

Twisted Light-Driven Exciton Dissociation for Enhanced Photoresponse in Monolayer MoS₂ Transistors

Ye-Ru Chen, Kristan Bryan Simbulan, Guan-Hao Peng, Yu-Chen Chang, I-Tong Chen, Han-Chieh Lo, Shao-Yu Chen, Shun-Jen Cheng, Ting-Hua Lu,* and Yann-Wen Lan*



Cite This: *ACS Nano* 2025, 19, 18282–18291



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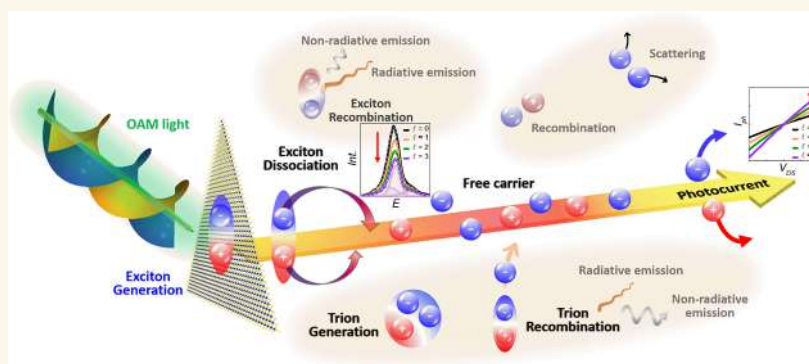
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ABSTRACT: Two-dimensional monolayer transition metal dichalcogenides (TMDs) exhibit strong exciton binding energy due to Coulomb interactions, making exciton dissociation challenging. However, the use of orbital angular momentum (OAM) light, or twisted light, enables momentum-conserving transitions, potentially enhancing exciton dissociation and improving optoelectronic performance. In this work, we simultaneously explore the optical and electrical characteristics of a field-effect transistor (FET) fabricated from molybdenum disulfide (MoS₂) when exposed to OAM-carrying illumination. A significant reduction in exciton luminescence rates is observed, whereas a substantial enhancement in the device's conductance is detected as the OAM order of light is increased. Light with OAM effectively slows exciton recombination, as confirmed by time-resolved photoluminescence, while concurrently strengthening the probability of exciton dissociation. This shift in the balance between exciton recombination and dissociation is inferred to as the driving force behind the improved free carriers in the device. In addition, light-carrying OAM slightly improves the material's light absorption by facilitating additional transitions that were normally inaccessible. The implications of our study extend to the potential improvement in the performance of phototransistors, showcasing the multifaceted benefits of harnessing OAM light for advanced applications in optoelectronics.

KEYWORDS: orbital angular momentum, transition metal dichalcogenides, MoS₂ FET, twisted light, phototransistor

Semiconductors, including silicon (Si), gallium nitride (GaN), zinc oxide (ZnO), perovskite, and two-dimensional (2D) materials, have been pivotal in shaping modern electronics by fueling the performance of transistors,¹ high-frequency devices,² optoelectronics,³ and solar cells.⁴ As researchers delve into novel materials and cutting-edge technologies, they aim to create smaller, faster, and more efficient devices. Among these materials, 2D TMD materials stand out due to their unique attributes, a 2D nature, direct bandgap, high exciton dissociation capability, advantages in absorption efficiency, flexibility, and the generation of free

carriers. In particular, MoS₂ FETs have garnered considerable interest due to their immense potential in nanoelectronics and optoelectronics.^{5–10} These studies have unveiled fascinating

Received: December 18, 2024

Revised: April 29, 2025

Accepted: April 30, 2025

Published: May 8, 2025



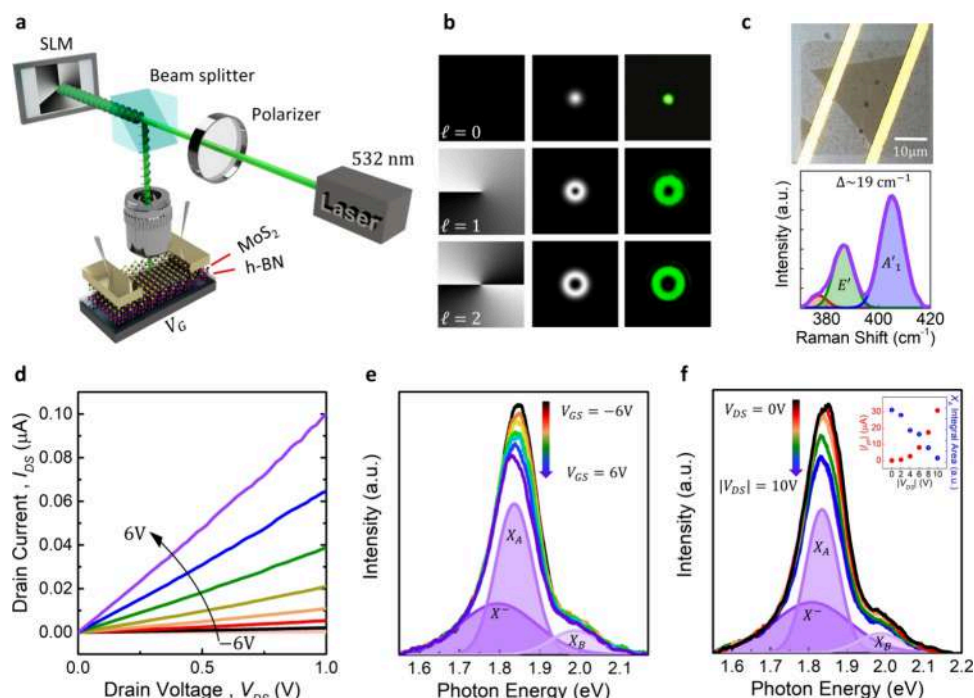


Figure 1. Experimental setup, device structure, and its electrical and optical properties. The experimental setup depicted in a), utilizes a 532 nm excitation source irradiated on an SLM to generate light that carries OAM, which is then focused on a MoS₂ FET using an objective lens. The laser spot size that irradiates the device under the condition of $l = 0$ is about 1 μm of the radius. The nine-square array in b), illustrates the characteristics of the LG beam with OAM ranging from 0 to 2 (top to bottom). The columns demonstrate the phase (left), simulated intensity (middle), and practical irradiation distribution (right) in the transverse plane. c), the top panel presents an optical microscope image of a single-layer MoS₂ on h-BN, while the bottom panel displays the Raman spectrum evidencing the presence of a monolayer MoS₂ in the FET. d), the electrical characteristics under a dark condition at V_{GS} ranging from -6 to 6 V with steps of 1 V. e), PL spectra are measured at different levels of V_{GS} , with the highest (lowest) exciton recombination rate observed at a relatively low (high) background carrier concentration. f), PL results while simultaneously applying V_{DS} with $V_{GS} = 0$. The inset shows the comparison between measured current flow and exciton recombination rate, which turns out to have a complementary relation to each other.

characteristics, revealing that the magnitude of the photocurrent is intricately tied to factors such as light intensity, wavelength, and polarization. These distinctive photoresponse properties have paved the way for a plethora of exciting applications, including the creation of optoelectronic devices such as photodetectors, photovoltaics, and integrated optical circuits.^{11–14} The capacity to seamlessly integrate electronic and optoelectronic functionalities within a single device positions MoS₂ FETs as a versatile platform poised to drive future technological advancements.

Orbital angular momentum (OAM) is an intriguing concept in the realm of optics, describing the phase rotation of a light wave around the propagation axis.¹⁵ Unlike spin angular momentum, which arises from the circular polarization of light, OAM derives its existence from the helical phase front of the wave in the spatial domain.¹⁶ This helical structure imparts a unique twist or corkscrew-like behavior to the wavefront, resulting in a photon that appears to rotate as it propagates. What distinguishes OAM is the presence of discrete quantized values, termed OAM states or modes. Each mode is characterized by an integer value, commonly represented as l , signifying the number of twists or helical wavefront within the light beam, and each OAM mode carries an orbital angular momentum of $l\hbar$ per photon.^{16–19}

The interaction of light with OAM produces practical effects, significantly influencing material absorption and emission properties, and altering their photonic response. OAM induces a substantial increase in open-circuit voltages

and short-circuit current in MoS₂ photovoltaic devices, intensifying light absorption and generating more photon-excited carriers.²⁰ One noteworthy outcome of this interaction is the selective excitation of excitonic transitions within MoS₂, manifesting as a discernible blue shift in exciton photoluminescence (PL).²¹ OAM can selectively couple to specific excitonic transitions, leading to improved optical absorption based on the OAM mode and the intrinsic excitonic characteristics of MoS₂.²² Beyond the 2D TMD materials, the enhancement of photonic absorption by OAM-carrying light has also been observed in silicon-nanowire FET, resulting in an increase in photocurrent.²³ Our previous research, also unveiled that the overall probability of trion-to-exciton formation can be modulated by OAM light.²⁴ From a more profound perspective, the photocurrents stemming from the helical phase of the OAM-carrying beam offer proof of the transfer of OAM to electrons and unveil the correlated phase-related data in photodetection.²⁵ Exploring the intricate interplay between OAM-carrying light and monolayer MoS₂ holds great promise for a variety of applications.

In this study, the OAM light of 532 nm is directed onto a MoS₂ FET to excite the MoS₂ channel, revealing a reduction in the radiative emission rate of excitons with increasing OAM order and a simultaneous gradual enhancement in photocurrent due to the rising OAM of light. Meanwhile, our time-resolved PL (TRPL) results indicate a prolonged exciton lifetime corresponding to the increment of the OAM of light. Accordingly, we explore the intricate relation between exciton

optical emission and the optoelectronic properties of MoS₂ FET. It reveals that an increase in the OAM carried by the excitation source suppresses the exciton optical emission rate while enhancing the exciton dissociation, ultimately leading to an improvement in the device's photoconductance.

RESULTS

OAM of Light and MoS₂ FET. We mainly harness the capabilities of a 532 nm laser with linear polarization as the excitation source, as shown in Figure 1a. The spatial light modulator (SLM) with a computer-generated hologram is used to generate light with OAM, and the light possessing OAM is directed toward the MoS₂ transistor. The monolayer MoS₂ transistor, supported by a gate insulator comprising ~5 nm h-BN on top of a 30 nm SiO₂ layer, is chosen for its reported reduction in the number of charge traps compared to only SiO₂.²⁶ The OAM of light originates from the azimuthal index (*l*) in the Laguerre-Gaussian (LG) profile, and the order for *l* periods of phase within a complete 2 π azimuthal angle, as depicted in the left column of Figure 1b. This periodicity in the phase leads to the formation of a wavefront with a unique twisted light beam. The winding of the azimuth introduces a singularity at the center of the beam, resulting in null intensity. The middle column of Figure 1b displays the simulated intensity distribution of the OAM-carrying light for various values of *l*, ranging from 0 to 2, and the right column provides practical irradiation intensity captured via charge-coupled device (CCD), revealing that as the order of *l* increases, the winding intensifies, giving rise to a more expansive donut-like beam. Figure 1c provides an optical microscopy image of monolayer MoS₂ FET (top), where the channel length is approximately 16 μ m, and the device area is ~320 μ m². The bottom in Figure 1c shows the Raman spectrum of two prominent phonon modes with their peak position difference around 19 cm⁻¹, revealing the evidence of monolayer MoS₂. The device is measured under dark conditions with back gate voltage (*V*_{GS}) varying from -6 to 6 V with steps of 1 V. The device's conductance exhibits gradual increments, indicative of modulation in the device's background carrier concentration and the intrinsic n-type characteristics, as demonstrated by the *I*_{DS} - *V*_{DS} curve presents in Figure 1d. Figures 1e and 1f showcase the monolayer MoS₂ PL results, providing insights into the modulation of excitonic transitions through external out-of-plane and in-plane electrical fields, respectively. Considering the diminishing dimensions of materials, the consequential reduction in Coulomb screening effects results in a substantial elevation of the exciton binding energy, often reaching magnitudes on the order of hundreds of meV.²⁷ The elevated binding energy effectively prevents the spontaneous and thermal dissociation of excitons.²⁸ Even with increased exciton density and environmental temperature, excitons in monolayer MoS₂ are more likely to undergo radiative recombination, resulting in PL compared to few-layer or bulk MoS₂. The determination of PL quantum yield (PLQY) depends on the ratio of radiative and nonradiative recombination rates for both trions and excitons relative to the overall generation rate *G*, which is associated with the incident power. The relation can be expressed as,²⁹

$$PLQY \propto \frac{1}{G} \left[\frac{n_X \tau_{Xr}}{\tau_{Xr} + \tau_{Xnr}} + \frac{n_T \tau_{Tr}}{\tau_{Tr} + \tau_{Tnr}} \right] \quad (1)$$

In this expression, *n*_X and *n*_T symbolize the carrier concentrations of excitons and trions, while τ_{Xr} (τ_{Xnr}) and τ_{Tr} (τ_{Tnr}) denote the radiative (nonradiative) lifetime of excitons and trions, respectively.

The rate of exciton recombination exhibits a nonlinear dependence on the background carrier concentration, a parameter controlled by the back gate voltage, which generates the out-of-plane electric field. Application of a positive back gate voltage ushers in electrons as the majority charge carriers, resulting in the generation of negative trions. Since the dominant recombination pathway for trions is nonradiative, these trions suppress the exciton's radiative recombination, leading to a decline in exciton emission while radiative trions persist. On the contrary, a negative back gate voltage suppresses electrons, diminishing the formation of negative trions. Under conditions of low background carrier concentration, the radiative recombination of excitons becomes highly probable, resulting in heightened PL intensity as shown in Figure 1e.

Beyond the influence of background carrier concentration, the dissociation rate of electron-hole pairs can be modulated by the application of an in-plane electric field, a phenomenon known as tunnel ionization. The exciton dissociation rate depends on the in-plane electrical field.²⁸ As the in-plane field tunes the band structure between the two terminals, electrons can tunnel through the potential barrier, which arises from the exciton binding energy, into the conduction region. The tunneled electrons elevate the free carrier concentration, a relation reported to be proportional to the exciton dissociation efficiency. The PL measurements are conducted with a simultaneously applied bias voltage (*V*_{DS}) to generate an in-plane field while extracting photons emitted from exciton recombination. The outcomes in Figure 1f reveal a decline in PL intensity as the potential difference (*V*_{DS}) between the two electrodes increases without applying *V*_{GS}. Concurrently, the device current under a specific bias voltage has been recorded, as shown in the inset, indicating that the photocurrent is inversely proportional to the A-exciton integral area, which denotes its recombination rate. This provides evidence supporting the idea that the increase in free carrier concentration can be attributed to the enhanced efficiency of exciton dissociation. It is essential to highlight that the observed trend remains consistent under both negative and positive bias voltages (refer to Supporting Information Figure S1, which also presents results from another device), and a constant number of incident photons is maintained for each measurement.

OAM-Carrying Light Affects Photon-Excited Free Carriers and Excitons. To explore the photoresponse of the MoS₂ device under the influence of twisted light, Figure 2a illustrates the *I*_{ph} - *V*_{DS} characteristic, revealing a consistent increase in device current as the OAM order of light varies from 0 to 3. To ensure the same quantity of impinging photons under each OAM condition, the total incident power is fixed. Note that the photocurrent is directly obtained from the equation *I*_{ph} = *I*_{illu} - *I*_{dark}. The *I*_{DS} - *t* measurement using another MoS₂ device is also detailed in Supporting Information Section 2. The measured device photocurrent (*I*_{ph}) correlates directly with the free carrier concentration (*n*) and is represented as,

$$I_{ph} = q(\mu_n n + \mu_p p)EWD \quad (2)$$

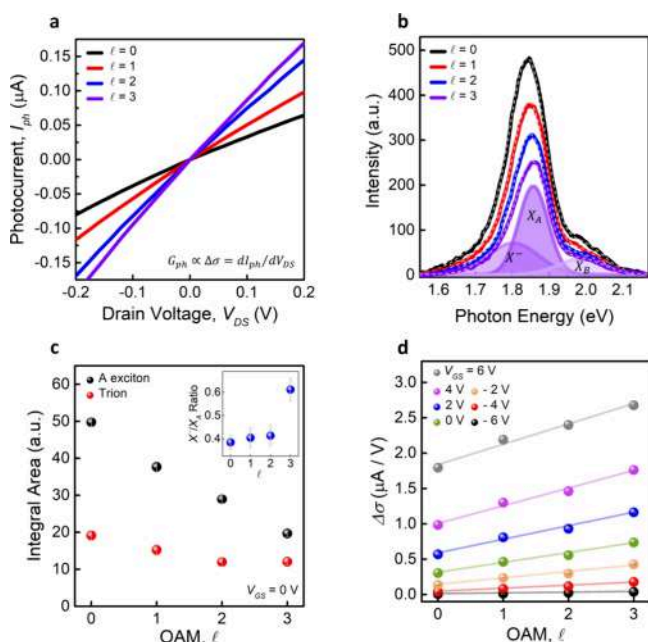


Figure 2. OAM-carried light affects photon-excited free carriers and excitons. a), the photocurrent (I_{ph}) versus applied bias voltage (V_{DS}) with different irradiation conditions ($l = 0$ to 3 , fixed incident power of $300 \mu\text{W}$). b), PL spectra measured at different OAM of light, indicating a decrease in the PL intensity as the OAM of the excitation source gets higher. The dashed line with a light color denotes the fitted curve. c), the fitted results from the PL spectra, reveal a decline in the exciton and trion integral area as the OAM of the incident light increments. The inset depicts the estimated trion-to-exciton concentration ratio, which shows a higher ratio of photons are emitted from the trion at higher OAM excitation sources. d), the device increased conductance extracted from different OAM of excitation sources at certain back gate voltages, indicating an enhancement of photoconductance while using higher OAM of light, which is improved by the modulation of carrier concentration.

Here, q is the elementary charge, μ_n (μ_p) signifies the carrier (hole) mobility, while E denotes the in-plane electrical field generated by the applied V_{DS} , W and D indicate the channel width and thickness, respectively. It is worth noting that electrons overwhelmingly dominate the current flow, and the contribution of hole carriers (p) is disregarded. The augmentation of photocurrent is then assumed to be attributed to the rise of n . The interaction between twisted light and 2D materials, as analyzed through the calculation of the exciton-light matrix, reveals the transfer of OAM from photons to the center-of-mass (CoM) momentum of excitons.²² The conservation of total angular momentum necessitates the exciton's CoM momentum to assume a magnitude of $l\hbar$, equivalent to the angular momentum of the incident photon. This additional momentum in the exciton state may shift the recombination path from direct to indirect bandgap and improve the dissociation probability of the exciton bound state, resulting in a decrease in the radiative recombination rate of excitons, as evident in Figure 2b. Figure 2c compares the photons emitted by excitons (X_A) and trions (X^-). The inset illustrates the trion-to-exciton concentration ratio ($\frac{X^-}{X_A}$), which can be related to the generation of free carriers.³⁰ As the OAM rises, the radiative recombination rate of excitons exhibits a gradual descent, and the ratio between trions and excitons increases. It

is important to note that the radiative recombination rate of trions remains relatively unaffected. The fitting parameters and the report table for the PL results with the order of OAM ranging from 0 to 3 are all provided in Supporting Information Section 3 and Table S1.

The photoconductance suggests that the photogenerated free carrier concentration, Δn , linked to the device conductance, leads to an enhancement of conductivity, $\Delta\sigma$, expressed as,

$$\Delta\sigma = q(\mu_n \Delta n) \quad (3)$$

This enhancement results from the combined influence of the photoconductivity and photogating effects. The photoconductivity effect enhances conductivity by generating additional photocarriers, while the photogating effect arises when photogenerated carriers are trapped in defect states or trap sites. These trapped carriers induce a persistent built-in electric field, effectively modulating the carrier concentration in the channel, similar to the effect of applying a gate voltage (V_{GS}). The dominant mechanism can be identified through the $I_{ph} - V_{GS}$ curve. If the photocurrent varies with gate voltage, the photogating effect dominates; conversely, if the $I_{ph} - V_{GS}$ curve remains flat (i.e., the photocurrent is independent of V_{GS}), the photoconductive effect is dominant. Within our operating gate voltage range ($\pm 6 \text{ V}$), the photogating effect is the primary contributor to photoconductance, as evidenced in the $I_{ph} - V_{GS}$ curve in Figure S4a. To minimize external influences, such as thermal effects that may arise under higher bias voltage (V_{DS}) conditions, the device conductance enhancement is extracted from the slope of the electrical characteristics within the $V_{DS} = \pm 0.2 \text{ V}$ range in Figure 2a. As shown in Figure 2d, the increase in device conductance varies with the applied gate voltage. The gate-dependent behavior of the device conductance indicates that the modulation of the background carrier concentration further influences the enhancement of the photocurrent under OAM light. The same results can be found in the other MoS_2 device as provided in Supporting Information Figure S5. To better understand how the OAM of light affects the results, it is important to consider the exposure area, as different OAM states result in varying exposure sizes. To eliminate this effect, we also examined cases where the power density remained constant across different OAM states. The results, shown in Supporting Information Figure S6, are consistent with those observed under fixed power intensity. Additionally, at higher OAM orders, we still observe a reduction in PL intensity, highlighting the significant influence of OAM light on exciton recombination rates.

Power Dependency of the Device's Conductance with Varying OAM of Light. The power-dependent behavior of device conductance, in Figure 3a (linear) and 3b (logarithmic), are elucidated in the absence of modulation of the background carrier concentration. The conductance exhibits a rapid upswing at low-power levels followed by a gentler increment at high-power, aligning with previously reported observations.^{20,23} This behavior is ascribed to the space-charge effect, wherein the excitation rate saturates as the intensity of the incident light surpasses a certain threshold. The space-charge effect refers to the accumulation of electric charges within a region of space. This phenomenon occurs when charged particles, such as electrons or ions, are present in a confined space, and their collective electric fields influence the motion and behavior of other charged particles in that

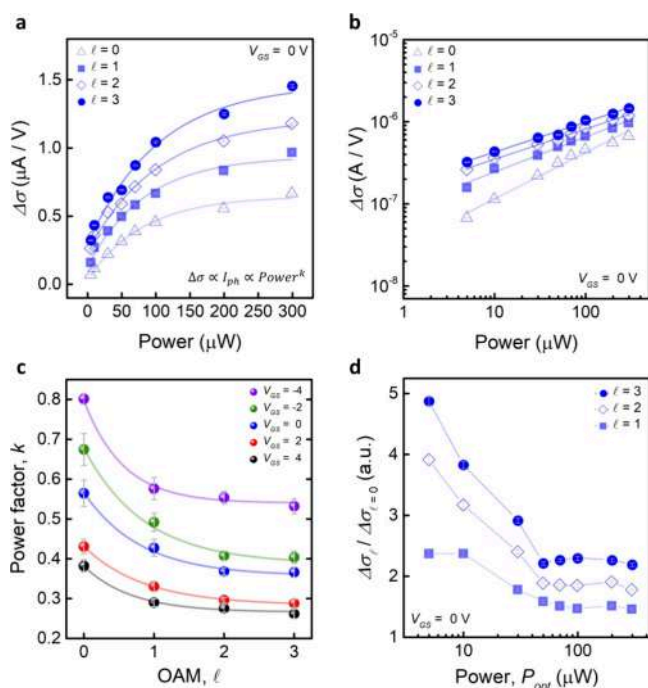


Figure 3. Power dependence of the device's conductance with varying OAM. a), the power dependency of the $\Delta\sigma$ under several light conditions (OAM ranging from 0 to 3 with different symbols) present in linear scale and b) in logarithmic scale. In c), the power factor k , which is extracted from the relation between conductance and incident power, decreases as the OAM-carried light and background carrier concentration increase. d), The OAM-enhancing rate of photoconductance compared to the Gaussian beam ($\frac{\Delta\sigma_l}{\Delta\sigma_{l=0}}$) with the absence of V_{GS} .

space. In a semiconductor, especially when it is subjected to an external electrical field, the generated carriers experience it. As these carriers move through the channel, they create an electric field due to their charge distribution. To gain a deeper insight into the device's power-dependent behavior with OAM of light, we analyze the interplay between $\Delta\sigma$, I_{ph} , and incident laser power (P_{opt}). This analysis allows the establishment of the power factor (k) through the power law, stating that $\Delta\sigma \propto I_{ph} \propto P_{opt}^k$. The k serves as a metric to gauge the material's photoresponse efficiency when concerning power dependence. A lower value of the power factor indicates an early manifestation of the space-charge effect,^{31–33} signifying a diminishing transformation efficiency of photocurrent with increasing incident power. The estimated values of the k are illustrated in Figure 3c, derived from linear fitting slopes of the double logarithmic-scale plot in Figure 3b, for different illumination conditions ($l = 0$ to 3) at $V_{GS} = 0$ V and other background carrier concentrations ($V_{GS} = -4$ to 4 V) shown in Supporting Information Figure S7. The outcomes reveal an early onset of the space-charge effect as both the OAM of light and the background carrier concentration increase. Using OAM light results in a higher number of excited electrons, similar to the effect of applying a higher V_{GS} . This increased electron population contributes to the space-charge effect and leads to a reduction in k . An elevated V_{GS} implies a greater concentration of electrons near the conduction band, increasing carrier scattering and thereby limiting conductance enhancement due to the pronounced space-charge effect. Similarly, when higher-order OAM light is employed, a lower k

is observed, likely due to the increased number of excited electrons in the conduction band compared to a Gaussian beam ($l = 0$).²³ Consequently, OAM light facilitates the enhancement of device conductance. The enhancing rate of photoconductance, employing OAM at $V_{GS} = 0$ V, is shown in Figure 3d. The ratio $\frac{\Delta\sigma_l}{\Delta\sigma_{l=0}}$ represents the elevation rate of OAM light relative to the fundamental mode. In the low-power range, there is a relatively high rate of photocurrent generation under OAM illumination. Nevertheless, this rate decreases rapidly, and the variation becomes stable as power changes in the high-power region. Note that similar results, but with a more enhanced ratio at low carrier concentration (negative V_{GS}), are demonstrated in Supporting Information Figure S8 due to the suppression of the space-charge effect. This reveals that OAM improves the photon-excited carrier concentration and provides more efficient photocurrent transformation, which is strongly relevant to the material's absorption and exciton dissociation. The role of OAM of light in generating an elevation of photocurrent will be further discussed in the following sections.

DISCUSSION

The transverse structure of twisted light not only endows it with OAM characteristics but also imparts in-plane linear momentum. The amplitude of the vector potential of twisted light in the angular spectrum representation^{21,22} is given by

$$A_l(r) = \sum_{q_{\parallel}} \mathcal{A}_l(q_{\parallel}) e^{iq_{\parallel}r} \quad (4)$$

where $\mathbf{q} = \mathbf{q}_{\parallel} + \mathbf{q}_0$ represents the linear momentum of each plane-wave component, with $\mathbf{q}_{\parallel}$ and $\mathbf{q}_0$ denoting the in-plane and out-of-plane components of $\mathbf{q}$ for normally incident twisted light, respectively. The weight function is expressed as $\mathcal{A}_l(q_{\parallel}) = F_l(q_{\parallel}) e^{ilq_{\parallel}}$, where $F_l(q_{\parallel})$ and $e^{ilq_{\parallel}}$ represent the OAM-dependent radial profile and azimuthal phase in $\mathbf{q}_{\parallel}$ -space, respectively. For twisted light with finite OAM, $F_l(q_{\parallel})$ exhibits a maximum magnitude at $q_{\parallel} \neq 0$, demonstrating that twisted light carries in-plane linear momentum.^{21,22} By the linear momentum conservation law, the plane-wave components with $\mathbf{q}_{\parallel} \neq 0$ induce nonvertical transitions and generate excitons with nonzero CoM momentum.^{21,22} The excitons with nonzero CoM momentum can couple to light with nonzero in-plane linear momentum,^{22,34} which is emitted at a tilted angle relative to the 2D material plane. These additional transitions induced by OAM light often exhibit longer lifetimes or slower decay rates for excitons, because the dipole moments of the exciton states with nonzero momenta do not have the full alignment with the transverse polarizations of the tilted emitted lights and possess slower spontaneous decay rates.^{22,35,36} The results are not only associated with weaker or suppressed optical emissions but also revealed the improvement of exciton dissociation efficiency and then evaluate the free carrier concentration. The free carrier concentration (n) of the device can be determined from the variation in threshold voltage (V_{TH}) observed in the $I_{ph} - V_{GS}$ curve as a function of the topological charge (as detailed in Supplementary Section 4). As the order of OAM increases, the free carrier concentration of the device gradually rises. The increase in free carrier concentration (Δn) induced by illumination can also be described as follows,²⁰

$$\Delta n = \eta \frac{P_{\text{opt}} \lambda}{A_{\text{ph}} h c} \tau \quad (5)$$

In the above equation, η denotes the absorption efficiency, P_{opt} and A_{ph} symbolize the incident power and the illumination area, λ indicates the excitation source wavelength, τ signifies the free carrier lifetime, h and c represent the Planck constant and the velocity of light, respectively. The measurements are conducted with a fixed incident power intensity across diverse OAM conditions. According to eq 5), while the wavelength of incident light is secured, the elevation of carrier concentration can be attributed to the effects of improved absorption efficiency or extended carrier lifetime. Figure 4a presents the

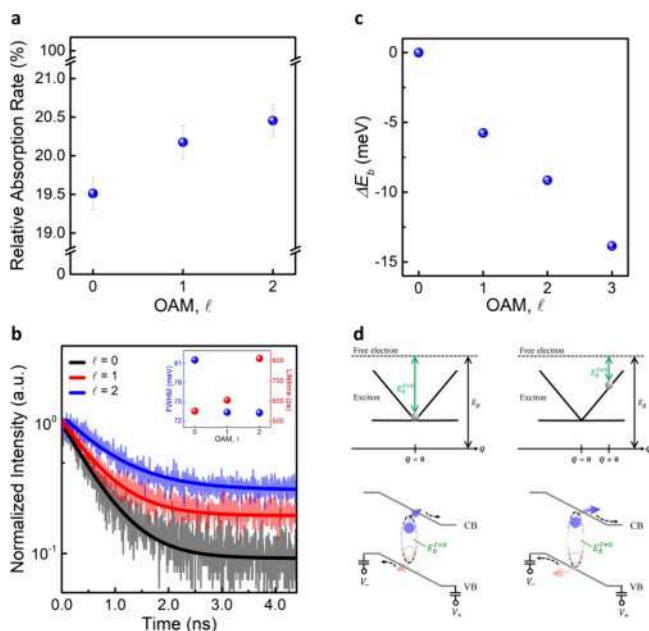


Figure 4. Optical absorption of MoS₂, exciton relative binding energy, and radiative lifetime. a), The estimated change of relative absorption rate from the reflectivity (R), $\frac{\Delta R}{R} = \frac{R_B - R_A}{R_B}$, in which R_A (R_B) denotes the reflectivity after the laser illuminates onto MoS₂/h-BN/SiO₂/Si (h-BN/SiO₂/Si) structures. b), The TRPL results show an extended lifetime of the exciton as the OAM of light increases. The inset figure presents a comparison between the exciton lifetime extracted from TRPL and the fwhm from the PL measurement. c), The estimated relative exciton binding energy change increased in the presence of OAM in light compared to the exciton generated by the fundamental Gaussian beam. d), the configuration of exciton CoM momentum (Q) with and without the excitation of OAM (top), and the illustration of exciton dissociation under applied in-plane field with expanded exciton radius that is excited by OAM of light (bottom).

results of the relative change in absorption efficiency under several OAM conditions (Details for obtaining the relative absorption efficiency are described in Method and Supporting Information Section 9). The result demonstrates that the incident OAM light with $l = 2$ enhances the absorption in MoS₂, with an augmentation of around 1% compared to the light without OAM ($l = 0$). One possible reason for this enhancement, as suggested by some studies, is that the interaction of OAM light with materials such as GaAs,³⁷ trapped ions,³⁸ metamaterials,³⁹ and semiconductor cuprite⁴⁰ can induce specific selection rules in dipole transitions, and

even promote quadrupole or hexapole transitions. OAM light excitation may induce additional transitions that cannot be triggered by fundamental Gaussian light. These additional transitions could be one of the possible reasons for the increased absorption we observed. In addition, if the parameters (λ , h , c) are held, and the laser power (P_{opt}) and power density ($\frac{P_{\text{opt}}}{A_{\text{ph}}}$) are further fixed, respectively. The results

also reveal increased photoconductance in both cases (Figure 2d and Figure S6d). Considering that the monolayer structure of MoS₂ tends to favor exciton formation over free carriers, the subsequent process of exciton dissociation into free carriers must be taken into account. Therefore, eq 5) does not completely describe the increased Δn , which is influenced by exciton dissociation. The photocurrent can be related to the rate at which exciton-dissociated free carriers drift out and are extracted by electrodes (free carrier extraction efficiency, η_{extract}), following the relation $I_{\text{ph}} \propto \Delta n \propto \eta_{\text{extract}}$. Accordingly, the extraction efficiency (η_{extract}) is proportional to the exciton dissociation efficiency (η_{diss}), and is also linked to the exciton recombination lifetime (τ_X),²⁸

$$\eta_{\text{extract}} \propto \eta_{\text{diss}} \propto \frac{\tau_X}{\tau_X + \tau_{\text{diss}}} \quad (6)$$

The τ_{diss} represents the exciton dissociation lifetime. Since the exciton dissociation rate ($\frac{1}{\tau_{\text{diss}}}$) is influenced by the applied in-plane field (bias voltage, V_{DS}), in our analysis, we fixed the range of V_{DS} within ± 0.2 V to retrieve the photoconductance. Under these conditions, the exciton dissociation rate is assumed to remain no significant change. Thus, the primary focus should be on determining the influence of OAM-carrying light on exciton recombination lifetime. Figure 4b provides the results of TRPL measurements conducted at room temperature, which turns out the exciton recombination lifetime by the combination of exciton radiative and nonradiative

recombination rates,⁴¹ $\tau_X = \left(\frac{1}{\tau_{\text{nr}}} + \frac{1}{\tau_{\text{nr}}} \right)^{-1}$. The PL emission spectrum analysis returns an extended exciton lifetime and narrower full width at half-maximum (fwhm) as presented in the inset (The raw PL results are provided in Supporting Information Figure S10). The prolonged lifetime for $l = 2$ is around 50% enhanced (from ~ 550 ps to ~ 800 ps) concerning the case of $l = 0$, as depicted in the inset of Figure 4b. According to the PLQY described in eq 1), these findings demonstrate that the prolonged exciton lifetime corresponds to a decrease in the radiative recombination rate as the OAM order increases. This suggests an enhancement in exciton dissociation efficiency, facilitating the conversion of exciton-bound states into free carriers, thereby improving the photocurrent.

Apart from observing the prolonged exciton recombination lifetime, we found a blue shift in the PL spectra that demonstrates a change in exciton recombination energy. The shifting is around 4 meV per OAM carried by light, as evident in Figure 2b and analyzed in Supporting Information Figure S11. The blue shift from the increment of OAM provides a fact that the exciton was excited to a higher energy state. To quantify the relative binding energy shift (ΔE_b), the analyzed A-exciton peak position is assumed to be correlated with its binding energy. The corresponding exciton binding energy excited by various OAM is compared with that induced by the Gaussian beam, allowing the assessment of $\Delta E_b = E_b^l - E_b^{l=0}$.

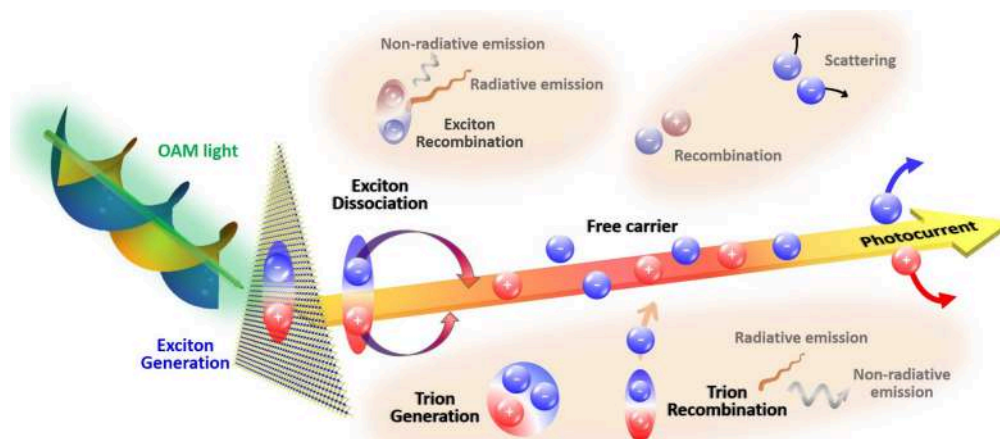


Figure 5. Mechanism of photocurrent enhancement by OAM of light. The illustration of the mechanism that OAM of light contributes to the enhancement of the conductance. In this, the rising OAM of light improves the exciton dissociation efficiency that generates more free carriers, causing the photocurrent to gradually increase as manifested in the device's conductance.

The ΔE_b decreases with the increasing order of OAM as illustrated in Figure 4c, indicating a weaker coupling between electron and hole induced by OAM of light.

We infer that the OAM of light generates excitons with CoM momentum containing OAM due to the conservation of angular momentum, as illustrated in Figure 4d. The representative schematics in the top row are based on exciton band structures obtained from the DFT-based Bethe-Salpeter equation (BSE) calculations for MoS_2 monolayers,²² in which, exciton quasi-particles are excited to energy states through the OAM light. The recombination pathways for OAM-carrying excitons are predominantly radiative but with a lower probability of optical emission due to the prolonged radiative recombination lifetime. Therefore, a higher order of OAM is employed to excite an exciton that results in a diminished optical emission rate but with slightly higher energy. Furthermore, Figure 4d further illustrates the potential mechanism behind the enhanced exciton dissociation, accompanied by an expanded distance (radius) between the electron and hole (bottom panel). The applied in-plane field distorts the band structure into a slide-like shape, increasing the likelihood of electron tunneling into the conduction band, representing exciton dissociation and subsequently forming free carriers.²⁸ The expanded exciton radius, excited by OAM-carrying light, increases the probability of excitons dissociating into free carriers, allowing both electrons and holes to tunnel through the potential barrier more effectively, which enhances the photocurrent. Figure 5 illustrates the potential evolution process for these excitons converting into free carriers and raising the photocurrent. For exciton recombination, the diminished screening effect fosters a strong Coulomb interaction between electron–hole pairs, resulting in a robust attractive force and the formation of highly efficient excitons with radiative recombination. However, still, a fraction of these excitons undergo nonradiative recombination due to exciton–phonon interactions. For trion formation, it involves the introduction of excess carriers, which may manifest as either electrons or holes. For example, in the case of a negatively charged trion, an extra electron is captured by the exciton, establishing a bound state comprised of two electrons and one hole. The recombination process for a trion is distinct from that of the exciton, resulting in the release of the excess electron and the recombination of the remaining electron–

hole pair.⁴² The released electron becomes a free carrier, probably collected by the electrodes under applied bias voltages. In the above processes, the electron–hole pairs ultimately undergo annihilation. However, the bound exciton quasi-particle can undergo dissociation, releasing the bound state and transforming both the electron and hole into free carriers. The OAM of light may enhance the generation rate of excitons by increasing absorption. Additionally, it endows excitons with additional momentum, thereby extending their lifetime and increasing the radius of excitons. This heightened momentum augments the likelihood of exciton dissociation, consequently elevating the free carrier generation rate and contributing to an increase in photocurrent. The application of bias voltage unveils this enhancement in device's conductance. Hence, the OAM of light simultaneously inhibits radiative exciton recombination and amplifies free carrier generation, ultimately enhancing the photoconductance in monolayer MoS_2 -based optoelectronic devices. As TMD materials scale down from bulk to few-layer and eventually to monolayer, the weakening of the screening effect results in an increase in exciton binding energy. This makes the dissociation of excitons into free carriers a critical consideration for optoelectronic applications. The enhancement of exciton dissociation and free carrier generation induced by OAM light in other two-dimensional materials holds great potential for practical applications. Moreover, studies have suggested that OAM light can influence free carrier drift velocity,^{43,44} implying a potential impact on free carrier lifetime. Free carrier lifetime emerges as a crucial parameter for understanding how OAM affects carrier transport, recombination, and mobility. Future research should integrate ultrafast optical and electrical measurements to provide deeper insights into OAM-induced charge transport effects. Since the interplay between the OAM of light and 2D/other materials still hides many undiscovered physical mechanisms, making it an intriguing area for future research.

CONCLUSIONS

The integration of OAM into the optical domain introduces an additional facet of control with implications for optoelectronic device applications. In the course of our investigation, we observed a reduction in the PL exhibited by the MoS_2 FET device as the OAM carried by the incident light increased. We

infer that the conservation of OAM imparts an augmented momentum to the generated excitons, which equivalently prolongs their radiative recombination lifetime. This phenomenon is intrinsically tied to the transformative facet that augments the probability of exciton dissociation and the creation of free charge carriers. Consequently, the infusion of OAM into light triggers an elevation in the efficiency of exciton dissociation, fueling the photoconductance within MoS₂-based optoelectronic devices. The OAM-driven MoS₂ FET exhibits versatility, making it a candidate for a wide range of future device applications.

METHODS

Synthesis of MoS₂. MoS₂ is synthesized on a sapphire substrate using the chemical vapor deposition (CVD) method at a temperature of 190 °C. In the synthesis process, molybdenum trioxide (MoO₃) powder is positioned in a quartz boat at the center of the heating zone in the furnace. Alongside, a sapphire substrate is placed adjacent to the quartz boat. Another quartz boat, containing sulfur (S) powder, is positioned at the upstream side of the furnace. The flow of argon gas is utilized to transport the vapors of both sulfur and MoO₃ toward the sapphire substrate for the synthesis process.

Device Fabrication. The fabrication process involves applying a coating of PMMA to both the MoS₂ and h-BN films, with a spin speed of 4000 rpm for 55 s. The PMMA/MoS₂/sapphire substrate undergoes immersion in an ammonia solution for 1 to 2 h to facilitate the separation of the PMMA/MoS₂ film from the sapphire substrate. Simultaneously, the h-BN/copper film is soaked in an ammonium persulfate solution to decompose the copper and enable the separation of the PMMA/h-BN film. Following the successive transfer of h-BN and MoS₂ onto the SiO₂/Si substrate, the protective PMMA layer is removed using acetone. A new layer of PMMA, with the same parameters, is applied for the e-beam lithography process to define the etching patterns. This is followed by argon plasma treatment to etch the unprotected patterns and remove the PMMA. Subsequently, the sample undergoes wet cleaning in an *N*-methyl-2-pyrrolidone solution to eliminate residues or contaminants on the MoS₂ surface. Another layer of PMMA is coated on the MoS₂/h-BN/SiO₂/Si substrate with a spin speed of 4000 rpm for 55 s to prepare it for the e-beam lithography process. Finally, Au (50 nm)/Ti (20 nm) electrodes are deposited using a thermal evaporator.

Laser Power Calibration Across Orders of OAM. To calibrate the OAM light power across different OAM orders, we employ two power meters placed at different positions: one at the sample side (Position 1) and another along the optical path between the sample and the SLM (Position 2). To ensure a consistent incident power on the device, we fix the power at Position 1 and record the corresponding power at Position 2. This method allows us to determine the necessary external power adjustments to maintain a constant power level at the sample side for each OAM order during the experiment. The calibrated laser power at Position 2 remains approximately 530 ± 3 μW across different OAM orders.

Reflectivity Measurement of the Excitation Source. The relative absorption efficiency was determined through a reflectivity experiment. In this experiment, a circularly polarized 532 nm excitation source passed through vortex phase plates (VPP) to impart OAM to the light. Subsequently, the light was directed toward a quarter-wave plate to be transformed into linear polarization before reaching the target. The reflected laser power was recorded after the excitation source was impinging on two different heterostructures: MoS₂/h-BN/SiO₂/Si and h-BN/SiO₂/Si, respectively.

TRPL Measurement. We conducted an investigation into the TRPL of the A-exciton in monolayer MoS₂. This involved using a pulse laser with a wavelength of 516 nm, a pulse width of less than 160 ps, and an average power of 4 mW, operating at a frequency of 80 MHz. To introduce light with OAM, we utilized a quarter-wave plate (QWP) in conjunction with a VPP, allowing us to control the phase of the transmitted circularly polarized incident laser beam. The

polarization was then transformed back to linear by passing through another QWP before impinging onto the sample. The spectrometer's slit was positioned at the emitting wavelength of the A-exciton, with a size of approximately 2.8 nm, for the TRPL measurements. The exposure time during measurements was set to 3 min, and the TRPL data was fitted using an exponential function to determine the radiative lifetime.

Statistical Analysis. We conduct all the statistical analyses by the software, OriginLab. The data presents in the figure plots are expressed as mean ± SD, and there is no preprocessing of the data.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsnano.4c18318>.

PL curve, photocurrent, fitted PL results, fixing power density, power dependence of photoconductance, enhancing rate of photoconductance, estimation of relative absorption efficiency, fwhm under TRPL, PL energy shift (PDF)

AUTHOR INFORMATION

Corresponding Authors

Ting-Hua Lu – Department of Physics, National Taiwan Normal University, Taipei 116, Taiwan; Email: thlu@ntnu.edu.tw

Yann-Wen Lan – Department of Physics, National Taiwan Normal University, Taipei 116, Taiwan; orcid.org/0000-0003-2403-1357; Email: ywlan@ntnu.edu.tw

Authors

Ye-Ru Chen – Department of Physics, National Taiwan Normal University, Taipei 116, Taiwan

Kristan Bryan Simbulan – Department of Mathematics and Physics, University of Santo Tomas, Manila 1008, Philippines; orcid.org/0000-0001-9102-5876

Guan-Hao Peng – Department of Electrophysics, National Yang Ming Chiao Tung University, Hsinchu 300, Taiwan

Yu-Chen Chang – Department of Physics, National Taiwan Normal University, Taipei 116, Taiwan

I-Tong Chen – Department of Physics, National Taiwan Normal University, Taipei 116, Taiwan

Han-Chieh Lo – Department of Physics, National Taiwan Normal University, Taipei 116, Taiwan

Shao-Yu Chen – Center of Atomic Initiative for New Materials, National Taiwan University, Taipei 116, Taiwan; orcid.org/0000-0003-3423-9768

Shun-Jen Cheng – Department of Electrophysics, National Yang Ming Chiao Tung University, Hsinchu 300, Taiwan; orcid.org/0000-0002-1721-7914

Complete contact information is available at: <https://pubs.acs.org/doi/10.1021/acsnano.4c18318>

Author Contributions

In this work, Y.-R.C. performed the experiments and data analysis. The device fabrication and preliminary test are conducted by H.-C.L. The optical setup for the experiments is prepared by Y.-C.C. and T.-H.L. The TRPL experiments conducted in NTNU is executed by I.-T.C. The reflectivity experiments that conducted in NTU is supported by S.-Y.C. The exciton radiative decay rate is suggested and calculated by G.-H.P. and S.-J.C. The manuscript is prepared and reviewed

by Y.-R.C., K.B.S., G.-H.P., S.-J.C., T.-H.L., and Y.-W.L. This project is conceived and led by T.-H.L. and Y.-W.L.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

We thank Dr. Wen-Hao Chang at NTNU for his suggestions and help with electrical experiments. We thank Yu-Fan Chiang at NTNU for his support of optical setups. We express our gratitude to the members of TLS 09A1 at the National Synchrotron Radiation Research Center's Taiwan Light Source for their support and assistance with prior test measurements. This work was also partially supported by the Taiwan Semiconductor Research Institute for the device fabrication, and the Semiconductor Fabrication Lab of the Consortia of Key Technologies, and the Nano-Electro-Mechanical-System Research Center, National Taiwan University. This work is supported by the National Science and Technology Council, Taiwan, under contract nos. MOST 108-2112-M-003-010-MY3, MOST 111-2628-M-003-002-MY3, MOST 111-2124-M-003-005-, NSTC 112-2124-M-003-002-, and NSTC 112-2112-M-003-014.

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