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Optical Control of Electronic Topological States

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TECHNICAL REPORT

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Optical Control of Electronic Topological States

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1. Accomplishments:

A new perspective on light-matter interaction for correlated electron systems:

An important goal of modern condensed-matter physics involves the search for states of matter with emergent properties and desirable functionalities. Although the tools for material design remain relatively limited, notable advances have been recently achieved by controlling interactions at heterointerfaces, precise alignment of low-dimensional materials, and the use of extreme pressures. In a Nature Perspective, we featured a paradigm based on controlling light–matter interactions, which provides a way to manipulate and synthesize strongly correlated quantum matter. We considered the case in which both electron–electron and electron–photon interactions are strong and give rise to a variety of phenomena.

Photon-mediated superconductivity, cavity fractional quantum Hall physics, and optically driven topological phenomena in low dimensions were among the frontiers discussed in this Perspective, which focused upon a field that we termed ‘strongly correlated electron–photon science.’ [Strongly correlated electron-photon systems, J. Bloch, A. Cavalleri, V. Galitski, M. Hafezi, and A. Rubio, Nature, 606, 41-48 (2022)]

Two-dimensional layered material and correlated state: Mott-moiré excitons:

There have been many exciting new results in optical measurement of correlated states in layered material. A hallmark example is the Mott states in moiré bilayers system. However, a systematic theory did not exist. We addressed this problem in our recent work [Mott-moiré excitons, T.-S. Huang, Y-Z Chou, C. L. Baldwin, F. Wu, M. Hafezi, arXiv:2207.13152 (2022)]. Specifically, we considered excitons in moiré systems for which the valence band is in the Mott-insulating regime. These “Mott-moiré excitons,” which are achievable in twisted TMD heterobilayers, are bound states of a magnetic polaron in the valence band and a free electron in the conduction band. We found significantly narrower exciton bandwidths in the presence of Hubbard physics, serving as a potential experimental signature of strong correlations. We also demonstrated the high tunability of Mott-moiré excitons through the dependence of their binding energies, diameters, and bandwidths on the moiré period. Our work provides guidelines

for future exploration of strongly correlated excitons in twisted TMD heterobilayers. Refer to the technical section, for our experimental progress on Mott-moire excitons.

Meanwhile, before entering to the many-body regime, we performed magneto-optical measurements of a negatively charged 2s exciton in WSe₂. Monolayer transition metal dichalcogenides host a variety of optically excited quasiparticles species that stem from two-dimensional confinement combined with relatively large carrier effective masses and reduced dielectric screening. The magnetic response of these quasiparticles gives information on their spin and valley configurations, nuanced carrier interactions, and insight into the underlying band structure. Recently, there have been several reports of 2s/3s charged excitons in TMDs, but very little is still known about their response to external magnetic fields. Using photoluminescence excitation spectroscopy, we observed the presence of the 2s charged exciton and reported for the first time its response to an applied magnetic field. We benchmarked this response against the neutral exciton and found that both the 2s neutral and charged excitons exhibit similar behavior with g-factors of $g_{2s0} = -5.20 \pm 0.11$ μB and $g_{2s-} = -4.98 \pm 0.11$ μB , respectively. [Magneto-Optical Measurements of the Negatively Charged 2s Exciton in WSe₂, J.C. Sell, J.R. Vannucci, D.G. Suarez-Forero, Cao, D.W. Session, H.-J. Chuang, K.M. McCreary, M.R. Rosenberger, B.T. Jonker, S. Mittal, and M. Hafezi, Phys.Rev.B(2022)] This work was in collaboration with NRL.

Optical properties and manipulation of electronic Quantum Hall states:

We theoretically proposed a scheme to create an electronic Floquet vortex state by irradiating a two-dimensional semiconductor with a laser light carrying nonzero orbital angular momentum. We analytically and numerically studied the properties of the Floquet vortex states, with methods analogous to the ones previously applied to the analysis of superconducting vortex states. We showed that such Floquet vortex states are similar to superconducting vortex states, and they exhibit a wide range of tunability. To illustrate the potential utility of such tunability, we showed how such states could be used for quantum state engineering. [Floquet vortex states induced by light carrying an orbital angular momentum, H. Kim, H. Deghani, I. Ahmadabadi, I. Martin, M. Hafezi, Phys. Rev. B 105, L081301 (2022)]. Further work on Floquet vortex states was consequently done in the area of optical conductivity and orbital magnetization. Motivated by earlier experimental demonstrations of Floquet topological insulators, there had been several theoretical proposals for using structured light, either spatial or spectral, to create other properties such as flat band and vortex states. Building upon our earlier work, we evaluated the orbital magnetization and optical conductivity as physical observables for such a system. We showed that the OAM of light induces nonzero orbital magnetization and current density. The orbital magnetization density increased linearly as a function of OAM degree, and non-linearly as a function of the frequency of the light field. In

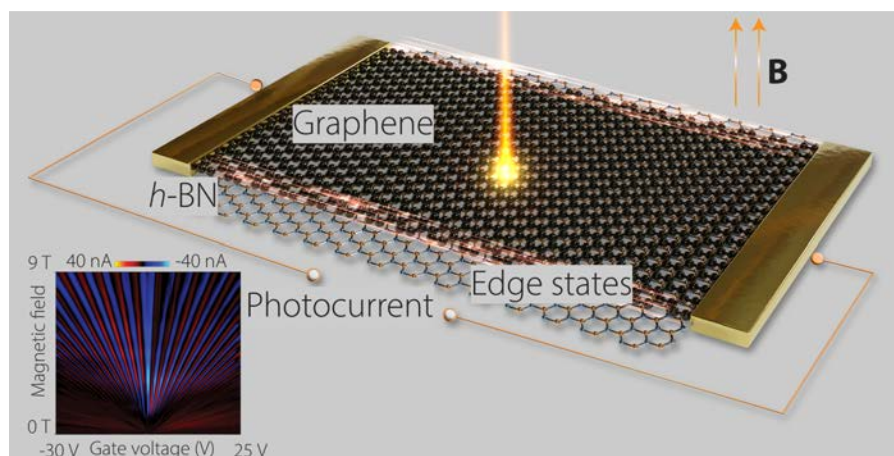
certain regimes, we found that orbital magnetization density is independent of the system size, width, and Rabi frequency of light. Furthermore, we studied the optical conductivity for various types of electron transitions between different states such as vortex, edge, and bulk that were present in the system. Based on conductance frequency peaks, a scheme for the detection of vortex states was proposed. [Optical conductivity and orbital magnetization of Floquet vortex states, I. Ahmadabadi, H. Dehghani, and M. Hafezi, arXiv:2204.09488 (2022)]

We've also completed a theoretical work with two-dimensional excitons from twisted light and refined our understanding of the fate of the photon's orbital angular momentum, in terms of a no-go theorem. As the bound state of two oppositely charged particles, excitons emerge from optically excited semiconductors as the electronic analogue of a hydrogen atom. In the two-dimensional (2D) case, realized either in quantum well systems or truly 2D materials such as transition metal dichalcogenides, the relative motion of an exciton is described by two quantum numbers: the principal quantum number n , and a quantum number j for the angular momentum along the perpendicular axis. Conservation of angular momentum demands that only the $j = 0$ states of the excitons are optically active in a system illuminated by plane waves. We considered the case for spatially structured light sources, specifically for twisted light beams with non-zero orbital angular momentum per photon. Under the so-called dipole approximation where the spatial variations of the light source occur on length scales much larger than the size of the semiconductor's unit cell, we showed that the photon (linear and/or angular) momentum is coupled to the center-of-mass (linear and/or angular) momentum of the exciton. Our study established that the selection rule for the internal states of the exciton, and thus the exciton spectrum, is independent from the spatial structure of the light source. [Two-dimensional excitons from twisted light and the fate of the photon's orbital angular momentum, T. Graß, U Bhattarcharya, J. Sell, M. Hafezi, Physical Review B 105, 205202 (2022)]

Optical flux pump in the quantum Hall regime:

A seminal *gedankenexperiment* by Laughlin described the charge transport in quantum Hall systems via the pumping of flux. We theoretically proposed an optical scheme that probes and manipulates quantum Hall systems in a similar way: when light containing orbital angular momentum interacts with electronic Landau levels, it acts as a flux pump which radially moves the electrons through the sample. We investigated this effect for a graphene system with annulus geometry and calculated the radial current in the absence of any electric potential bias. Remarkably, the current is robust against disorder, and in the weak excitation limit, the current showed a power-law scaling with intensity characterized by the novel exponent $2/3$. [Optical flux pump in the quantum Hall regime, B. Cao, T. Graß, G. Solomon, and M. Hafezi, PhysRevB.103.L241301, (2021)] Refer to the technical section, for our experimental progress on optical flux pump.

Meanwhile, our experimental work with chiral transport of hot carriers in graphene in the quantum Hall regime delivered interesting results: Photocurrent (PC) measurements can reveal the relaxation dynamics of photo-excited hot carriers beyond the linear response of conventional transport experiments, a regime important for carrier multiplication. In graphene subject to a magnetic field, PC measurements can probe the existence of Landau levels with different edge chiralities exclusive to relativistic electron systems. Here, we reported the accurate measurement of PC in graphene in the quantum Hall regime. Prominent PC oscillations as a function of gate voltage on samples' edges were observed. These oscillation amplitudes formed an envelope which depended on the strength of the magnetic field, as did the PCs' power dependence and their saturation behavior. We explained these experimental observations through a model using optical Bloch equations, incorporating relaxations through acoustic-, opticalphonons and Coulomb interactions. The simulated PC agreed with our experimental results, leading to a unified understanding of the chiral PC in graphene at various magnetic field strengths, and providing hints for the occurrence of a sizable carrier multiplication. [Chiral transport of hot carriers in graphene in the quantum Hall regime, B. Cao, T.Graß, O. Gazzano, K. Ashokbhai Patel, J. Hu, M. Muller, T. Huber, L. Anzi, K. Watanabe, T. Taniguchi, D. Newell, M. Gullans, R. Sordan, M. Hafezi, and G Solomon, arXiv:2110.01079v1 (2021)] Accepted for publication in ACS Nano.



Magnetic field and Fermi level dependence of the photocurrent on the edge, Exhibiting the Landau fan. The excitation power used is $1 \mu\text{W}$.

2. Impacts

In a Nature Perspective, we featured a paradigm based on controlling light–matter interactions, which provides a way to manipulate and synthesize strongly correlated quantum matter. We considered the case in which both electron–electron and electron–photon interactions are strong and give rise to a variety of novel phenomena. Photon-mediated superconductivity, cavity fractional quantum Hall physics, and optically driven topological phenomena in low dimensions were among the frontiers discussed in this Perspective, which focused upon a field that we termed ‘strongly correlated electron–photon science.’ [Strongly correlated electron-photon systems, J. Bloch, A. Cavalleri, V. Galitski, M. Hafezi, and A. Rubio, Nature, 606, 41-48 (2022)].

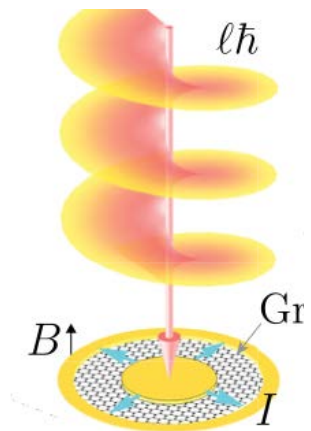
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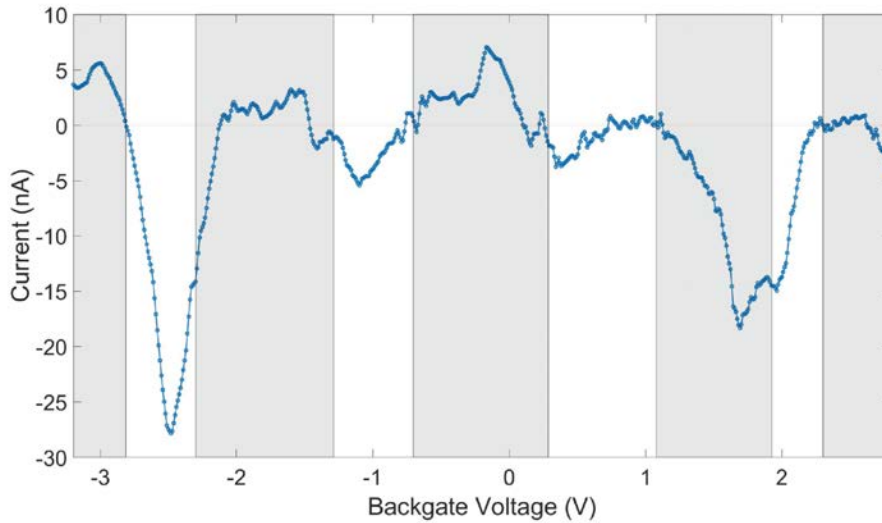
We continue to experience some delays in our work due to covid. Infrastructural concerns such as loss of staff at the University exacerbate supply chain issues to make wait times for the purchase or repair of equipment unusually lengthy. Several post-doctoral or graduate student researchers have had travel and visa delays that impeded significantly their ability to join us and therefore our progress.

4. Technical Updates

Manipulation of quantum Hall state with vortex beams:

Following our theoretical proposal [Optical flux pump in the quantum Hall regime, B. Cao, T. Graß, G. Solomon, and M. Hafezi, PhysRevB.103.L241301, (2021)], we have been conducting photocurrent experiments by illuminating an annulus geometry graphene sample with a laser field carrying optical angular momentum. Our preliminary results shows that indeed by switching the helicity of such vortex beams, the photocurrent in the radial direction changes polarity. This is a direct evidence that the electronic states are not only susceptible to the amplitude of the electric field but also its phase. Moreover, this shows that the electronic wavefunction is phase coherent around the hole of the annulus. We are currently collectcting more data (various magnetic field, backgate sweeps, bias fields, etc.) and performing control experiments to increase our confidence in our claims.

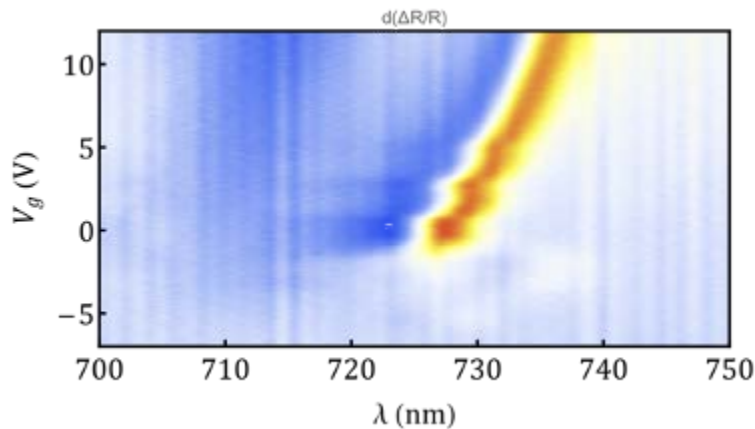




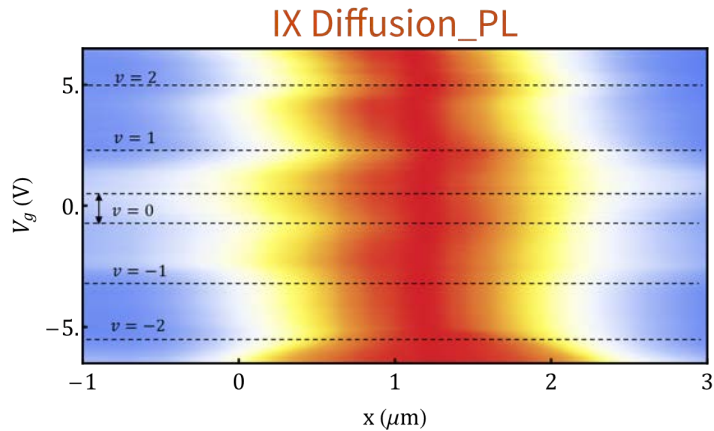
The difference between photocurrent from the OAM = +1 beam and the OAM = -1 beam for $B = 9T$. The shaded regions correspond to the peaks in σ_{xx} with included peak widths.

Mott-Moiré excitons:

Following our theoretical work in understanding the nature of Mott-excitons in TMDs. [Mott-moiré excitons, T.-S. Huang, Y-Z Chou, C. L. Baldwin, F. Wu, M. Hafezi, arXiv:2207.13152 (2022)], we have been performing experiments on bilayer WS₂/WSe₂. We have reproduced the incompressibility signature in our samples (see below), similar to previously reported data, around 4 Kelvin temperature. We are currently studying these Mott-excitons in lower temperatures that is only accessible in a dilution fridge (30mKelvin). Our goal is to understand the interplay between charge order and spin orders in such systems. For that, we are currently performing diffusion and lifetime measurements, and we believe that by going to such low temperatures we can differentiate between charge and spin orders.



WSe2 reflectivity spectrum shows a blueshift for integer fillings. This could be explained because of screening effect from the other layer (electron doping of the WS2 layer) is suppressed due to charge order. Such a picture may be modified when the spin physics becomes important at lower temperatures.



Interlayer exciton diffusion map for a range of backgate voltage (Fermi level). The Interlayer diffusion length is strongly suppressed under integer charge filling factors. The asymmetry in the right side is because of the material's surface inhomogeneity.

[end]