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Research Scholarship for International Graduate Students

Two-Dimensional Semiconducting Transition Metal Dichalcogenides: Light-Matter Interaction and Photodevice Application

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Abstract

The single-layer form of 2D transition metal dichalcogenides (TMDs) exhibits direct bandgap, high photoluminescence (PL) quantum efficiency, and spin-valley coupling-related properties, making them an excellent platform to investigate interesting optical properties. This work takes further experimental investigation and describe the initially unexplored phenomena arising from the interaction of light carrying orbital angular momentum (OAM) with 2D TMDs. Monolayer (ML) molybdenum disulfide (MoS_2) – a prototypical 2D TMD – was subjected to interaction with incident light having distinct properties. Consequently, it was observed that an impinging light with OAM had caused a selective photoexcitation of the exciton quasiparticles manifested by the blue peak energy shifts of the recorded PL intensity. The effects of light with OAM were further investigated and found to have controlled the photovoltaic properties of a MoS_2-channeled photodevice. Such observations imply that light with certain properties may facilitate onto the ML MoS_2 additional degrees of freedom useful for data storage, enhanced energy harvesting, and sensing applications.

Twisted Light

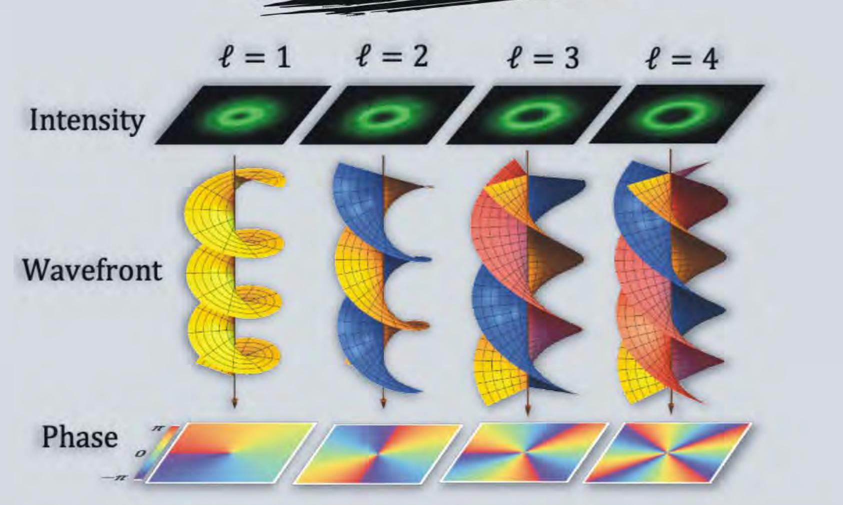


Figure 1. Twisted Light intensity, wavefront, and phase patterns at different ℓ

- Has **orbital angular momentum of $\ell\hbar$** per photon
- Enhances **quadrupole transitions** in atoms & semiconductors
- Can easily be generated in laboratories

1-layer MoS_2

- Exhibits a bandgap w/energy within the **visible light range**
- Enables formation of tightly bound **excitons**
- High optical absorption coefficient

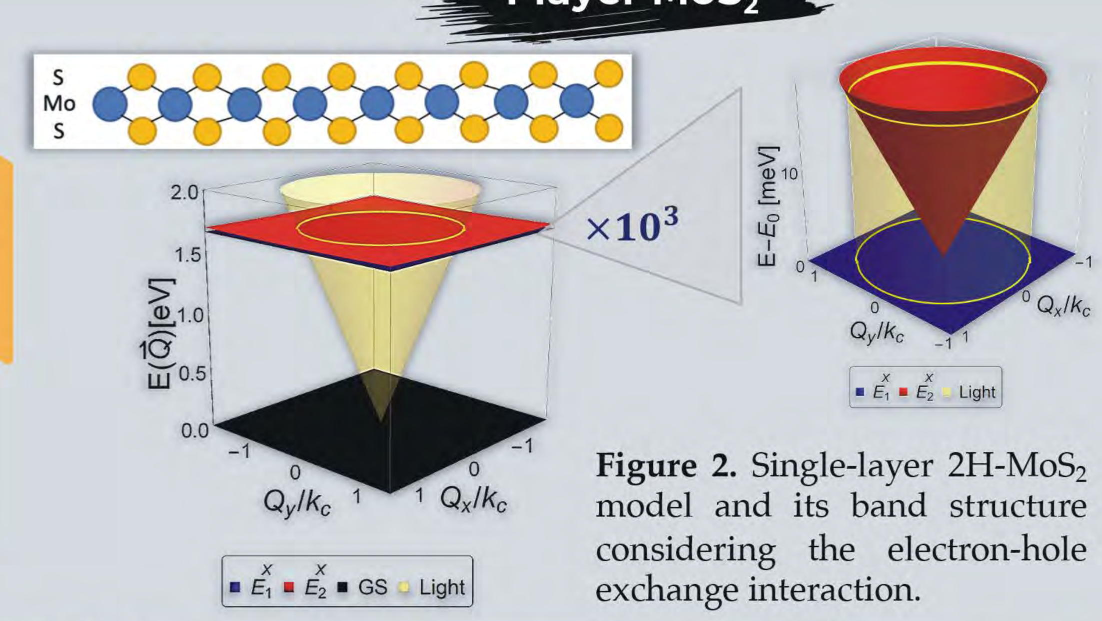


Figure 2. Single-layer 2H-MoS_2 model and its band structure considering the electron-hole exchange interaction.

Experiment Setup

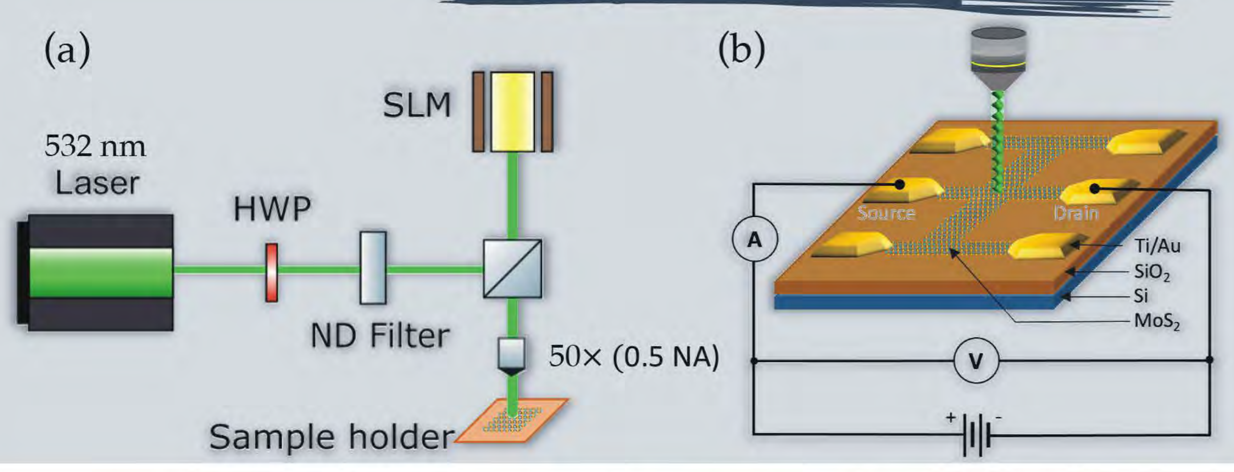
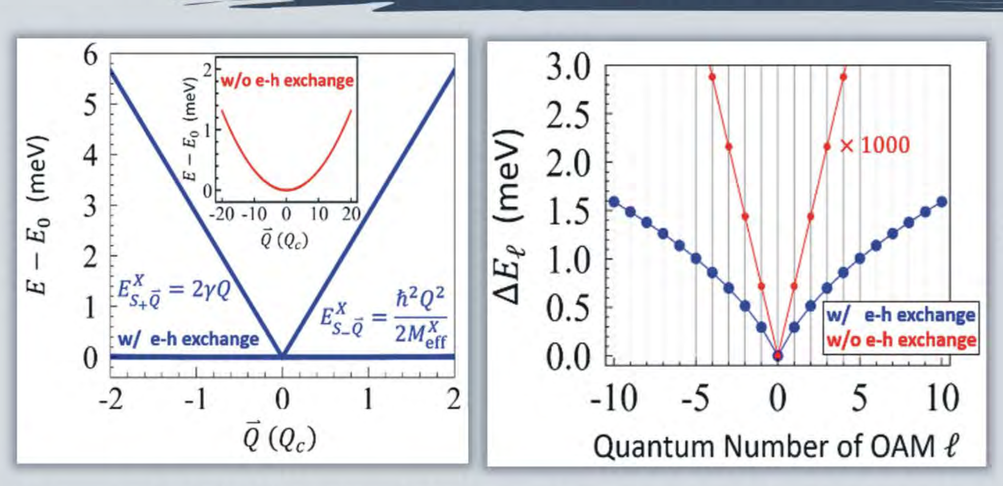


Figure 3. (a) Optical measurement setup and the (b) electrical measurement setup. The spatial light modulator (SLM) converts plane wave light to twisted light.

Twisted Light-Induced Spectral Shifts



(Shifts in the upper band)

$$\Delta E_\ell^{S+} \equiv \left(\frac{\sqrt{2\pi}\gamma}{W_0}\right)\left[\frac{1}{2^{|\ell|+1}}\frac{(2|\ell|+1)!!}{|\ell|!}-\frac{1}{2}\right]$$

(Shifts in the lower band)

$$\Delta E_\ell^{S-} \equiv \left(\frac{\hbar^2}{2M_{\mathrm{eff}}^X W_0^2}\right)|\ell| \propto |\ell|.$$

Figure 4. (a) The simulated exciton energy dispersion of single-layer MoS_2. (b) The calculated PL spectral shifts induced on the single-layer MoS_2 by the incident twisted light with well-defined $\ell\hbar$.

- **As one may notice, a signature of the linear exciton dispersion is the non-linear trend of the ℓ −dependent PL spectral shift.**

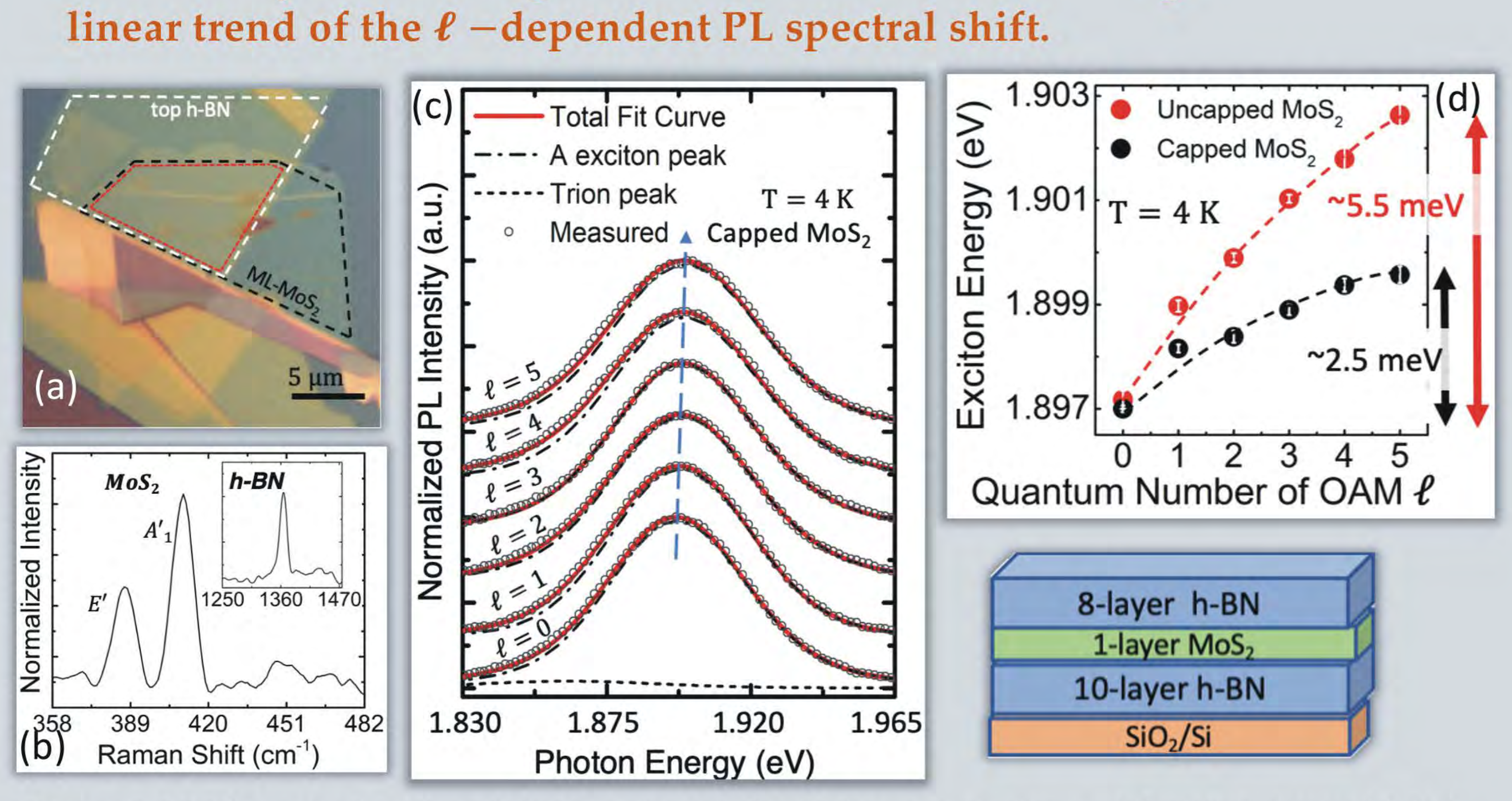
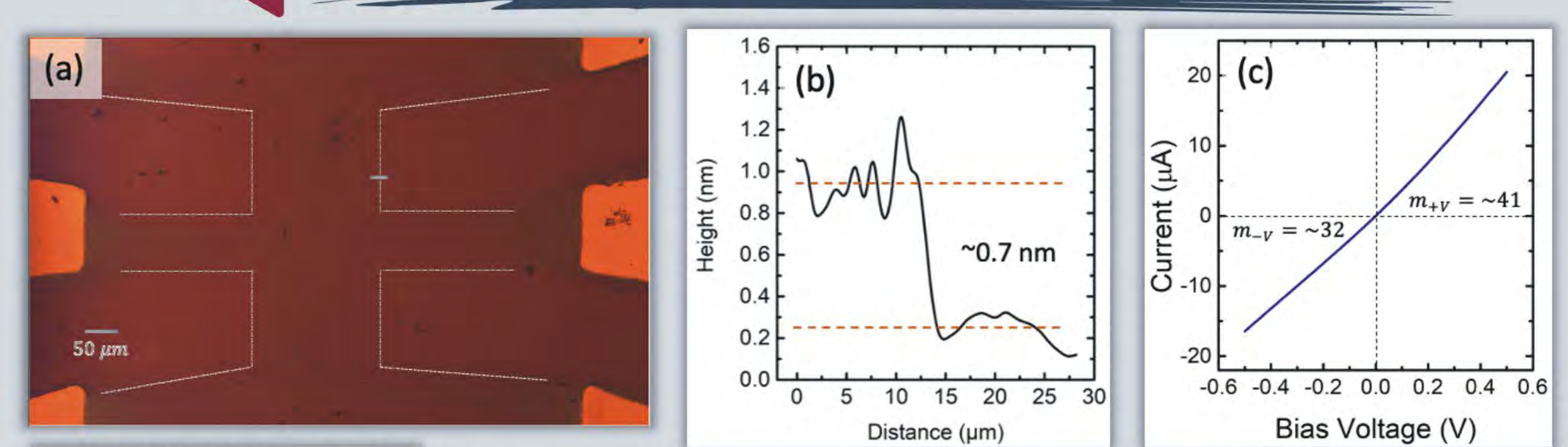


Figure 5. (a) The optical image of the sample and the (b) Raman spectrum of the MoS_2 layer. (c) The PL spectra of the 20 μW laser excited h-BN capped MoS_2 sample at different ℓ at 4 K. (d) The summary of the peak spectral shifts of the uncapped versus h-BN capped sample at various ℓ at 4 K.

Twisted-Light-Enhanced Photovoltaic



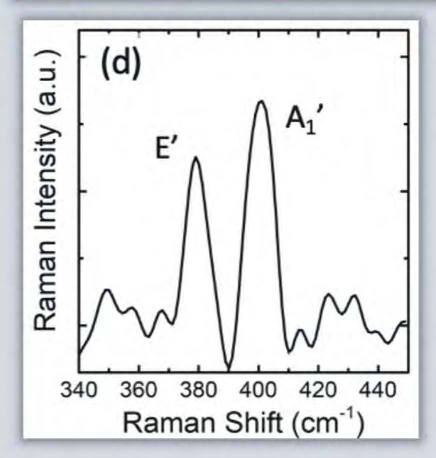


Figure 6. Device structure and characteristics. (a) Optical image of the single-layer MoS_2 channel device. **(b)** The height profile of the region crossing the border between the MoS_2 film and the SiO_2 substrate as indicated by a white bar in **(a)**. **(c)** The current-voltage curve in dark condition, evidencing the existing p-n junction in the device. **(d)** Raman spectra of the MoS_2 channel.

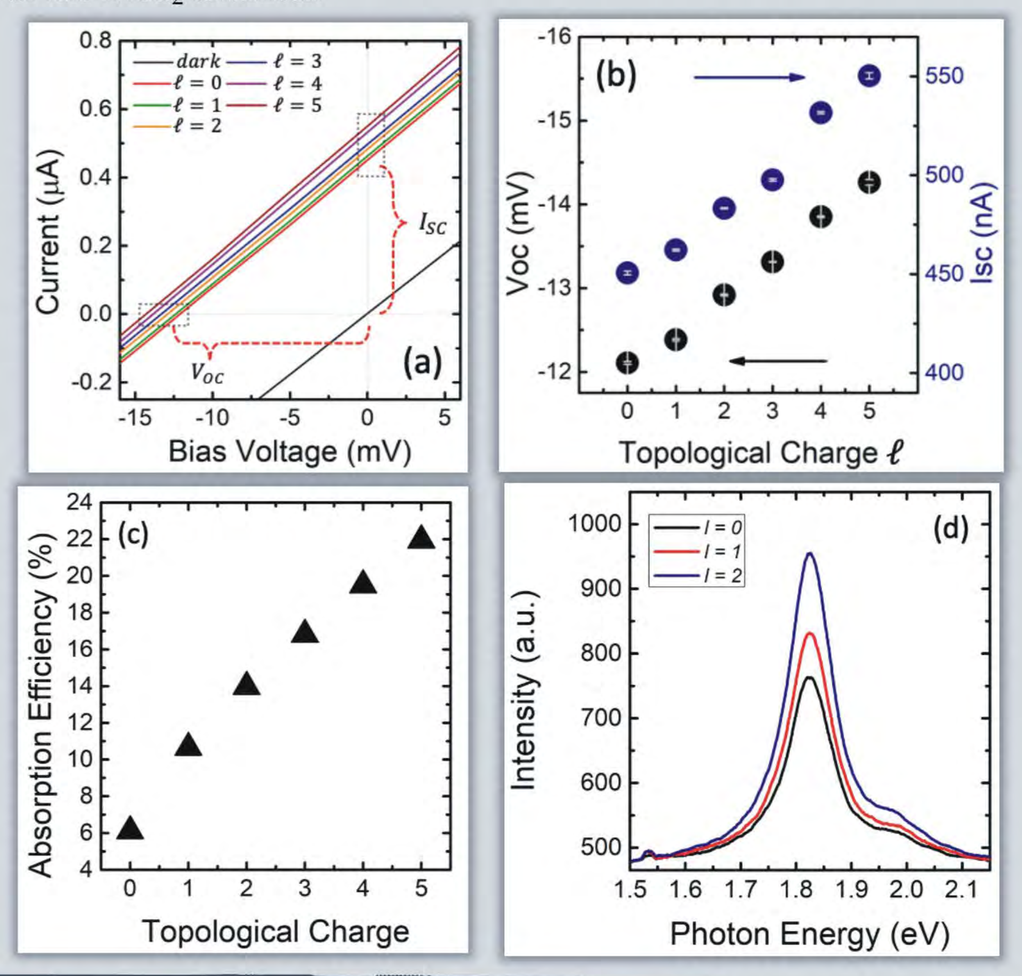


Figure 7. Enhancement of the photovoltaic effect in single-layer MoS_2. (a) Drain-source current versus bias voltage (I-V) characteristic of the device under dark and under illumination of light carrying different ℓ (20 μW laser power, 532 nm wavelength); $\ell = 0$ refers to the fundamental mode beam. **(b)** The corresponding short-circuit currents (I_{SC}) and open-circuit voltages (V_{OC}) at various ℓ conditions. **(c)** The estimated absorption efficiency of the MoS_2 channel. **(d)** Photoluminescence (PL) spectra of the triangular MoS_2 sample illuminated by incident light with selected values of ℓ and fixed laser power (20 μW).

Summary and Selected Publications

This work reported optical properties of valley excitons in ML-MoS_2 due to its interaction with twisted light. Noticeable blue shifts were observed as the magnitude of the $|\ell|$ of the twisted light was incremented. The study revealed the signature of the predicted lightlike exciton dispersion of the valley exciton in 2D MoS_2. Enhancements of the photovoltaic properties of a monolayer MoS_2 – channeled device was demonstrated, which may be a manifestation of an improved absorption in MoS_2 due to the OAM of light.

Selected Publications: [1] Selective photoexcitation of finite-momentum excitons in monolayer MoS_2 by twisted light. ACS Nano, 15 (2), 3481-3489. (2021); [2] Twisted Light-Enhanced Photovoltaic Effect. ACS Nano, 15, 14822-14829. (2021)