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Valley-Dependent Photogating-Controlled Quantum Transport in Monolayer Transition Metal Dichalcogenides

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Abstract:

In this work, we develop a theoretical framework to investigate trap-charge-induced photogating as the sole electrostatic control mechanism for quantum transport and different basis for negative differential resistance (NDR) in monolayer WSe₂ and MoS₂. The valley sensitive optical response, photocarrier generation, carrier trapping, and resulting photogating potential are coupled to quantum transport using the Landauer–Büttiker formalism and an extended transfer-matrix approach. The transport response is systematically examined over photogating voltages (ΔV_{pg}) of 0 to 8 mV. The calculations reveal a strongly nonlinear current response, with photogating-induced currents of the order of 0.2 μ A and maximum peak to valley ratio, mPVR, values of approximately 1.01–1.03. For ML – WSe₂, the calculated absorption coefficient reaches $17.82 \times 10^5 \text{ cm}^{-1}$, with a trapped-carrier density of approximately $1.3 \times 10^{12} \text{ cm}^{-2}$. The observed NDR arises from photogating-induced modulation of resonant transmission states in the absence of an externally imposed source–drain driving potential. Our findings establish photogating as a direct means of controlling nonlinear quantum transport and provide a basis for light-controlled low-power switching, signal modulation, sensing, memory, and neuromorphic applications.

Key word: photogating, trapped carriers, transmission, monolayer TMDCs, negative differential resistance (NDR)

Introduction:

Two-dimensional (2D) semiconductors have attracted considerable attention because of their atomically thin geometry, reduced dielectric screening, and strong sensitivity to external stimuli providing exceptional opportunities for controlling electronic and optoelectronic properties at the nanoscale. Among them, monolayer transition-metal dichalcogenides (ML-TMDCs), including MoS₂, MoSe₂, WS₂, and WSe₂, are particularly attractive owing to their direct band gaps spanning the visible to near-infrared range, strong light–matter interactions, pronounced spin–orbit coupling, and valley-dependent degrees of freedom. Unlike graphene, ML-TMDCs possess substantial direct band gaps that enable effective control of charge

transport and high on/off current ratios, making them promising for field-effect transistors, diodes, photodetectors, and other electronic and optoelectronic devices [1–4]. Their strong Coulomb interactions also give rise to tightly bound excitons that strongly influence their optical response [5–11]. Moreover, their electronic and optical properties can be engineered through dielectric-environment modulation, external electric fields, and defect engineering, providing opportunities for low-power electronics, optoelectronics, and emerging neuromorphic architectures [12–21].

Photogating provides an important different mechanism for converting optical excitation into an electrical response in low-dimensional semiconductors. In this process, photogenerated carriers become trapped at defects, impurities, or interfaces, generating an effective electrostatic potential that modulates the energy landscape and charge transport. Unlike conventional photocurrent generation based primarily on the collection of photocarriers, photogating allows trapped charge to act as an effective or virtual gate, potentially influencing transport even after the initially generated free carriers have recombined or been extracted. The high surface-to-volume ratio and weak dielectric screening of 2D materials make them particularly sensitive to such trap states, whose density and lifetime strongly influence the resulting photogating potential. Trap-mediated photogating has consequently attracted interest for high-gain photodetection, optical memory, self-driven photodetectors, and optoelectronic neuromorphic applications [19–23].

Experimental studies demonstrate that photogating can produce substantial electrical modulation in TMDC-based structures. In multilayer $\text{MoS}_2/\text{HfO}_2$, for example, oxygen vacancies in HfO_2 can trap photogenerated holes, producing a photocurrent of approximately $2.1 \mu\text{A}$, compared with 62.3 nA for $\text{MoS}_2/\text{SiO}_2$ under comparable conditions [24]. Sahoo et al. reported a responsivity of approximately $4.2 \times 10^3 \text{ A/W}$ by controlling carrier trapping in ML – MoS_2 [25], while photogating-induced currents of approximately $0.17 - 1.7 \mu\text{A}$ have been reported for MoS_2 -based phototransistors [26]. For WSe_2/WO_x , a responsivity exceeding 3000 A/W at an optical power of 1.1 nW , corresponding to a photocurrent greater than approximately $3.3 \mu\text{A}$, was demonstrated [27]. In addition, photogating associated with in-gap trap states has been identified as a dominant contribution to the photocurrent in MoSe_2 , while Han et al. studied trapped-charge distributions, electrostatics, and quantum transport in a self-consistent numerical treatment [28]. Despite these advances, photogating has generally been studied as a modulation mechanism for a current established by an externally biased device, with the drain–source voltage retained as a transport-driving parameter. In contrast, the our work considers the photogating potential generated by trapped photocarriers as the sole electrostatic driving and control potential, providing a basis for investigating non-monotonic quantum transport and NDR without an externally imposed source–drain driving potential.

Negative differential resistance (NDR) is an important nonlinear transport phenomenon with potential applications in switching, memory, logic, and high-frequency devices. In 2D materials, NDR can arise from band-to-band tunneling, resonant tunneling, or alignment of discrete quantum states. Experimental and theoretical studies have reported NDR in $\text{MoS}_2/\text{WSe}_2$ heterostructures [29], $\text{WSe}_2/\text{h} - \text{BN}/\text{p}^+ - \text{MoS}_2$ structures [30], and $\text{MoS}_2 -$

WSe₂ – Graphene heterostructures [31]. Reported peak to valley ratio(PVR) values include 1.8 at room temperature and 3.8 at 7 K for Graphene/h – BN/Graphene [32], 1.06–4.2 for theoretical Graphene/h – BN/Graphene structures [33], approximately 2.6 for graphene/black – phosphorus heterostructures at biases above 1.5 V [34], and 1.42 and 1.43 for WTe₂/ZrS₂ and WTe₂/HfS₂, respectively [35]. For ML – WSe₂/MoS₂ planar superlattice diodes, low-bias resonant-tunneling NDR with mPVR values of 1.30–1.61 has also been predicted [36]. Importantly, a large PVR is not the only criterion for technological relevance: NDR with a very small peak-to-valley voltage difference can be advantageous for high-frequency and low-power operation [37]. Light-triggerable NDR has further demonstrated the possibility of coupling optical excitation to nonlinear transport and switching [38]. These results motivate a different question: can trapped photoinduced charge, acting as an internally generated electrostatic potential, control quantum transmission and produce NDR without an externally applied source–drain driving potential? To the best of our knowledge, this specific mechanism has not been systematically investigated in ML-TMDCs. As mentioned before in the present research, the photogating potential (V_{pg}) is therefore treated as the principal control parameter, and NDR corresponds to a regime in which ($dI/V_{pg} < 0$).

Accordingly, this study develops a theoretical framework for trap-charge-induced photogating and its effect on quantum transport in ML-TMDCs, with particular emphasis on WSe₂ and MoS₂. The optical absorption, photocarrier generation, trapped-carrier density, and resulting photogating potential are determined within the adopted theoretical framework and subsequently incorporated into quantum transport through the Landauer–Büttiker formalism and the extended transfer-matrix approach. No externally applied source–drain voltage is used as the origin of the calculated current; instead, trapped photocarriers and their associated photogating potential provide the sole electrostatic mechanism for driving and modulating transport. The resulting current and NDR are therefore investigated as functions of V_{pg} , with the photogating potential considered over the range $\Delta V_{pg} = 0 - 8$ mV. The complete analytical derivations of the ML-TMDC optical absorption coefficient and the transmission coefficient under photogating are provided in Supplementary Sections 1 and 2, respectively. Overall, the study establishes a light-controlled route to nonlinear quantum transport in which trapped charge acts as an internally generated electrostatic control potential, with potential relevance to low-power optical switching, optoelectronic memory, sensing, signal modulation, and optoelectronic neuromorphic architectures.

Theoretical Model and Method:

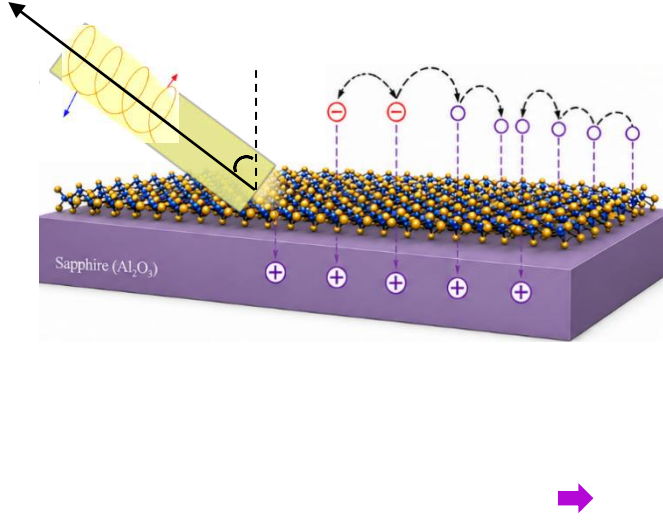


Figure 1. Schematic illustration of the photogating effect in ML-TMDCs under circularly polarized illumination.

Photoexcitation generates electron–hole pairs, which are subsequently separated and trapped, leading to a modulation of the effective potential (ΔV_{PG}) and, consequently, the transmission.

In this study, we aim to develop a physical model based on ML-TMDCs, in which defects and impurities act as sources of trap states. Accordingly, we estimate the photogating potential in terms of the structural parameters of the system. The underlying mechanism is schematically illustrated in Fig. 1. We first consider the photogating potential induced by trapped charge carriers. Specifically, trapped carriers generate a net charge within the system, which acts as an additional electrostatic gate and induces a local electric field. This field modifies the free-carrier density and, consequently, the electrical conductivity of the system. Upon illumination, electron–hole pairs are generated, a fraction of which becomes trapped in the available trap states. The density of these trapped carriers, n_t , can be approximated as:

$$n_t = \eta' G \tau_t, \quad (1)$$

Here, G denotes the generation rate of electron–hole pairs induced by optical illumination, η' represents the fraction of photogenerated electron–hole pairs that become trapped, and τ_t is the average carrier lifetime in the trap states. Thus, under steady-state conditions, the trapped-carrier density can be approximately described by Eq. (1), which provides a commonly used first-order approximation. Since n_t is defined as the trapped-carrier density per unit volume, an important consideration for two-dimensional systems is that trap densities are commonly

reported as areal densities. Therefore, the corresponding areal trapped-carrier density, $n_t^{(2D)}$, can be obtained from the volumetric density n_t using the effective thickness of the monolayer, t_{mono} , as $n_t^{(2D)} = n_t \times t_{\text{mono}}$. A key step in obtaining a quantitative estimate of the trapped-charge density is therefore to determine the electron–hole pair generation rate, G . The generation rate depends on the incident optical intensity, the absorption coefficient, and the quantum efficiency. G can be written as [39]:

$$G = \left(\frac{I}{\hbar\omega}\right) \cdot \alpha \cdot \eta, \quad (2)$$

In photogating and photoluminescence experiments, the incident optical intensity is typically on the order of a few W/m^2 , whereas in laser-based experiments, it can reach values of several hundred W/m^2 .

Here, I denotes the incident optical intensity. More precisely, since optical intensity has the dimensions of energy per unit area per unit time, rather than the number of photons, I should be divided by the energy of each photon, $(\hbar\omega)$, to obtain the incident photon flux. Thus, $(I/\hbar\omega)$ represents the photon flux of the incident radiation. In photogating and photoluminescence experiments, the incident optical intensity is typically on the order of a few W/m^2 , whereas in laser-based experiments, it can reach values of several hundred W/m^2 . Here, α denotes the absorption coefficient of the ML-TMDCs, which depends on the wavelength of the incident light and the intrinsic properties of the material. Owing to their strong excitonic optical transitions, ML-TMDCs exhibit remarkably high optical absorption. As detailed in Supplementary Section (1), the absorption coefficient of these materials is derived fully analytically from the Hamiltonian governing their electronic states, explicitly accounting for the contributions of different valleys and spin states. Based on Eq. (A9), the absorption coefficient of the ML-TMDCs is obtained in the following form:

$$\alpha_{TMDC} = \left(\frac{e^2 v_F^2 (m_r)^{3/2}}{\sqrt{2} c_0 \epsilon_0 n \hbar^2}\right) |F(x)|^2 \left[\left(\frac{\sqrt{\hbar\omega - E_g}}{\hbar\omega}\right)\right] \Theta(\hbar\omega - E_g) \quad (3)$$

Once the absorption coefficient of the structure has been determined, the only remaining unknown parameter in Eq. (2) is the quantum yield of the layer, denoted by η . The values of η for different ML-TMDCs, together with the other relevant parameters, are reported in Table 1. The values highlighted in red correspond to the parameters adopted in the present study [40,41].

Table 1: Electronic properties and structural parameters of ML-TMDCs

Material	Layer thickness (d (nm))	Mobility (μ ($\text{cm}^2/\text{V}\cdot\text{s}$))	Efficiency (η)	Electron effective mass (m_e^*/m_0)	Hole effective mass (m_h^*/m_0)	Refractive Index (n , ~ 650 nm)
ML – MoS ₂	0.65	0.7	1	0.35–0.47(0.41)	0.43–0.54(0.485)	4.0–4.5(4)
ML – MoSe ₂	0.60	0.5	0.97	0.55–0.60	0.65–0.70	4.8–5.1
ML – WS ₂	1.00	0.9	1	0.27–0.33	0.36–0.40	4.2–4.4
ML – WSe ₂	0.73	0.9	1	0.28–0.32(0.30)	0.34–0.38(0.36)	4.5–4.8(4.5)

To calculate the trapped-carrier density using Eq. (2), an estimate of the trapped-carrier lifetime, (τ_t), is required. The parameter τ_t is therefore one of the key parameters governing the dynamics of photogating and the photoresponse of ML-TMDCs. It depends on several characteristics of the trapping centers, including the trap energy depth, trap density, and defect type, as well as the surface area of the structure, system temperature, and carrier transport and recombination dynamics. Under a first-order rate-equation approximation, the trapped-carrier lifetime can be estimated from $\tau_t = 1/\gamma$, where γ is the recombination rate coefficient. The value of this parameter is typically determined from time-resolved photocurrent measurements or by fitting appropriate dynamical models to the experimental data. Importantly, γ , and consequently τ_t , do not possess a single universal value across different ML-TMDCs, as these quantities depend strongly on the material, defect states, interfaces, and experimental conditions. Experimental studies of ML-TMDCs have shown that carrier recombination can involve both fast components on the timescale of a few picoseconds and slower components extending beyond hundreds of picoseconds, highlighting the important role of defect-mediated trapping and recombination processes, particularly Auger-related processes [42]. In photogating phenomena, however, carriers trapped in deep defect states or interfacial states can exhibit substantially longer lifetimes, resulting in persistent or slow photoresponses. In some ML-TMDC-based devices, response times ranging from milliseconds to seconds have been reported, with trapped-carrier lifetimes extending beyond 10^4 s [43]. Therefore, for an accurate description of photogating, the trapped-carrier lifetime should ideally be determined experimentally based on the nature of the trapping centers and the specific TMDC/substrate structure. The resulting trapped-charge density, in turn, gives rise to a local electric field that can be modeled as an effective gate potential, V_{pg} , given by $V_{pg} = en_t^{(2D)}d/C$, where e is the elementary charge, d is the effective thickness of the material, and C is the effective capacitance between the trapped charges and the semiconductor. This effective potential acts as an equivalent gate voltage and thereby modifies the electrical conductivity of the material.

An important aspect of the present work is to investigate the modulation of the charge transport by trapped carriers through the photogating potential. Once the photogating potential has been determined, its effect on the transport current can be evaluated using an appropriate quantum transport formalism. One of the most widely used theoretical approaches for calculating the electrical conductance and current through a mesoscopic or nanoscale structure in the presence of a potential is the Landauer–Büttiker formalism [19,36,44]. For ML-TMDCs under a photogating potential (V_{pg}), the current can be expressed within the Landauer–Büttiker formalism as:

$$I_{SD} = I_0 \int_{-\mu_S}^{+\mu_D} dE |E| T_{\tau}^{s_z}(E, V_{pg}), \quad (5)$$

where I_0 is the constant prefactor given by $I_0 = 2geW/v_F h^2$, with $g = 4$ accounting for the spin and valley degeneracies, W denoting the width of the device, and v_F representing the Fermi velocity. Here, $T_{\tau}^{s_z}(E, V_{pg})$ is the energy- and photogating-dependent transmission probability for carriers in the specified valley τ and spin state s_z . The photogating potential does not

directly contribute to the transport current through the trapped carriers themselves. Rather, the trapped charge generates an electrostatic potential that modifies the electronic energy landscape of the ML-TMDC structure. In particular, V_{pg} shifts the relevant energy levels relative to the chemical potential and consequently modifies the energy range and transmission probability of the free charge carriers. As a result, the photogating effect can significantly alter the carrier transmission and, hence, the electrical current. The magnitude of this photoinduced modulation depends on the trapped-carrier density and, ultimately, on the incident optical intensity. The resulting expression for the current can then be evaluated numerically through appropriate approximations and numerical integration. Before calculating the current, however, an estimate of the carrier transmission probability through the structure under the influence of V_{pg} is required. Accordingly, the transmission coefficient of the structure is derived in Supplementary Section (2). Based on Eq. (B20), the transmission coefficient is obtained in the following form:

$$t(E, V_{pg}) = \frac{2P_0 \cos \theta_0}{M_{22}P_0 e^{\mp i\theta_0} + M_{11}P_e e^{\pm i\theta_e} - M_{12}P_0 P_e e^{\pm i(\theta_e - \theta_0)} - M_{21}} \quad (5)$$

Thus, the transmission probability can be defined as $T_t^{sz}(E, V_{pg}) = |t(E, V_{pg})|^2$.

1. Results and discussions.

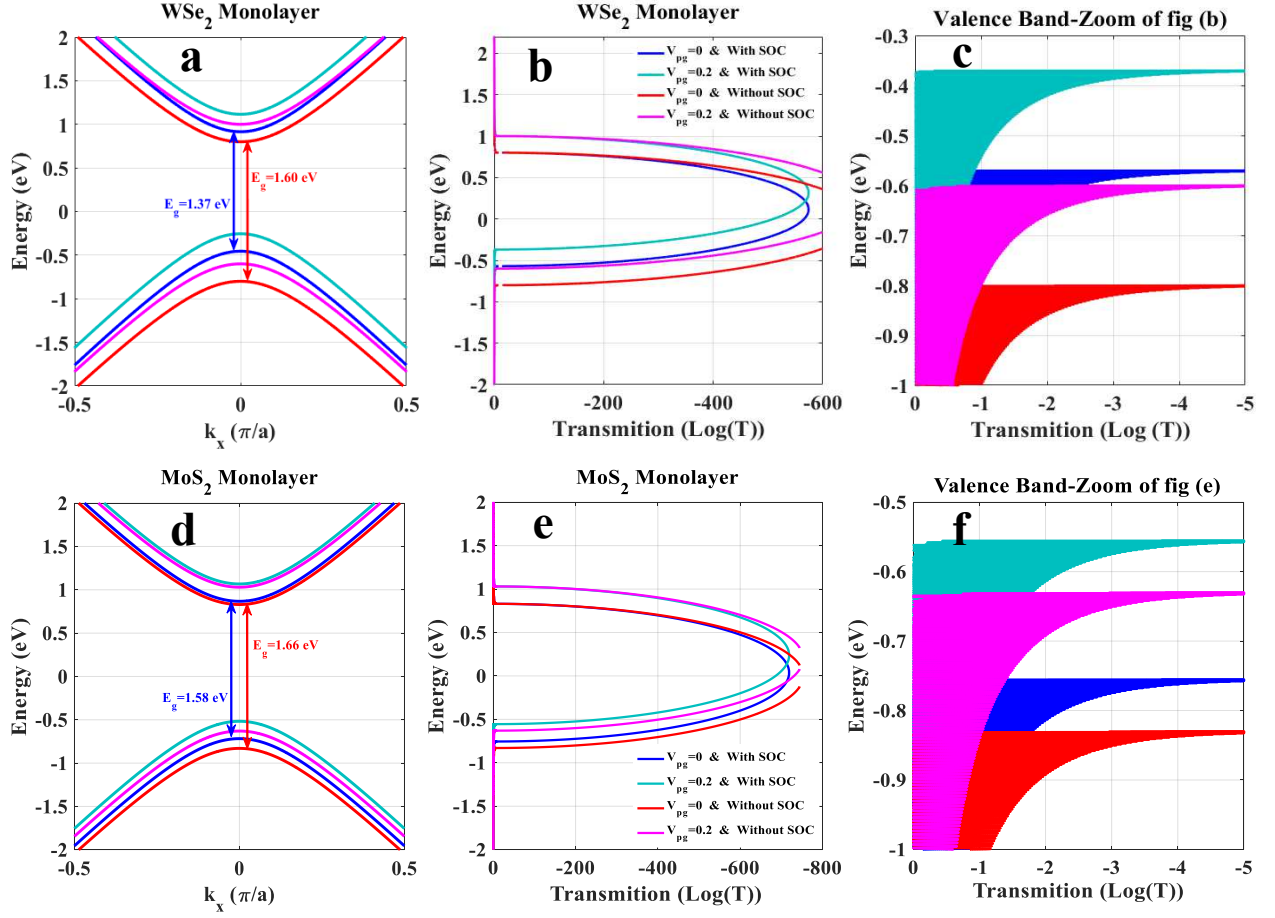


Figure 2. Band structures and corresponding transmission spectra of ML – WSe₂ and ML – MoS₂ with and without spin–orbit coupling (SOC) and under photogating. (a,d) Band structures showing the SOC-induced valence-band splitting and band-gap modification. (b,e) Transmission spectra under different SOC and photogating conditions, illustrating the photogating-induced shift of the transmission edges. (c,f) Enlarged views of the valence-band regions and corresponding transmission spectra.

One of the most important effects in ML-TMDCs is spin–orbit coupling (SOC), which gives rise to pronounced modifications in their electronic and optoelectronic properties compared with conventional semiconductors. Figure 2(a) shows the band structure of ML – WSe₂ in the presence and absence of spin–orbit interaction. It can be seen that the inclusion of SOC leads to a pronounced splitting of the valence band at $k_x = 0$, whereas the conduction band undergoes only minor changes. This behavior originates from the orbital character of the electronic bands. The valence-band maximum is predominantly composed of the $d_{x^2-y^2}$ and d_{xy} orbitals of tungsten, together with the p orbitals of selenium, and therefore experiences strong spin–orbit coupling owing to the relatively large atomic number of tungsten. In contrast, the conduction-band minimum is predominantly derived from the d_{z^2} orbital, which is less sensitive to spin–orbit interaction; consequently, only relatively small changes are observed in the conduction band. The results show that the valence-band splitting is approximately

230 meV, which is in good agreement with previously reported theoretical and experimental values for ML-WSe₂. This spin-orbit-induced splitting reduces the band gap from approximately $E_g = 1.60$ eV to $E_g = 1.37$ eV. Although this reduction in the band gap is relatively modest, it can have a significant impact on the distribution of electronic states near the Fermi level and, consequently, on the quantum-transport properties of the system. From a physical perspective, in ML-TMDCs, the combination of broken inversion symmetry and strong spin-orbit coupling, arising from the presence of heavy elements, gives rise to spin-valley locking, such that the K and K' valleys are associated with opposite spin polarizations. This intrinsic spin-valley coupling provides an important basis for a wide range of spintronic and valleytronic applications of ML-TMDCs. Figure 2(b) presents the corresponding transmission spectrum calculated from the electronic band structure showing that, the transmission gap coincides with the band gap of the electronic structure, and the transmission probability remains nearly zero within this energy range. Upon inclusion of spin-orbit interaction, the transmission edges associated with the spin-orbit-split valence bands become separated, resulting in two distinct energy thresholds for the onset of transmission. This behavior arises because the transmission function is directly related to the number of available electronic modes that can contribute to transport. When SOC lifts the degeneracy of the valence-band states, the transmission channels associated with the two spin states become available at different energies. Consequently, the onset of transmission occurs at two distinct energy thresholds. This behavior demonstrates the dependence of quantum transport on the spin polarization of the charge carriers. Figure 2(c) shows an enlarged view of the valence-band region and the corresponding transmission spectrum. Figure clearly demonstrates that the energy splitting induced by SOC is directly reflected in the transmission spectrum. In other words, each spin-orbit-split branch of the band structure corresponds to an independent transmission channel in the transmission spectrum. These results demonstrate that the transmission spectrum provides a direct manifestation of the electronic structure of the material, with the modifications induced by spin-orbit coupling being directly reflected in its quantum-transport characteristics.

Upon switching photogating potential on ($V_{PG} = 0.2$ V), a noticeable shift in the energy bands and, consequently, in the transmission spectrum is observed. As shown in Figs. 2(b) and 2(c), photogating primarily induces an energy shift of the bands, thereby moving the onset energies of the transmission channels to new values. This behavior arises because the photogating potential acts as an effective electrostatic potential that modifies the carrier energies within the active region of the structure, without directly altering the intrinsic crystal symmetry or the underlying spin-orbit coupling mechanism. Consequently, although the positions of the valence- and conduction-band edges are shifted, the energy separation between the spin-split branches remains approximately unchanged. The transmission spectrum follows the same trend, with the transmission channels undergoing a shift along the energy axis, while the overall spectral profile and the number of available transmission channels remain essentially unchanged. These results indicate that photogating primarily serves to tune the energy thresholds for carrier transmission rather than to generate additional transmission channels. In other words, the energy required to activate each transmission channel can be controlled by the photogating potential, whereas the number of conducting modes, determined by the underlying

band structure, remains essentially unchanged. Furthermore, the close view of the valence-band region in Fig. 2(c) clearly shows that, even in the presence of photogating, the two transmission channels associated with the two spin-split branches are preserved. This finding demonstrates that photogating and spin–orbit coupling represent two physically distinct but complementary effects: SOC is responsible for the splitting of the spin states and the formation of distinct transmission channels, whereas photogating primarily tunes the energy positions of these channels. Such independent control of the transmission thresholds provides a means of selectively tuning the carrier-transmission energies without eliminating the underlying spin polarization, which is particularly relevant to the design of tunable spintronic and valleytronic devices. From the application view point, this feature is important because it enables the control of spin-polarized carrier transport through band-structure engineering, without requiring direct modification of the intrinsic spin–orbit coupling.

The results obtained for ML – MoS₂ exhibit a trend similar to that observed in ML – WSe₂, Notable difference that the strength of spin–orbit coupling is considerably weaker. This difference can be attributed primarily to the lower atomic number of molybdenum compared with tungsten. Consequently, the valence-band splitting in ML – MoS₂ is only about 80 meV, corresponding to approximately one-third of the value obtained for ML – WSe₂. As a result, the associated modifications in the transmission spectrum are less pronounced, and the energy separation between the transmission channels originating from the two spin-split branches is reduced. Nevertheless, the direct correspondence between the electronic band structure and the transmission function remains unchanged, such that changes in the electronic structure are directly reflected in the transport characteristics. In ML – MoS₂, the application of a photogating potential produces a behavior similar to that observed in ML – WSe₂. As shown in Figs. 2(e) and 2(f), the transmission edges are shifted to new energy values by turning the photogating potential on, whereas the energy separation between the two transmission channels associated with spin splitting remains approximately constant. Because the spin–orbit-induced splitting is smaller in ML – MoS₂, the energy shift induced by photogating becomes comparable in magnitude to the spin-splitting energy. Consequently, the feature of carrier transmission can be unchanged with bigger sensitivity through the photogating potential. This feature suggests that, in materials with weaker spin–orbit coupling, photogating can play a more pronounced role in tuning the conduction window and controlling the transport characteristics. Therefore, the photogating-induced shift of the transmission thresholds is expected to be directly manifested in the current–voltage characteristics. By varying the photogating potential, the on-state current, threshold voltage, and on/off current ratio of ML-TMDC-based devices can potentially be controlled effectively. This behavior is examined in details below through an analysis of the corresponding current–voltage characteristics.

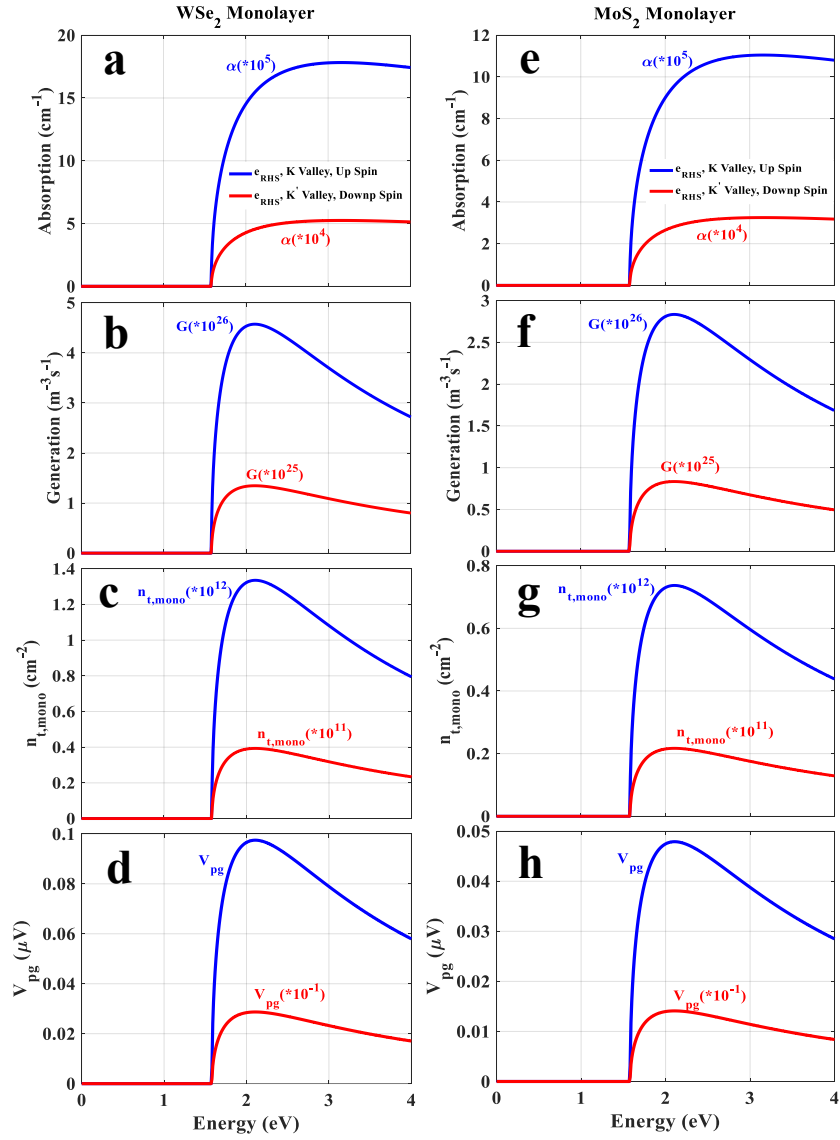


Figure 3. Photon-energy dependence of the optical absorption coefficient, photocarrier generation rate, trapped-carrier density, and photogating potential for ML – WSe₂ (left column) and ML – MoS₂ (right column). The blue and red curves correspond to $\hat{e}_{\text{RHS}}$ excitation coupled to the K valley with spin-up and the K' valley with spin-down configurations, respectively. The four rows represent (a) the absorption coefficient, (b) the photocarrier generation rate, (c) the trapped-carrier density, and (d) the photogating potential.

Figure (3) illustrates the evolution of the optical absorption coefficient, photocarrier generation rate, trapped-carrier density, and, ultimately, the photogating potential as a function of the incident photon energy for ML – WSe₂ and ML – MoS₂. These four quantities represent successive stages of a single physical process, which begins with photon absorption and ultimately leads to the formation of an electrostatic potential equivalent to that induced by an external gate. As discussed above, several factors can influence the magnitude of the photogating potential, among which the optical absorption spectrum of the semiconductor plays a particularly important role. In Supplementary Section (1), we analytically investigated the optical response of ML-TMDCs, with particular emphasis on the absorption coefficient, and derived an explicit expression for the absorption spectrum of these materials within a two-level-system framework. Several parameters can affect both the magnitude and spectral

dependence of the optical absorption, including the spin–valley degree of freedom and the polarization state of the incident light. For ML – WSe₂, as shown in Fig. 3(a), the absorption coefficient for right-handed circularly polarized light ($\hat{\mathbf{e}}_{\text{RHS}}$) coupled to the K and K' valley begins approximately at the bandgap energy, ($E = 1.37$ eV), and increases rapidly with increasing photon energy. This behavior can be understood from the absence of available electronic states for interband transitions when the photon energy is below the bandgap, resulting in negligible optical absorption. Once the photon energy exceeds the bandgap, interband transitions become energetically allowed, and the number of available electronic states increases according to the two-dimensional density of states, leading to a pronounced increase in the absorption coefficient. As evident from the figure, for $\hat{\mathbf{e}}_{\text{RHS}}$ excitation coupled to the K valley, the absorption coefficient reaches a maximum value of $\alpha_{\text{max}} = 17.82 \times 10^5 \text{ cm}^{-1}$ at $E = 3.15$ eV, corresponding to the strongest absorption in the considered spectral range. Notably, this maximum occurs within the visible-light energy range, highlighting the strong optical response of ML – WSe₂ in this region. Another important feature evident in Fig. 3(a) is the pronounced difference between the two absorption curves corresponding to $\hat{\mathbf{e}}_{\text{RHS}}$ excitation coupled to the K and K' valleys. This difference originates directly from the valley-dependent selection rules characteristic of two-dimensional TMDC semiconductors. Owing to the strong spin–orbit coupling and the broken inversion symmetry in monolayer TMDCs, right- and left-handed circularly polarized light couple selectively and predominantly to the K and K' valleys, respectively. In particular, when $\hat{\mathbf{e}}_{\text{RHS}}$ excitation is considered for the K' valley, the maximum absorption coefficient decreases markedly to $\alpha_{\text{max}} = 5.25 \times 10^4 \text{ cm}^{-1}$, corresponding to an approximately 34-fold reduction relative to the K-valley configuration. These results clearly demonstrate that the polarization state of the incident light plays a crucial role in determining the optical absorption and, consequently, the magnitude of the photogating response in ML-TMDCs.

This behavior of the absorption spectrum is directly observed in the photocarrier generation rate, since, according to Eq. (2), the generation rate is directly proportional to the absorption coefficient. Therefore, any increase in the absorption coefficient, in the absence of additional limiting mechanisms, necessarily leads to an increase in the generation rate of electron–hole pairs. In calculating the photocarrier generation rate under optical illumination, an incident light intensity of $I = 500 \text{ W/m}^2$ was assumed. Based on the absorption spectrum, a higher carrier generation rate is expected for $\hat{\mathbf{e}}_{\text{RHS}}$ excitation locked to the K valley than for the K' valley. As shown in Fig. 3(b), the generation rate reaches a maximum value of $G = 4.5 \times 10^{26} \text{ m}^{-3}\text{s}^{-1}$ at $E = 2.15$ eV for the K-valley configuration, whereas the corresponding value for the K' valley is $G = 1.4 \times 10^{25} \text{ m}^{-3}\text{s}^{-1}$, which is approximately 32 times smaller. The shift of the peak in the G spectrum relative to that of the absorption coefficient can be understood from the photon-flux term ($I/\hbar\omega$) appearing in the above relation. At higher photon energies, although the absorption coefficient approaches a nearly saturated regime, the energy carried by each photon increases, thereby reducing the number of incident photons available for a fixed optical intensity. Consequently, the carrier generation rate does not continue to increase with the absorption coefficient but instead reaches a maximum and subsequently decreases. This competition between the increasing absorption coefficient and the decreasing

photon flux at higher photon energies accounts for both the emergence of a maximum in the generation-rate spectrum and its shift toward lower photon energies relative to the peak of the absorption spectrum.

Since only a fraction of the photogenerated carriers are captured by the trap states present in the semiconductor, the trapped-carrier density is expected to closely follow the behavior of the carrier generation rate. Initially, the increase in photocarrier generation leads to the progressive filling of available trap states and, consequently, to a rapid increase in the trapped-charge density. However, beyond the photon energy corresponding to the maximum generation rate, the number of photogenerated carriers decreases, thereby reducing the probability of further trap-state filling. As a result, the trapped-carrier density exhibits a non-monotonic dependence on photon energy and reaches a maximum at a specific photon energy. Assuming that approximately 20% of the photogenerated carriers are captured by trap states ($\eta' = 0.2$) and that the carrier lifetime in the trapped states is $\tau_t = 0.2 \mu s$, Eq. (1) predicts an energy dependence of the trapped-carrier density that closely follows the behavior of G . According to Table (1), the thickness of $ML - WSe_2$ is $d = 0.73 \text{ nm}$. As shown in Fig. 3(c), for $\hat{e}_{RHS}$ excitation coupled to the K valley, the trapped-carrier density reaches a maximum value of $n_{t,mono} = 1.3 \times 10^{12} \text{ cm}^{-2}$, whereas for the K' valley it decreases to $n_{t,mono} = 0.4 \times 10^{11} \text{ cm}^{-2}$. These values are consistent with experimentally reported trap densities in TMDC-based field effect transistors. In particular, trap densities of $2.40 \times 10^{12} \text{ cm}^{-2}$ for WSe_2 and $4.79 \times 10^{11} \text{ cm}^{-2}$ for MoS_2 have been experimentally extracted from gate-voltage hysteresis in TMDC-FETs [45]. Furthermore, trap densities exceeding 10^{12} cm^{-2} have been reported for WSe_2 devices with surface oxide layers [27]. These experimental observations provide reasonable support for the magnitude of the trapped-carrier densities obtained in the present model.

From a physical perspective, as discussed above, the trapped charges act as an internal electrostatic gate. Owing to their relatively long lifetime, these trapped charges can persist in the material even after the photogenerated free carriers are recombined or extracted, thereby establishing a quasi-static electric field that modifies the electronic potential view and induces a shift in the Fermi level. Consequently, the photogating potential is directly proportional to the trapped-charge density, and its energy dependence closely follows the corresponding variation in the trapped-charge density, as presented in Fig. 3(d). Considering the parameters adopted in the present model, together with the energy dependence of the absorption spectrum and trapped-charge density, the calculated photogating potential is found to lie in the microvolt range. For $\hat{e}_{RHS}$ illumination at $E = 2.15 \text{ eV}$ coupled to the K valley, the photogating potential reaches approximately $V_{pg} = 0.1 \times 10^{-6} \text{ V}$, whereas considering coupling to the K' valley it reduces to almost $V_{pg} = 0.2 \times 10^{-8} \text{ V}$. This pronounced valley-dependent variation demonstrates that the control of polarization state of the incident light can provide an effective means of tuning the magnitude of the photogating potential that, consequently paves a way to control the optoelectronic response.

Based on the results presented in Fig. 3 and the foregoing analysis, an important conclusion can be drawn regarding the valley-selective optical response of monolayer TMDCs. Right-handed circularly polarized light couples effectively to the K valley, whereas, conversely, left-handed circularly polarized light couples predominantly to the K' valley. This valley-selective optical coupling constitutes an important selection rule in monolayer TMDCs and provides a direct means of controlling valley-dependent optoelectronic responses through the polarization state of the incident light. Besides, a comparison of the results obtained for ML – WSe₂ and ML – MoS₂ in Fig. 3 further reveals a clear difference in their optical and photogating responses. For ML – MoS₂, the maximum absorption coefficient is obtained at $E = 3.15$ eV, with $\alpha_{\max} = 11.05 \times 10^5 \text{ cm}^{-1}$. Considering the monolayer thickness of $d = 0.65$ nm, the corresponding photogating potential is approximately $V_{\text{pg}} = 0.05 \times 10^{-6} \text{ V}$ for $\hat{e}_{\text{RHS}}$ excitation coupled to the K valley and $V_{\text{pg}} = 0.14 \times 10^{-8} \text{ V}$ for the K' valley. The resulting photogating potential for ML – MoS₂ is therefore approximately one-half of that obtained for ML – WSe₂ under the same conditions. A comparison of the calculated quantities for the two monolayers shows that ML – WSe₂ generally exhibits larger values across the entire sequence of calculations, from optical absorption to photogating potential. This behavior suggests that ML – WSe₂, owing to its stronger light–matter interaction and more efficient photocarrier generation, has a greater capability for generating trapped charges and establishing a photogating field. It may therefore represent a more promising material platform for applications such as photogating-based phototransistors, high-gain photodetectors, and valley-controlled optoelectronic devices. Finally, because the magnitude of the photogating potential depends on a number of structural, optical, and electronic parameters, and considering the range of photogating potentials reported for related TMDC-based structures, we adopt $\Delta V_{\text{pg}} = 0 - 8 \text{ mV}$ as the range of photogating potentials in the subsequent calculations. This range enables us to systematically investigate the effect of photogating over a sufficiently broad parameter space and to assess its influence on the current–voltage characteristics of the proposed structures.

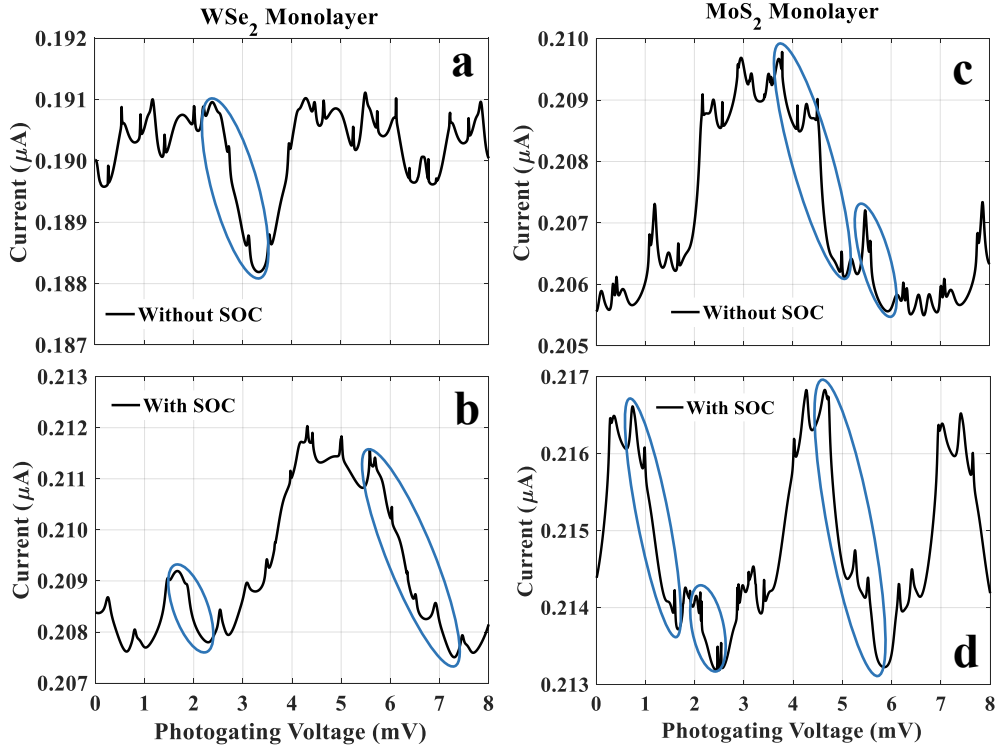


Figure 4. Photogating-induced current as a function of the photogating potential V_{pg} for the two considered ML-TMDC structures, calculated without SOC in (a, c) and with SOC in (b, d). The non-monotonic current response reflects the photogating-induced modulation of carrier transmission.

From the heart of quantum transport physics, results depicted in Fig. 4 represents one of the most important aspects of the present study, as it addresses a relatively unexplored problem: the effect of trap-charge-induced photogating on ballistic transport in ML-TMDCs. Figure (4) shows that the electrical currents of ML – WSe₂ and ML – MoS₂ exhibit a pronounced nonlinear dependent of the photogating potential over the range of $V_{pg} = 0 - 8$ mV. This pronounced nonlinearity indicates that the current is not determined solely by the externally applied bias, but is also directly governed by the modification of the electronic structure caused by photogating. Within the Landauer–Büttiker formalism, the current is expressed by Eq. (4); consequently, any modification of the transmission spectrum can directly lead to changes in the resulting current response. Since the photogating potential in the present work is derived from trapped charges, the observed current modulation primarily reflects changes in the carrier transmission probability arising from the rearrangement of the electrostatic potential landscape, rather than merely an increase in the number of available charge carriers.

In all four curves presented in Fig. 4, the current exhibits a distinctly non-monotonic dependence on the photogating potential, characterized by multiple peaks and valleys. This behavior can be attributed primarily to three factors: the variation of the photogating potential, which shifts the transmission window within the transmission spectrum; the presence of localized or trap-related states, which modify the available density of states; and the inherently quantum-mechanical nature of carrier transport in ML-TMDCs. Unlike conventional classical

diodes, the peaks observed in these curves arise from the sequential opening and closing of quantum transmission channels. Whenever a quasibound state becomes energetically aligned with the transmission window, the carrier transmission probability is enhanced, leading to an increase in the transmission coefficient and, consequently, in the quantum conductance. The current therefore reaches a local maximum. As the photogating potential is increased further, this state moves out of resonance with the transmission window, reducing the tunneling probability and causing the current to decrease. Thus, without any reduction in the number of available carriers, a decrease in their transmission probability alone can give rise to negative differential resistance (NDR). In the present structure, this process is controlled by the photogating potential rather than solely by the externally applied bias, which constitutes one of the key novel aspects of this work. In fact, the observed current oscillations provide direct evidence of quantum transport and the successive realignment of electronic states. In other words, photogating effectively modifies the local electrostatic potential and thereby alters the band structure and tunneling probability, ultimately leading to pronounced modulation of the current response. The presence of numerous small ripples in all four curves is another notable feature of these results. Since transport within the Landauer–Büttiker framework is treated as fully coherent, multiple reflections of the electron wave between different regions of the structure can give rise to quantum oscillations resembling Fabry–Pérot interference. These interference effects produce fine-scale modulations in the transmission probability and, consequently, appear as small ripples superimposed on the overall current response. Such features further reflect the coherent wave nature of carrier transport in the considered ML-TMDC structures.

As shown in Fig. 4, the photogating-induced current is on the order of $0.2 \mu\text{A}$, without the involvement of any external gate or additional control parameter in its generation. The emergence of such a current in response to a relatively small variation of the photogating potential over the range $V_{\text{pg}} = 0 - 8\text{mV}$ demonstrates the pronounced sensitivity of carrier transport in ML-TMDCs to trapped charges. Although the absolute current magnitude lies within the moderate range compared with that reported for some high-gain two-dimensional phototransistors, the generation of a current on the order of hundreds of nanoamperes without an externally applied gate is nevertheless significant from the perspective of low-power optoelectronic devices, photodetectors, and optical switching applications. From this standpoint, the photogating potential can be regarded as an effective mechanism for directly controlling carrier transport and tuning the electrical response of the considered structures. In particular, even small variations in V_{pg} can produce appreciable changes in the current, thereby enabling optical control of the electrical current without requiring an external gate. The results reveal that the inclusion of spin–orbit coupling produces a remarkable enhancement of the current in both materials. This behavior can be attributed to the modification of the electronic density of states and, consequently, to changes in the number of available transmission channels, together with shifts in the positions of the transmission resonances. The effect is substantially stronger in ML – WSe₂, owing to its considerably larger spin–orbit splitting in the valence band, which is approximately 230 meV, compared with about 80 meV for ML – MoS₂. Accordingly, the current in ML – WSe₂ is expected to exhibit a stronger response to

photogating-induced modifications of the electronic structure. As further evident from Fig. 4, the modified peak-to-valley ratio (mPVR) for the two structures lies approximately in the range of 1.01–1.03. A more effective photogating-induced shift of the resonant states enhances the difference between the peak and valley currents, thereby increasing the mPVR. Although the magnitude of this parameter is smaller than that reported for conventional tunnel diodes and various other resonant-tunneling structures, it is worth to note that the underlying NDR mechanism in the present system is fundamentally different. Here, NDR does not originate from conventional multilayer tunneling barriers; rather, it results from trap-charge-induced photogating and the consequent modulation of the transmission spectrum. Therefore, the obtained mPVR values demonstrate that photogating can effectively modulate quantum carrier transport in ML-TMDC-based structures. The dependence of both the mPVR and the transmitted current on V_{pg} thus provides a basis for optically tunable transport characteristics and the development of devices in which light serves as a direct control parameter for electrical current.

Conclusion:

Finally, Our results demonstrate that trap-charge-induced photogating can provide an effective route for controlling quantum transport in ML – WSe₂ and ML – MoS₂ through an internally generated electrostatic potential. Both materials exhibit a pronounced non-monotonic current response to relatively small variations of V_{pg} , confirming the strong sensitivity of ballistic transport to trapped charge. The observed NDR is governed by the photogating-induced realignment of resonant transmission states with the transport window, rather than by conventional bias-driven carrier injection. The stronger response obtained for ML – WSe₂ is consistent with its larger spin–orbit-induced valence-band splitting and stronger valley dependent optical response. Although the resulting NDR contrast is modest, its significance lies in the optical controllability and fundamentally different origin of the nonlinear transport, in which light-generated trapped charge directly regulates quantum transmission. These findings establish a physical basis for optically controlled nonlinear transport in ML-TMDCs and motivate further exploration of photogating-based low-power switching, signal processing, sensing, memory, and neuromorphic devices.

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Data Availability

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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Supplementary Section 1:

Optical Properties of ML-TMDCs

*Calculation of the Absorption Coefficient of ML-TMDCs

Analytical investigation of the optical properties of ML-TMDCs, including the absorption coefficient and oscillator strengths between different energy states, based on a Hamiltonian formalism, is one of the main objectives of this study. In the following, employing a semiclassical approach in conjunction with perturbation theory, we derive fully analytical expressions for the interaction Hamiltonian and the absorption coefficient of the considered structure. We begin with the massive Dirac-like Hamiltonian governing ML-TMDCs and define the Fermi velocity of the monolayer as $v_f = at/\hbar$. The Hamiltonian for the structure under consideration can then be written as follows [1–3]:

$$H = \hbar v_f (\tau k_x \sigma_x + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z + \frac{\lambda s \tau}{2} (I_2 - \sigma_z) \quad (A1)$$

$\hat{\sigma}_{x,y,z}$ denotes the Pauli matrices, a is the lattice constant of each ML-TMDC with a hexagonal crystal structure, t is the effective hopping energy, Δ is the monolayer band gap, and 2λ represents the spin–orbit coupling (SOC) splitting. The indices s and τ denote the spin and valley indices, respectively, with $\tau = \pm 1$ corresponding to the K and K' valleys. The parameters of the ML-TMDC Hamiltonian for the different materials considered in this study are summarized in Table A1.

Table A1. Structural parameters of various ML-TMDC semiconductors

Material	$z = a \text{ (Å)}$	$\Delta \text{ (eV)}$	$t \text{ (eV)}$	$2\lambda \text{ (eV)}$
ML – MoS ₂	3.193	1.66	1.10	0.15
ML – MoSe ₂	3.313	1.47	0.94	0.18
ML – WS ₂	3.197	1.79	1.37	0.43
ML – WSe ₂	3.310	1.60	1.19	0.46

In the presence of an electromagnetic field, the momentum is modified according to $\mathbf{p} \rightarrow \mathbf{p} + \frac{e}{c} \mathbf{A}$, where $\mathbf{p}$ denotes the electron momentum operator and $\mathbf{A}$ is the vector potential. Accordingly, the Hamiltonian of the monolayer in the presence of the electromagnetic field can be written as follows:

$$\hat{H} = -i\hbar v_f \left[\tau \sigma_x \left(\partial_x - \frac{e}{i\hbar c} A_x \right) + \sigma_y \left(\partial_y - \frac{e}{i\hbar c} A_y \right) \right] + \frac{\Delta}{2} \hat{\sigma}_z - \frac{\lambda \tau s}{2} (\hat{\sigma}_z - \hat{1}) \quad (A2)$$

Thus, the total Hamiltonian of the system in the presence of the electromagnetic field can be expressed as $\hat{H}_t = \hat{H}_{\text{TMDC}} + \hat{H}_{\text{em}}$. This Hamiltonian can be conveniently decomposed into a

non-interacting Hamiltonian ($\hat{H}_0$) and an interaction Hamiltonian ($\hat{H}_{\text{int}}$), such that it can be rewritten as $\hat{H}_t = \hat{H}_0 + \hat{H}_{\text{int}}$. Finally, by defining $\boldsymbol{\sigma}$ for the K valley as $\boldsymbol{\sigma} = \sigma_x \mathbf{i} + \sigma_y \mathbf{j}$, the interaction Hamiltonian for this valley can be simplified to $\hat{H}_{\text{int}} = \frac{ev_F}{c}(\boldsymbol{\sigma} \cdot \mathbf{A})$. Likewise, by defining $\boldsymbol{\sigma}'$ for the K' valley as $\boldsymbol{\sigma}' = -\sigma_x \mathbf{i} + \sigma_y \mathbf{j}$, the corresponding interaction Hamiltonian for this valley is obtained as $\hat{H}_{\text{int}} = \frac{ev_F}{c}(\boldsymbol{\sigma}' \cdot \mathbf{A})$.

The effect of the electromagnetic field on the solid-state system is treated as a perturbation, driving the system from its initial state ψ_i to a final state ψ_f after a time t . Within first-order perturbation theory, only the term linear in the electromagnetic field is retained, given by $H_A = H_{\text{int}}$. We then examine the transition between the initial and final states induced by the incident light. By considering the time-dependent Schrödinger equation and carrying out the necessary calculations, the final form of Fermi's golden rule is obtained as follows:

$$a_f(t_f) = -\frac{i}{\hbar} \int_{t_i}^{t_f} H_{\text{int},fi} e^{-i(E_{0i}-E_{0f})t/\hbar} dt \quad (\text{A3})$$

Consequently, $P_{fi} = |a_f(t_f)|^2$ represents the transition probability of a carrier from the initial state ψ_i to the final state ψ_f . We assume that a classical optical field $\mathbf{E}$ is incident on the system and is described by:

$$\mathbf{E} = -E_0 \cos(\mathbf{k}' \cdot \mathbf{r} - \omega t) \hat{\mathbf{e}}' \quad (\text{A4})$$

where $\hat{\mathbf{e}}'$ denotes the polarization vector of the incident electromagnetic field and ω is the corresponding angular frequency. If the electromagnetic wave is incident on the x-y plane at an θ angle (Fig. 1), the coordinates of the incident wave are denoted by primed coordinates, whereas the coordinates of the structure are represented by unprimed coordinates. Accordingly, based on the considerations presented above, if the propagation direction of the electromagnetic wave is $\hat{\mathbf{x}}'$ along ($\mathbf{k}' = k\hat{\mathbf{x}}'$), the circular polarization vectors in the primed coordinate system for right- and left-handed circularly polarized light are given by $\hat{\mathbf{e}}'_{\text{RHS}} = (\hat{\mathbf{y}}' + i\hat{\mathbf{z}}')/\sqrt{2}$ and $\hat{\mathbf{e}}'_{\text{LHS}} = (\hat{\mathbf{y}}' - i\hat{\mathbf{z}}')/\sqrt{2}$, respectively.

Using the three relations given above, together with the interaction Hamiltonian of the structure and the defined classical electromagnetic field, we proceed to solve the problem. We define the vector potential associated with the incident field as $\mathbf{A}_{\text{opt}} = \frac{ic\mathbf{E}}{\omega}$. For the K valley, substituting this vector potential and the classical optical field into $\hat{H}_{\text{int}}$ yields the following interaction Hamiltonian:

$$\hat{H}_{\text{int}} = \frac{E_0 ev_F}{2i\omega} [e^{i(\mathbf{k}' \cdot \mathbf{r} - \omega t)} + e^{-i(\mathbf{k}' \cdot \mathbf{r} - \omega t)}] \boldsymbol{\sigma} \cdot \hat{\mathbf{e}}' \quad (\text{A5})$$

When light is incident on the system, it can induce electric dipole, electric quadrupole, and magnetic dipole transitions in the material.

We assume that the incident radiation lies in the visible and infrared spectral ranges, with a wavelength of approximately $\lambda = 400 - 600$ nm, which is much larger than the unit-cell size of the ML-TMDCs, $\Lambda = 0.3 - 0.4$ nm, i.e., $\lambda \gg \Lambda$. Therefore, we can infer the relation $\mathbf{k} \cdot \mathbf{r} \ll 1$, and the interaction Hamiltonian takes the form $\hat{H}_{\text{int}} \cong \mathbf{p} + i(\mathbf{k} \cdot \mathbf{r})\mathbf{p} \cong \mathbf{p}$. Under these physical conditions, the electric-dipole contribution is the dominant transition mechanism. Accordingly, we employ the electric-dipole approximation. Finally, $\hat{H}_{\text{int}}$ is obtained in the form $\hat{H}_{\text{int}} = \frac{E_0 e v_F}{2i\omega} [e^{i\omega t} + e^{-i\omega t}] \boldsymbol{\sigma} \cdot \hat{\mathbf{e}}'$.

We now return to the calculation of the transition probability. To evaluate this quantity, we first need to calculate $H_{\text{int,fi}}$ in the form $\hat{H}_{\text{int,fi}}(\mathbf{x}) = \frac{E_0 e v_F}{2i\omega} [e^{i\omega t} + e^{-i\omega t}] \langle \psi_{\text{of}}(\mathbf{q}) | \boldsymbol{\sigma} \cdot \hat{\mathbf{e}}' | \psi_{\text{oi}}(\mathbf{q}) \rangle$. Furthermore, according to the definition of $E_{\text{of}} - E_{\text{oi}} = \hbar\omega_{\text{fi}}$, the above integral can be rewritten as:

$$\int_{t_i}^{t_f} H_{\text{int,fi}} e^{-i(E_{\text{oi}} - E_{\text{of}})t/\hbar} dt = \int_{t_i}^{t_f} \left(\frac{E_0 e v_F}{2i\omega} [e^{i\omega t} + e^{-i\omega t}] \langle \psi_{\text{of}}(\mathbf{q}) | \boldsymbol{\sigma} \cdot \hat{\mathbf{e}}' | \psi_{\text{oi}}(\mathbf{q}) \rangle \right) e^{i\omega_{\text{fi}} t} dt \quad (\text{A6})$$

By defining $F(\mathbf{x})$ as $F(\mathbf{x}) = \langle \psi_{\text{of}}(\mathbf{q}) | \boldsymbol{\sigma} \cdot \hat{\mathbf{e}}' | \psi_{\text{oi}}(\mathbf{q}) \rangle$, and noting that the wavefunctions have no explicit time dependence, the above expression can be written as $\int_{t_i}^{t_f} H_{\text{int,fi}} e^{-i(E_{\text{oi}} - E_{\text{of}})t/\hbar} dt = \frac{E_0 e v_F}{2i\omega} F(\mathbf{x}) \int_{t_i}^{t_f} [e^{i(\omega_{\text{fi}} + \omega)t} + e^{i(\omega_{\text{fi}} - \omega)t}] dt$. Taking t as the transition time and employing the rotating-wave approximation, or equivalently considering the resonant transitions, together with the properties of the Dirac delta function, we obtain the transition probability in the following form:

$$P_{\text{fi}} = \left| \frac{E_0 e v_F F(\mathbf{x})}{\hbar\omega} \right|^2 \frac{\pi t}{2} \delta(\omega_{\text{fi}} - \omega) \quad (\text{A7})$$

The total transition probability P_t is obtained by integrating over all the states involved in the transition and is defined as $P_t = \left| \frac{E_0 e v_F}{\hbar\omega} \right|^2 \frac{t}{8\pi} \int_{-\infty}^{+\infty} |F(\mathbf{x})|^2 \delta(\omega_{\text{fi}} - \omega) d\mathbf{K}$.

Interband absorption is the dominant absorption mechanism and is particularly important in crystals with a direct band gap. The dependence of the absorption coefficient on the frequency of the incident electromagnetic radiation can be determined from the definition of the absorption coefficient. Considering the transition in the vicinity of $\mathbf{K} = 0$, introducing the reduced mass as $\frac{1}{m_r} = \frac{1}{m_c} + \frac{1}{m_v}$, and applying the divergence theorem to convert the volume integral into a surface integral, while accounting for the two-dimensional nature of the structure, the transition probability is obtained as

$$P_t = \frac{1}{2\sqrt{2}} |E_0 e v_F F(\mathbf{x})|^2 \left(\frac{(m_r)^{3/2}}{\hbar^2} \right) \left(\frac{\sqrt{\hbar\omega - E_g}}{(\hbar\omega)^2} \right) t \quad (\text{A8})$$

We next establish the relationship between the transition probability and the absorption coefficient. The absorption coefficient is defined as the transition rate per unit photon flux, given by $\alpha = \left(\frac{1}{\Phi}\right) \frac{dP_{tot}}{dt}$. The photon flux is, in turn, defined as the number of incident photons per unit area per unit time, i.e., $\Phi = \frac{\langle S \rangle}{\hbar\omega}$. It can also be determined from the time-averaged magnitude of the Poynting vector (S), normalized by the incident photon energy, according to $\langle S \rangle = \frac{1}{2} c_0 \epsilon_0 n |E_0|^2$. Therefore, based on these definitions, the absorption coefficient can be directly expressed in terms of the transition probability. Substituting the transition probability obtained for the present structure from Eq. (A8) yields the following fully analytical expression for the absorption coefficient:

$$\alpha_{TMDC} = \left(\frac{e^2 v_F^2 (m_F)^{3/2}}{\sqrt{2} c_0 \epsilon_0 n \hbar^2} \right) |F(x)|^2 \left[\left(\frac{\sqrt{\hbar\omega - E_g}}{\hbar\omega} \right) \right] \Theta(\hbar\omega - E_g) \quad (A9)$$

A key feature of ML-TMDCs is their unique optical response. Owing to their electronic structure and the associated optical selection rules, light with $\hat{e}'_{RHS}$ polarization couples selectively to the K valley, whereas light with $\hat{e}'_{LHS}$ polarization couples selectively to the K' valley. As discussed above, for the K valley, the matrix σ , which was defined for an individual monolayer, takes the form $\sigma = \sigma_x \hat{x} + \sigma_y \hat{y}$. Similarly, when the K' valley is considered, σ' is defined as $\sigma' = -\sigma_x \hat{x} + \sigma_y \hat{y}$. Furthermore, as derived above, $F(x)$ is given by $F(x) = \langle \psi_{of}(q) | \sigma \cdot \hat{e}' | \psi_{oi}(q) \rangle$. We can therefore obtain $F(x)$ in the unprimed coordinate system by transforming the primed coordinates using the corresponding coordinate-transformation matrix. After carrying out the necessary algebra, $F(x)$ is obtained in the following form:

$$F(x) = \langle \psi_{of}(q) | \sigma \cdot \hat{e}' | \psi_{oi}(q) \rangle = \begin{cases} F_{RHS}(x) = \frac{1}{\sqrt{2}i} (\pm \sin \theta F_1(x) + F_2(x)) & \text{for K(K') valley} \\ F_{LHS}(x) = \frac{1}{\sqrt{2}i} (\mp \sin \theta F_1(x) + F_2(x)) & \text{for K(K') valley} \end{cases} \quad (A10)$$

If the functions $g_{1(2)}$ are defined as $g_{1(2)} = [\psi_{of,A}^*(q) \psi_{oi,B}(q)] \pm [\psi_{of,B}^*(q) \psi_{oi,A}(q)]$, and the relation $F_{1(2)}(x) = \int_{-\infty}^{+\infty} dx g_{1(2)}$ is satisfied, then $F_{RHS}(x)$ can be simplified and expressed as $F_{RHS}(x) = \frac{1}{\sqrt{2}i} (\sin \theta F_1(x) + F_2(x))$. We can next consider the K' valley for light with the same right-handed circular polarization. Following the same procedure and using the definitions introduced above, we ultimately obtain the relation $F_{RHS}(x) = \frac{1}{\sqrt{2}i} (-\sin \theta F_1(x) + F_2(x))$ for this particular case. The same analysis can then be repeated for left-handed circularly polarized light.

In the subsequent calculations, we need to derive analytical expressions for the coefficients $F_{1(2)}$ consistent with the specific structure under consideration. Owing to their crystal structure, ML-TMDCs consist of two sublattices, denoted by A and B. Accordingly, the wavefunction of these materials is represented by a two-component spinor, $\psi_k = \begin{pmatrix} \psi_{k,A} \\ \psi_{k,B} \end{pmatrix}$, and the corresponding

eigenfunctions and eigenvalues have been extensively reported in the literature [4,5]. By employing these material-specific properties together with Fourier transformations, we evaluate the integral in two-dimensional polar coordinates and ultimately obtain the following analytical expression:

$$F_1 = \frac{\pi^3}{3} \left(\frac{\Delta_{s\tau}}{\hbar v_F} \right)^2 \left[1 - \left(1 + \left(\frac{2\hbar v_F}{\Delta_{s\tau}} \right)^2 k_{\text{cutoff}}^2 \right)^{3/2} \right] \quad (\text{A11})$$

Here, the effective band gap is defined as $\Delta_{s\tau} = \Delta - \lambda s\tau$, and the relation $k_{\text{cutoff}} = \frac{\pi}{a_{\text{lattice}}}$ has also been used. This model therefore describes the carriers in the vicinity of the K and K' points in momentum space, with the corresponding wave functions represented in spinor form. The same procedure can subsequently be applied to F_2 , which ultimately leads to the same expression given above. Consequently, we obtain $F_1 = F_2$.

Oscillator Strength (f_{fi}):

An important optical parameter of the structure is the quantum-mechanical oscillator strength. In quantum mechanics, the dimensionless quantity f_{fi} is referred to as the oscillator strength associated with the transition frequency ω_{fi} and is defined as $f_{fi} = \left(\frac{2m\omega_{fi}}{e^2\hbar} \right) |\mathbf{d}_{fi}|^2$, where $\mathbf{d}_{fi}$ denotes the electric-dipole matrix element between the initial and final states. The central task in this section is therefore to calculate the dipole moment for the system under consideration, namely, to evaluate $\mathbf{d}_{fi} = \langle \psi_f | \mathbf{er} | \psi_i \rangle$. To calculate this quantity, we first evaluate the commutator of the position operator $\mathbf{r}$ with the Hamiltonian H , i.e., $[\mathbf{r}, \hat{H}_t(x)]$. Considering the K valley and rewriting the Hamiltonian of the structure, the Hamiltonian takes the form $\hat{H}_t = v_F \boldsymbol{\sigma} \cdot \mathbf{p} + \frac{\Delta}{2} \hat{\sigma}_z - \frac{\lambda s}{2} (\hat{\sigma}_z - \hat{1})$. Since the position operator $\mathbf{r}$ commutes with the spin components, $[\mathbf{r}, \hat{\sigma}_z] = 0$, we obtain $[\mathbf{r}, \hat{H}_t(x)] = [\mathbf{r}, v_F \boldsymbol{\sigma} \cdot \mathbf{p}] = i\hbar v_F \boldsymbol{\sigma}$.

Sandwiching this relation between the initial and final states of the system yields $\langle \psi_{of} | [\mathbf{r}, \hat{H}_t(x)] | \psi_{oi} \rangle = i\hbar \langle \psi_{of} | v_F \boldsymbol{\sigma} | \psi_{oi} \rangle$. On the other hand, expanding the first term on the right-hand side of this relation gives $\langle \psi_{of} | [\mathbf{r}, \hat{H}_t(x)] | \psi_{oi} \rangle = (E_i - E_f) \langle \psi_{of} | \mathbf{r} | \psi_{oi} \rangle$. Comparing this result with the second part of the relation ultimately leads to the following expressions for the dipole-moment matrix elements in the K and K' valleys:

$$\begin{cases} \mathbf{d}_{fi} = \langle \psi_{of} | \mathbf{er} | \psi_{oi} \rangle = \frac{e\hbar v_F}{i\hbar\omega_{fi}} \langle \psi_{of} | \boldsymbol{\sigma} | \psi_{oi} \rangle & \text{for K valley} \\ \mathbf{d}'_{fi} = \langle \psi_{of} | \mathbf{er} | \psi_{oi} \rangle = \frac{e\hbar v_F}{i\hbar\omega_{fi}} \langle \psi_{of} | \boldsymbol{\sigma}' | \psi_{oi} \rangle & \text{for K' valley} \end{cases} \quad (\text{A12})$$

Consequently, the oscillator strength f_{fi} , which plays a crucial role in determining the optical absorption, is obtained as

$$\begin{cases} f_{fi} = \left(\frac{2mv_F^2}{\hbar\omega_{fi}} \right) | \langle \psi_{of} | \boldsymbol{\sigma} | \psi_{oi} \rangle |^2 & \text{for K valley} \\ f'_{fi} = \left(\frac{2mv_F^2}{\hbar\omega_{fi}} \right) | \langle \psi_{of} | \boldsymbol{\sigma}' | \psi_{oi} \rangle |^2 & \text{for K' valley} \end{cases} \quad (A13)$$

Finally, using the definitions of $\boldsymbol{\sigma}$ and $\boldsymbol{\sigma}'$ for the K and K' valleys, respectively, together with the expressions for $F_{1(2)}$, the modified expression for the oscillator strength is obtained as

$$f_{fi} = f'_{fi} = \left(\frac{2mv_F^2}{\hbar\omega_{fi}} \right) (|F_1(x)|^2 + |F_2(x)|^2) \quad (A14)$$

In conclusion, a fully analytical framework has been developed here for investigating the optical properties of ML-TMDCs.

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Supplementary Section 2:

Transport Properties of ML-TMDCs

In this section, we again employ the effective massive Dirac-like Hamiltonian governing ML-TMDC semiconductors (Eq. A1) and use the extended transfer-matrix method to investigate the carrier transmission probability through the structure in the presence of the photogating potential. As illustrated in Fig. B1, the monolayer is assumed to extend in the x-y plane, with carrier transport considered along the in-plane x direction. As shown schematically, carriers are incident on the structure at an angle θ_0 and emerge from the structure at an angle θ_e .

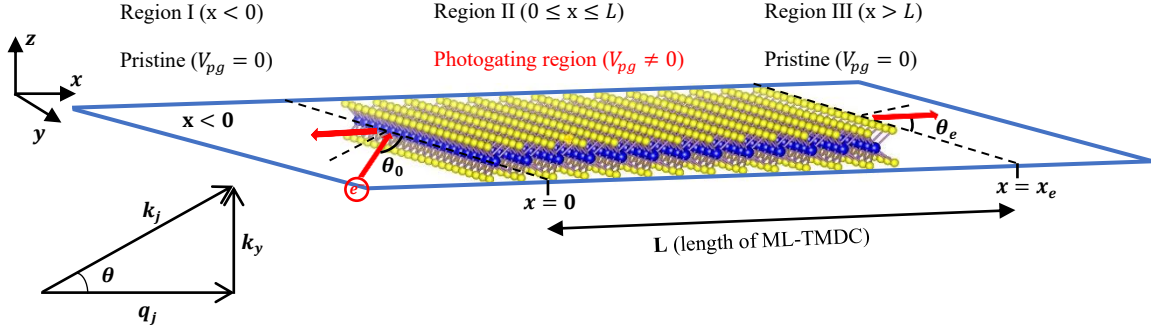


Figure B1. Schematic illustration of carrier transport through a photogated ML-TMDC, showing the incident and transmitted carrier wave vectors and the corresponding angles θ_0 and θ_e .

$$H = \hbar v_f (\tau k_x \sigma_x + k_y \sigma_y) + \frac{\Delta}{2} \sigma_z + \frac{\lambda s \tau}{2} (I_2 - \sigma_z) + e V_{pg} I_2 \quad (B1)$$

The wave vector components along the x- and y-directions are defined as $k_x = -i \partial_x$ and $k_y = -i \partial_y$, respectively. This Hamiltonian acts on a state described by the two-component pseudospin wavefunction $\Psi = (\psi_A, \psi_B)$, where ψ_A and ψ_B correspond to the two sublattices of ML-TMDCs. Since carrier propagation is considered only along the x-direction, the wavefunction can be written in the form $\psi_{A,B}(x, y) = \psi_{A,B}(x) e^{ik_y y}$. Substituting this wavefunction into the above Hamiltonian, the resulting problem for ML-TMDCs can be solved as follows:

$$\begin{pmatrix} \frac{\Delta}{2} + e V_{pg} & \hbar v_f (\tau k_x - i k_y) \\ \hbar v_f (\tau k_x + i k_y) & \lambda \tau s - \frac{\Delta}{2} + e V_{pg} \end{pmatrix} \begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix} e^{ik_y y} = E \begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix} e^{ik_y y} \quad (B2)$$

E denotes the energy of the incident charge carriers, while $k_+ = \left[\frac{(E + \frac{\Delta}{2} - e V_{pg}) - \lambda \tau s}{\tau \hbar v_f} \right]$ and $k_- =$

$\left[\frac{(E - \frac{\Delta}{2} - e V_{pg})}{\tau \hbar v_f} \right]$ are defined as the wave vectors within the monolayer, and $k_0 = \frac{E}{\tau \hbar v_f}$ corresponds

to the wave vector of the incident carrier in the $x < 0$ region. By expressing the above equation

as two coupled equations and differentiating with respect to x , a second-order differential equation for the A and B sublattices can be obtained by defining $k_j^2 = k_+ k_-$ as:

$$\begin{cases} \frac{d^2}{dx^2} \psi_A(x) + (k_j^2 - k_y^2) \psi_A(x) = 0 \\ \frac{d^2}{dx^2} \psi_B(x) + (k_j^2 - k_y^2) \psi_B(x) = 0 \end{cases} \quad (B2)$$

If q_j denotes the x-component of the wave vector within the monolayer, it is obtained as

$$q_j = \begin{cases} \text{sgn}(k_+) \sqrt{k_j^2 - k_y^2} & , k_j^2 > k_y^2 \\ i \sqrt{k_y^2 - k_j^2} & , \text{otherwise} \end{cases} \quad (B3)$$

For the wavefunctions, we assume solutions of the form $\psi_A(x) = Ae^{iq_j x} + Be^{-iq_j x}$ and $\psi_B(x) = Ce^{iq_j x} + De^{-iq_j x}$. Here, A and C denote the forward-propagating spinor components, whereas B and D represent the backward-propagating spinor components. Substituting these wavefunctions into Eq. (B2) yields the relations $C = \frac{ik_-}{iq_j + \frac{1}{\tau}k_y} A$ and $D = -\frac{ik_-}{iq_j - \frac{1}{\tau}k_y} B$. To establish the relationship between the wavefunctions $\psi_{A,B}(x)$ and $\psi_{A,B}(x + \Delta x)$, we assume the basis functions $\varphi(x) = \begin{pmatrix} \varphi_1(x) \\ \varphi_2(x) \end{pmatrix}$ to be of the following form:

$$\begin{cases} \varphi_1(x) = Ae^{iq_j x} + Be^{-iq_j x} \\ \varphi_2(x) = Ae^{iq_j x} - Be^{-iq_j x} \end{cases} \quad (B4)$$

We rewrite the wavefunctions in terms of the basis states. Using the definitions given by Eqs.

$P = \frac{k_-}{k}$, $\sin(\theta) = \frac{k_y}{k}$, and $\cos(\theta) = \frac{q}{k}$, the wavefunctions can be expressed as follows:

$$\begin{cases} \psi_A(x) = \varphi_1(x) \\ \psi_B(x) = \frac{i}{\tau} P \sin(\theta) \varphi_1(x) + P \cos(\theta) \varphi_2(x) \end{cases} \quad (B5)$$

If the matrix $R(E, V_{pg})$ is defined as the transformation matrix in the form $\begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix} = R(E, V_{pg}) \begin{pmatrix} \varphi_1(x) \\ \varphi_2(x) \end{pmatrix}$, then we obtain:

$$R(E, V_{pg}) = \begin{pmatrix} 1 & 0 \\ \frac{i}{\tau} P \sin(\theta) & P \cos(\theta) \end{pmatrix} \quad (B6)$$

Next, we consider the relationship between the wavefunction $\begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix}$ and the wavefunction $\begin{pmatrix} \psi_A(x + \Delta x) \\ \psi_B(x + \Delta x) \end{pmatrix}$ within the ML-TMDC plane:

$$\begin{cases} \varphi_1(x + \Delta x) = Ae^{iq(x+\Delta x)} + Be^{-iq(x+\Delta x)} \\ \varphi_2(x + \Delta x) = Ae^{iq(x+\Delta x)} - Be^{-iq(x+\Delta x)} \end{cases} \quad (B7)$$

Substituting these relations into $\begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix} = R(E, V_{pg}) \begin{pmatrix} \varphi_1(x) \\ \varphi_2(x) \end{pmatrix}$, and subsequently expressing the resulting equation in matrix form as $\begin{pmatrix} \psi_A(x+\Delta x) \\ \psi_B(x+\Delta x) \end{pmatrix} = T(\Delta x, E, V_{pg}) \begin{pmatrix} \varphi_1(x) \\ \varphi_2(x) \end{pmatrix}$, we obtain:

$$T(\Delta x, E, V_{pg}) = \begin{pmatrix} \cos(q\Delta x) & i\sin(q\Delta x) \\ iP \left[\frac{1}{\tau} \sin(\theta) \cos(q\Delta x) + \cos(\theta) \sin(q\Delta x) \right] & -P \left[\frac{1}{\tau} \sin(\theta) \sin(q\Delta x) - \cos(\theta) \cos(q\Delta x) \right] \end{pmatrix} \quad (B8)$$

Using the two transformation relations for the matrices $R(E, V_{pg})$ and $T(\Delta x, E, V_{pg})$, the relationship between the wavefunctions $\begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix}$ and $\begin{pmatrix} \psi_A(x+\Delta x) \\ \psi_B(x+\Delta x) \end{pmatrix}$ can then be written as:

$$\begin{pmatrix} \psi_A(x+\Delta x) \\ \psi_B(x+\Delta x) \end{pmatrix} = T_j(\Delta x, E, V_{pg}) R_j^{-1}(E, V_{pg}) \begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix} \quad (B9)$$

Finally, the relationship between $\psi_{A,B}(x)$ and $\psi_{A,B}(x + \Delta x)$ is formulated in terms of the extended transfer matrix $M(\Delta x, E, V_{pg}) = T(\Delta x, E, V_{pg}) R^{-1}(E, V_{pg})$. This matrix shifts the wavefunction $\begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix}$ by a distance Δx and transforms it into $\begin{pmatrix} \psi_A(x+\Delta x) \\ \psi_B(x+\Delta x) \end{pmatrix}$. Accordingly, the individual elements of the matrix $M(\Delta x, E, V_{pg})$ are obtained as follows:

$$M(\Delta x, E, V_{pg}) = \begin{pmatrix} \frac{\cos(\theta) \cos(q\Delta x) + \frac{1}{\tau} \sin(\theta) \sin(q\Delta x)}{\cos(\theta)} & i \frac{\sin(q\Delta x)}{P \cos(\theta)} \\ i \frac{P_j \sin(q\Delta x)}{\cos(\theta)} & \frac{\cos(\theta) \cos(q\Delta x) - \frac{1}{\tau} \sin(\theta) \sin(q\Delta x)}{\cos(\theta)} \end{pmatrix} \quad (B10)$$

One of the most important quantities that should be considered in the study of both the transport and optical properties is the wavefunction of the structure. The two-component wavefunction at the position x_0 , denoted by $\begin{pmatrix} \psi_A(x_0) \\ \psi_B(x_0) \end{pmatrix}$, can be determined from the boundary conditions, as illustrated in Fig. B1. We assume that a free electron with energy E is incident on the structure from the $x_0 < 0$ region at an angle θ_0 . In this region, the total wavefunction is given by the superposition of the incident and reflected waves. Therefore, at $x_0 = 0$, we have:

$$\psi_A(0) = \psi_i(E) + \psi_r(E) = (1 + r)\psi_i(E) \quad (B11)$$

As shown in Fig. B1, $\psi_i(E)$ represents the wave packet of the incident electron at $x_0 = 0$. In deriving the above relations, the relation $\psi_r(E) = r\psi_i(E)$ has been used, where r is the overall reflection amplitude of the structure. Based on the proposed definition of the wavefunction, $\psi_A(x) = Ae^{iqx} + Be^{-iqx}$, the corresponding relation at the point of incidence is given by

$\psi_A(0) = A + B$. By combining Eq. (B11) with the above description, the relations $A = \psi_i(E)$ and $B = \psi_r(E) = r\psi_i(E)$ are obtained. Accordingly, the basis functions can be rewritten in the following form:

$$\begin{cases} \varphi_1(0) = (1 + r)\psi_i(E) \\ \varphi_2(0) = (1 - r)\psi_i(E) \end{cases} \quad (\text{B12})$$

On the other hand, we previously obtained the relation $\psi_B(x) = \frac{i}{\tau}P\sin(\theta)\varphi_1(x) + P\cos(\theta)\varphi_2(x)$. At the point $x_0 = 0$, and using the above relations, we obtain

$$\psi_B(0) = P_0[\frac{i}{\tau}\sin\theta_0 + \cos\theta_0]\psi_i(E, k_y) + P_0r[\frac{i}{\tau}\sin\theta_0 - \cos\theta_0]\psi_i(E, k_y) \quad (\text{B13})$$

Equation $\psi_{A(B)}(0)$ can be expressed in a compact matrix form as $\begin{pmatrix} \psi_A(0) \\ \psi_B(0) \end{pmatrix} = \begin{pmatrix} 1+r \\ P_0[(\cos\theta_0 + \frac{i}{\tau}\sin\theta_0) - r(\cos\theta_0 - \frac{i}{\tau}\sin\theta_0)] \end{pmatrix} \psi_i(E)$.

We now consider the other end of the structure, where the carrier exits the structure at position $x = x_e$ after undergoing various interactions, with an exit angle of θ_e . Assuming that t denotes the transmission amplitude of the structure, the corresponding wavefunction can be written as $\psi_A(x_e) = t\psi_i(E, k_y)$. Based on the definition of the wavefunctions in terms of the basis functions at position $x = x_e$, and assuming the relation $\varphi_{1,2}(x_e) = t\psi_i(E)$, the wavefunction at the end of the structure, i.e., at the carrier-exit position, can be expressed in the following compact matrix form $\begin{pmatrix} \psi_A(x_e) \\ \psi_B(x_e) \end{pmatrix} = \begin{pmatrix} t \\ tP_e(\cos\theta_e + \frac{i}{\tau}\sin\theta_e) \end{pmatrix} \psi_i(E)$.

Using the extended transfer matrix obtained for this structure, the matrix $M(\Delta x, E, V_{pg})$ relates the wavefunction $\begin{pmatrix} \psi_A(x_0) \\ \psi_B(x_0) \end{pmatrix}$ to $\begin{pmatrix} \psi_A(x_e) \\ \psi_B(x_e) \end{pmatrix}$, such that $\Delta x = x_e - x_0$. Therefore, we obtain the relation $\begin{pmatrix} \psi_A(x_e) \\ \psi_B(x_e) \end{pmatrix} = M(\Delta x, E, V_{pg}) \begin{pmatrix} \psi_A(x_0) \\ \psi_B(x_0) \end{pmatrix}$. By combining the relations obtained for $\begin{pmatrix} \psi_A(x_0) \\ \psi_B(x_0) \end{pmatrix}$ and $\begin{pmatrix} \psi_A(x_e) \\ \psi_B(x_e) \end{pmatrix}$, we arrive at:

$$\begin{pmatrix} t \\ tP_e(\cos\theta_e + \frac{i}{\tau}\sin\theta_e) \end{pmatrix} = \begin{pmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{pmatrix} \begin{pmatrix} 1+r \\ P_0[(\cos\theta_0 + \frac{i}{\tau}\sin\theta_0) - r(\cos\theta_0 - \frac{i}{\tau}\sin\theta_0)] \end{pmatrix} \quad (\text{B14})$$

Solving the above equation and using $\det M = 1$, the reflection coefficient r and transmission coefficient t of the structure, which are key quantities for characterizing its transport properties, are obtained for the K (K') valleys as follows:

$$r(E, k_y) = \frac{M_{22}P_0e^{\pm i\theta_0} - M_{11}P_0e^{\pm i\theta_e} - M_{12}P_0P_0e^{\pm i(\theta_e + \theta_0)} + M_{21}}{M_{22}P_0e^{\mp i\theta_0} + M_{11}P_0e^{\pm i\theta_e} - M_{12}P_0P_0e^{\pm i(\theta_e - \theta_0)} - M_{21}} \quad (\text{B15})$$

$$t(E, k_y) = \frac{2P_0 \cos \theta_0}{M_{22}P_0 e^{\mp i\theta_0} + M_{11}P_e e^{\pm i\theta_e} - M_{12}P_0 P_e e^{\pm i(\theta_e - \theta_0)} - M_{21}} \quad (\text{B16})$$

Finally, the wave function at an arbitrary position x can be written as $\begin{pmatrix} \psi_A(x) \\ \psi_B(x) \end{pmatrix} = M(\Delta x, E, V_{pg}) \begin{pmatrix} \psi_A(x_0=0) \\ \psi_B(x_0=0) \end{pmatrix}$. Accordingly, the wavefunctions corresponding to the different A and B sublattices are obtained as follows:

$$\begin{cases} \psi_A(x) = \psi_i(E) [(1+r)M_{11} + P_0 \left(\cos \theta_0 + \frac{i}{\tau} \sin \theta_0 \right) - r(\cos \theta_0 - \frac{i}{\tau} \sin \theta_0)] M_{12} \\ \psi_B(x) = \psi_i(E) [(1+r)M_{21} + P_0 \left(\cos \theta_0 + \frac{i}{\tau} \sin \theta_0 \right) - r(\cos \theta_0 - \frac{i}{\tau} \sin \theta_0)] M_{22} \end{cases} \quad (\text{B17})$$

Here, M_{mn} denote the elements of the matrix $M(\Delta x, E, V_{pg})$. These equations are used to describe the spatial evolution of the two-component pseudospin wave function in ML-TMDCs for a given incident electron wave packet $\psi_i(E)$.

Supplementary Files

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- [Supplementary1.docx](#)
- [Supplementary2.docx](#)