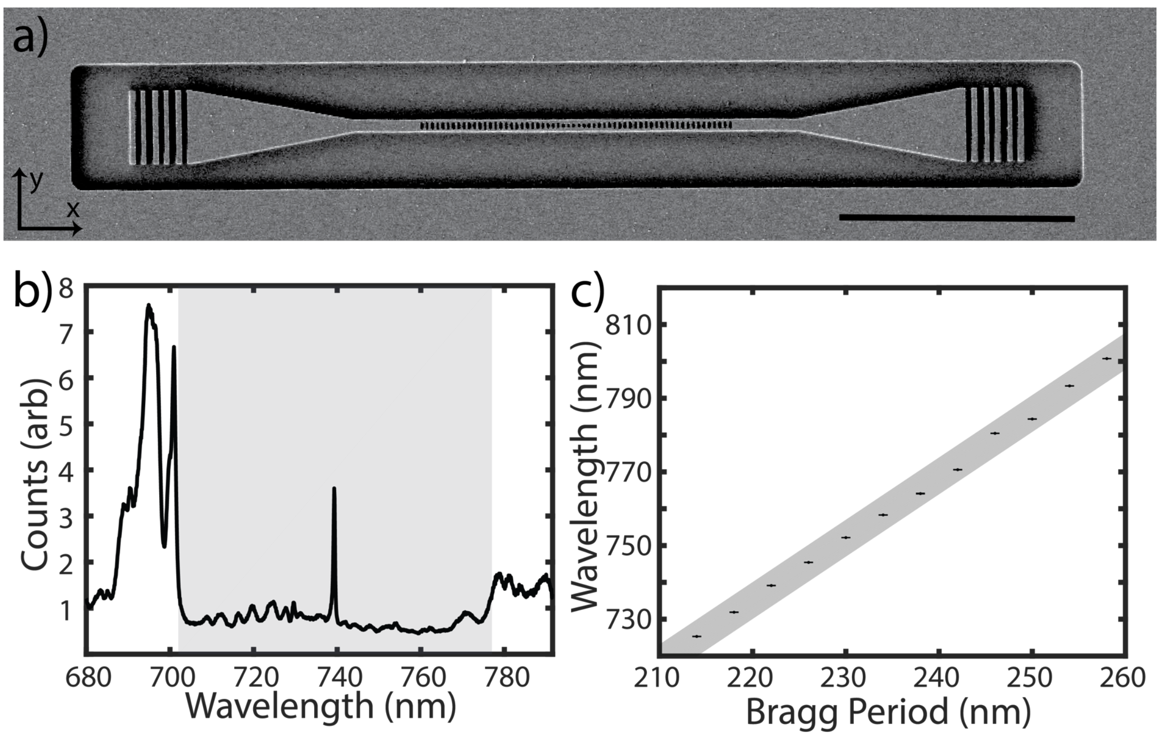

Figure 1: Bare cavity resonances: *(a) An SEM of a fabricated SiN nanobeam prior to encapsulation. The nanobeam resonators are probed via the two grating couplers on the ends of the nanobeams. The scale-bar is 10 μm. (b) Example cavity transmission spectrum as measured through the gratings. The shaded portion highlights the low transmission region from the Bragg reflectors, with the cavity peak at the center. (c) The observed cavity resonances scale linearly with the Bragg period, while holding the ratio between radii and periodicity constant.*

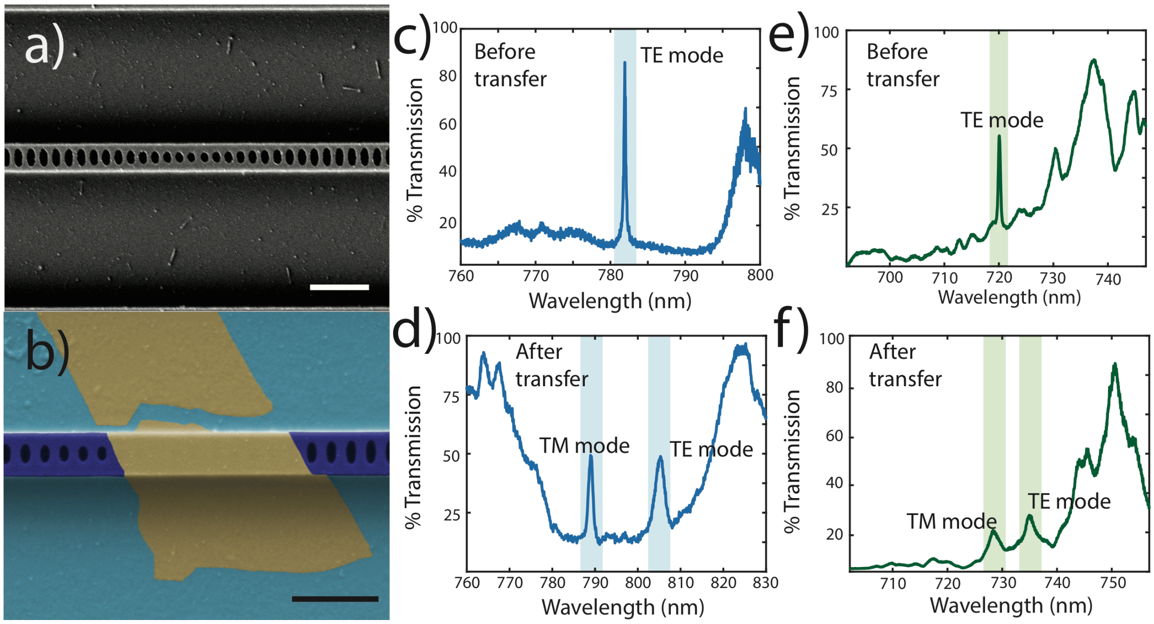

Figure 2: Transmission through SiN nanobeam before and after transfer of WSe_2: *(a) A SEM of the defect region of the nanobeam. (b) False colored SEM of a nanobeam with monolayer WSe_2. The SiN is shown in dark blue, the silicon oxide is shown in light blue, and the WSe_2 is shown in gold. The scale-bar in both figures corresponds to $1\mu m$. (c), (e) The transmission spectrum before transferring WSe_2 for devices 1 and 2, respectively. (d), (f) The transmission spectra after WSe_2 transfer for devices 1 and 2, respectively.*

Strong photon antibunching in weakly nonlinear two-dimensional exciton-polaritons

A deterministic and scalable array of single photon nonlinearities in the solid state holds great potential for both fundamental physics and technological applications, but its realization has proved extremely challenging. Despite significant advances, leading candidates such as quantum dots and group III-V quantum wells have yet to overcome their respective bottlenecks in random positioning and weak nonlinearity. Here we consider a hybrid light-matter platform, marrying an atomically thin two-dimensional material to a photonic crystal cavity, and analyze its second-order coherence function. We identify several mechanisms for photon antibunching under different system parameters, including one characterized by large dissipation and weak nonlinearity. Finally, we show that by patterning the two-dimensional material into different sizes, we can drive our system dynamics from a coherent state into a regime of strong antibunching with second-order coherence function $g^{(2)}(0) \sim 10^{-3}$, opening a possible route to scalable, on-chip quantum simulations with correlated photons.

Deterministic positioning of colloidal quantum dots on silicon nitride nanobeam cavities

Engineering an array of precisely located cavity-coupled active media poses a major experimental challenge in the field of hybrid integrated photonics. We deterministically position solution-processed colloidal quantum dots (QDs) on high quality-factor silicon nitride nanobeam cavities and demonstrate light-matter coupling. By lithographically defining a window on top of an encapsulated cavity that is cladded in a polymer resist, and spin coating the QD solution, we can precisely control the placement of the QDs, which subsequently couple to the cavity. We show rudimentary control of the number of QDs coupled to the cavity by modifying the size of the window. Furthermore, we demonstrate Purcell enhancement and saturable photoluminescence in this QD-cavity platform. Finally, we deterministically position QDs on a photonic molecule and observe QD-coupled cavity super-modes. Our results pave the way for precisely controlling the number of QDs coupled to a cavity by engineering the window size, the QD dimension, and the solution chemistry and will allow advanced studies in cavity enhanced single photon emission, ultralow power nonlinear optics, and quantum many-body simulations with interacting photons. Note that, while the proposed research was meant to work with 2D material, this work is enabled by the cavities designed under the proposal.

Ultrathin van der Waals metalenses

Ultrathin and flat optical lenses are essential for modern optical imaging, spectroscopy, and energy harvesting. Dielectric metasurfaces comprising nanoscale quasi-periodic resonator arrays are promising for such applications, as they can tailor the phase, amplitude, and polarization of light at subwavelength resolution, enabling multi-functional optical elements. To achieve 2π phase coverage, however, most dielectric metalenses need a thickness comparable to the wavelength, requiring fabrication of high-aspect-ratio scattering elements. Here, we report ultrathin dielectric metalenses made of van der Waals (vdW) materials, leveraging their high refractive indices and the incomplete phase design approach to achieve device thicknesses down to $\sim\boldsymbol{\lambda}/10$, operating at infrared and visible wavelengths. These materials have generated strong interest in recent years due to their advantageous optoelectronic properties. Using vdW metalenses, we demonstrate near diffraction-limited focusing and imaging, and exploit their layered nature to transfer the fabricated metalenses onto flexible substrates to show strain-induced tunable focusing. Our work enables further downscaling of optical elements and opportunities for integration of metasurface optics in ultra-miniature optoelectronic systems.

Interlayer Exciton Emission Enhanced by Photonic Crystal Cavity

The advent of van der Waals heterostructures marks the emergence of a new class of synthetic materials with novel and unique properties, unattainable in their constituent materials. The two-dimensional architecture of these heterostructures makes them naturally suited for integration with a wide variety of planar nanophotonic structures, including optical cavities for next-generation low-power optoelectronic devices and explorations of fundamental physical effects in these new systems. Here, we report the coupling of the interlayer exciton in a transition metal dichalcogenide heterobilayer with a gallium phosphide photonic crystal defect cavity. The exciton-cavity coupling is found to be in the weak regime, resulting in ~10-fold increase in the photoluminescence intensity for interlayer exciton in resonance with the cavity. The order of magnitude enhancement of the photoluminescence yield offsets the low oscillator strength of the interlayer exciton, adding a new tool for probing the underlying physics of this novel excitonic system.

Chiral plasmons in quantum anomalous Hall insulators

We investigate the edge plasmons in magnetically doped topological insulator thin films and find the Berry curvature splits the edge plasmons propagating along the opposite direction of the edge. When the bulk is insulating, only one unidirectional edge plasmon mode survives and propagates along the edge, whose direction can be changed by external fields. In the long-wavelength limit, the unidirectional edge plasmon is acoustic and essentially determined by the anomalous Hall conductivity. For arge wave vector, the group velocity of the chiral edge plasmon would change sign, which originates from the k-quadratic correction to the effective mass. The impacts of the Fermi level and the wave vector on the bulk and edge plasmons are discussed. Our work provides a quantitative understanding of the recent observation of the chiral edge plasmon in quantum anomalous Hall insulators and some insight into the application of realistic topological materials in chiral plasmonics.

Auger Photocurrent Spectroscopy of Excitons in Monolayer Semiconductors

In Auger spectroscopy, the internal transitions of atoms relaxing after X-ray absorption cause the ejection of electrons from higher energy levels into vacuum, where they are collected, and their energy distribution measured. A similar process, called Auger scattering, occurs in photoexcited semiconductors[1] when the energy released by recombination of an electron-hole pair is transferred to a third carrier. In the solid, however, this "missing" energy is normally dissipated non-radiatively and can only be inferred from energy balance. Here, we introduce a new technique, Auger photocurrent spectroscopy, which allows the measurement of the energy of the Auger-excited carriers by tunneling. The semiconductor we use is monolayer WSe_2, and a thin hexagonal boron nitride (BN) dielectric barrier plays a role analogous to the vacuum in Auger spectroscopy: its wide bandgap allows it to function as an energy filter, transmitting the high-energy Auger carriers but not the low-energy quasi-equilibrium carriers. The signal is the current flowing through the BN from the WSe_2 to a graphite electrode under illumination. By mapping this photocurrent as a function of gate voltage and photon energy, we observe the Auger scattering of holes from multiple WSe_2 excitonic species and investigate the interplay between band filling and the renormalization of exciton energies with doping. An interesting consequence of the renormalization is negative differential photoconductance. Our findings illustrate how van der Waals heterostructures enable new approaches to studying carrier dynamics and interactions in solids. The paper is current in preparation for Nature Nanotechnology.

Nonlinear Optical Spectroscopy of Interacting Valley Exciton and Trions in 2D Semiconductors

Monolayer transition metal dichalcogenide (TMD) are the subject of intense investigation due to their exceptional electronic, optical and mechanical properties. The optical response of these two-dimensional (2D) semiconductors is dominated by strongly bound excitons with valley-degrees of freedom. Here, we report on the resonant nonlinear optical response of monolayer $MoSe_2$ encapsulated in hexagonal boron nitride (hBN) at low temperature (30 K). We identify nonlinear interactions that govern both the exciton and negatively charged trion. In our high-quality samples with exciton spectral linewidth of 5.6 meV, we obtain a linear transmission change of 35% and a resonant $\chi^{(3)} = 7.4\, x\, 10^{-5}$ esu ($\sim 10^{-12} m^2/V^2$). We also observed valley dependent correlations between the exciton and trion, measured by two-color continuous wave nonlinear spectroscopy. The valley dependent exciton-trion correlations can be explained in terms of trion formation and bandgap renormalization. Our work sheds light on previous work, which observed exciton-trion interactions but was unable to determine their physical origin. This understanding of the resonant $\chi^{(3)}$ is important for future scientific and technological applications of monolayer TMD semiconductors, where the resonant nonlinear response can be used for atomically thin modulators, frequency shifters and amplifiers that can be directly integrated on-chip with photonic waveguides. The paper is current in preparation for Nature Physics.

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[1] T. K. Fryett, Y. Chen, J. Whitehead, Z. M. Peycke, X. Xu, and A. Majumdar, "Encapsulated Silicon Nitride Nanobeam Cavity for Hybrid Nanophotonics," *ACS Photonics,* 2018/04/18 2018.