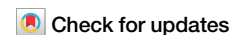




Intertwined bulk photocarrier and interfacial barrier dynamics in van der Waals point-contact Schottky junctions



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Schottky junctions based on transition-metal dichalcogenides underpin next-generation optoelectronic devices, yet the nanoscale carrier dynamics that ultimately govern their performance remain largely inaccessible. Here, we employ optical pump–probe time-resolved atomic force microscopy (OPP-TR-AFM) to investigate the spatiotemporal response of a WSe₂/PtIr van der Waals Schottky junction. Under backside illumination, photocarriers generated in bulk WSe₂ produce a local junction response through coupled bulk carrier transport and interfacial barrier modulation. We show that the time-resolved current is consistently explained by the competition between long-lived hole diffusion governed by the occupancy of bulk trap states and thermionic emission current modulated by the surface photovoltage. This mechanism is supported by transient absorption spectroscopy, light-modulated current–voltage measurements, and numerical simulations. Furthermore, spatial mapping reveals pronounced nanoscale variations in the coupled dynamics of carrier transport and interfacial barrier modulation. These results demonstrate that OPP-TR-AFM provides a new framework for resolving the coupled behavior of bulk carriers and interfacial barrier dynamics in van der Waals Schottky junctions with simultaneous spatial and temporal resolution, establishing it as a powerful approach for investigating the microscopic processes that play a crucial role in the development of next-generation optoelectronic devices.

Transition metal dichalcogenides (TMDCs) have attracted considerable attention as promising materials for next-generation optoelectronic devices owing to their strong light–matter interactions, tunable electronic structures, and excellent carrier transport properties^{1–3}. In particular, TMDC-metal Schottky junctions play a central role in a wide range of optoelectronic technologies, including photodetectors and energy-harvesting devices^{4,5}. Recently, WSe₂-based Schottky junctions have demonstrated nanosecond-scale photoresponses, highlighting their potential for high-speed optoelectronic applications^{6–8}. However, the microscopic processes governing carrier transport and photocurrent generation remain poorly understood.

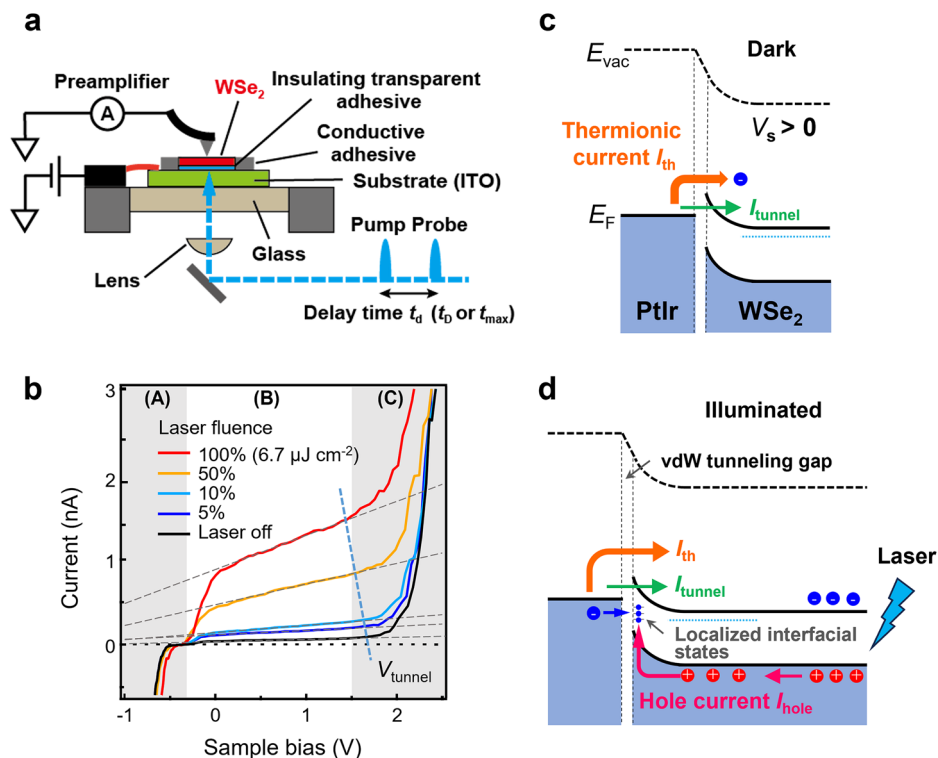
A central challenge is that photocurrent response in TMDC Schottky junctions is determined not only by carrier generation and transport within the semiconductor but also by the dynamic response of the interfacial energy landscape, including band bending, surface photovoltage (SPV), and trap-mediated processes^{9–15}. Because these processes evolve on nanometer and

nanosecond scales, conventional spatially averaged techniques, such as pump–probe spectroscopy and device-level electrical measurements, have limited ability to directly resolve the local interplay among them.

To address this challenge, we employed optical pump–probe time-resolved atomic force microscopy (OPP-TR-AFM), which combines optical pump–probe excitation with local electrical measurements to achieve simultaneous nanosecond temporal resolution and nanometer spatial resolution. A local van der Waals Schottky junction was formed between a conductive AFM probe and the surface of bulk WSe₂¹⁶, providing a junction geometry relevant to practical Schottky optoelectronic devices. In the backside-illumination configuration, photoexcited carriers are generated predominantly within the bulk WSe₂ and subsequently diffuse toward the interface. Consequently, the measured photocurrent reflects the coupled dynamics of bulk carrier transport and local interfacial barrier modulation.

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Fig. 1 | OPP-TR-AFM configuration and carrier transport model of the WSe₂/PtIr van der Waals Schottky junction. **a** Schematic illustration of the experimental configuration. Optical excitation was performed under backside illumination through a transparent substrate. The illuminated region of the WSe₂ crystal was electrically isolated from the ITO substrate by a transparent insulating adhesive layer, while the WSe₂ crystal was electrically connected to the ITO substrate only through a conductive adhesive applied outside the illuminated region. **b** Light-modulated I - V characteristics measured at different excitation fluences. The bias dependence is divided into three regimes: (A) the forward-bias region, (B) the reverse-bias region, and (C) the tunneling regime. The OPP-TR-AFM measurements presented in this study were performed primarily in region (B). The threshold bias for the onset of tunneling conduction is denoted by V_{tunnel} . **c** Schematic energy-band diagram of the WSe₂/PtIr junction in the dark. In the reverse-bias region, the current is dominated by the thermionic emission current I_{th} over the effective Schottky barrier. At sufficiently high bias, the tunneling current I_{tunnel} also contributes to the measured current. **d** Schematic illustration of the carrier transport processes under optical excitation. Photoexcited carriers are generated predominantly within the bulk WSe₂ region and subsequently diffuse toward the WSe₂/PtIr interface. Photoexcited holes contribute to the hole diffusion current I_{hole} , whereas charge accumulation in localized states near the interface modulates the effective Schottky barrier, thereby changing the thermionic emission current I_{th} .



To elucidate the origin of the observed response, we combined OPP-TR-AFM measurements with transient absorption spectroscopy (TAS) covering timescales from picoseconds to microseconds¹⁷, light-modulated current-voltage (I - V) measurements¹⁸, and numerical simulations. We demonstrate that the time-resolved current arises from the competition between long-lived hole diffusion governed by bulk trap occupancy and thermionic emission current modulated by the surface photovoltage. Furthermore, spatial mapping reveals pronounced nanoscale variations in these coupled processes. Together, these findings establish OPP-TR-AFM as a powerful approach for investigating the interplay between bulk carrier transport and interfacial barrier dynamics, providing new insight into the microscopic operation of van der Waals Schottky optoelectronic devices.

Results and discussion

Experimental configuration and operating principle

Figure 1a schematically illustrates the OPP-TR-AFM measurement system used in this study (see Supplementary Fig. 1 for details). A local van der Waals Schottky junction was formed by gently bringing a PtIr-coated conductive AFM cantilever into point contact with the surface of bulk WSe₂, providing a local junction geometry relevant to practical Schottky optoelectronic devices. Optical excitation was performed under backside illumination through a transparent substrate (see Methods). In this illumination geometry, photoexcited carriers are generated predominantly within the bulk WSe₂ and subsequently transported through the crystal, with a fraction reaching the local WSe₂/PtIr interface. Consequently, the measured time-resolved current reflects both carrier transport within the bulk and the temporal evolution of the interfacial Schottky barrier.

Figure 1b shows the light-modulated I - V characteristics of the WSe₂/PtIr junction measured under periodic light ON/OFF modulation. This measurement enables simultaneous evaluation of the current responses under dark and illuminated conditions. The I - V characteristics can be

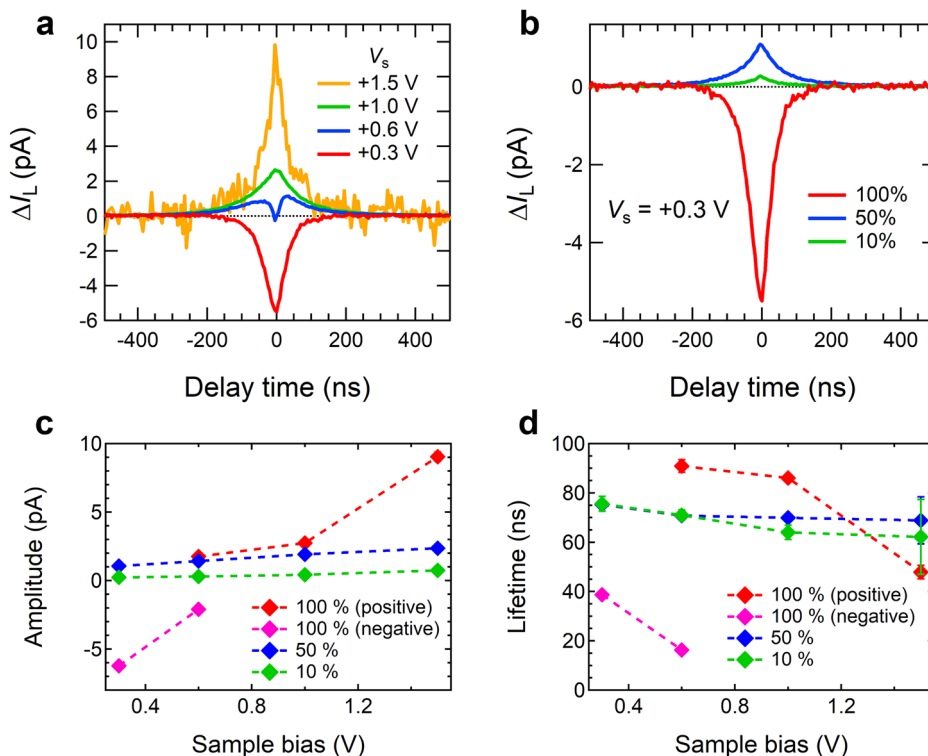
divided into three bias regimes: (A) the forward-bias region (sample bias, $V_s < -0.3$ V), (B) the reverse-bias region ($-0.3 < V_s < +1.5$ V), and (C) the tunneling regime ($V_s > +1.5$ V).

Under dark conditions, the I - V characteristics exhibit the rectifying behavior characteristic of a Schottky junction, consistent with the WSe₂ surface behaving effectively as an n -type semiconductor. Analysis of the forward-bias characteristics in region (A) based on the thermionic emission model^{19–21} yielded an effective Schottky barrier height of ~ 0.65 eV (Supplementary Fig. 2). In the reverse-bias region (B), the thermionic emission current I_{th} over the Schottky barrier is the dominant current component (Fig. 1c). At higher bias, in the tunneling regime (C), the tunneling current I_{tunnel} becomes dominant, suggesting that the depletion region near the WSe₂ surface is relatively narrow.

Under illumination, the onset voltage for tunneling conduction V_{tunnel} shifts systematically toward lower sample bias with increasing excitation fluence (Fig. 1b). This behavior indicates that optical excitation induces an SPV, thereby modifying the electrostatic potential near the interface^{22,23}. We attribute this shift to the transient accumulation of photoexcited holes in localized states near the interface, which partially relaxes the band bending and consequently reduces the sample bias required to initiate tunneling conduction.

Based on these observations, the carrier-transport model considered in this study is illustrated in Fig. 1d. Photoexcited holes generated in the bulk are transported toward the interface through trap-mediated diffusion and contribute to the hole current I_{hole} . Meanwhile, photoexcited charges accumulated in localized states near the interface partially relax the band bending, thereby modulating the Schottky barrier and changing I_{th} . Here, the interfacial localized states refer to localized electronic states that may form at the metal-semiconductor interface, such as those arising from local structural disorder at the WSe₂/PtIr contact or metal-induced gap states²⁴.

Fig. 2 | Bias- and excitation-fluence dependences of the time-resolved current in the WSe₂/PtIr junction. **a** Bias-voltage dependence of the time-resolved current $\Delta I_L(t_d)$ measured at an excitation fluence of $3.4 \mu\text{J cm}^{-2}$ with a contact force of 12 nN. **b** Excitation-fluence dependence of $\Delta I_L(t_d)$ measured at a sample bias of $V_s = +0.3 \text{ V}$ with a contact force of 12 nN. An excitation fluence of 100% corresponds to $3.4 \mu\text{J cm}^{-2}$. **c, d** Bias-voltage dependences of the amplitudes and lifetimes obtained by exponential fitting of $\Delta I_L(t_d)$ measured at excitation fluences of 10%, 50%, and 100%.



As shown below, the OPP-TR-AFM measurements exhibit characteristic bias- and excitation-fluence-dependent behaviors, particularly in bias region (B). To elucidate these behaviors, we discuss the origins of the delay-time, bias, and excitation-fluence dependences of the time-resolved current in terms of the contributions from I_{hole} and I_{th} modulated by the SPV.

OPP-TR-AFM measurements

Next, time-resolved current signals were measured using pump–probe pulse pairs in combination with the delay-time modulation method²⁵ (see Methods and Supplementary Fig. S1). The delay-time modulation method enables highly sensitive detection of the temporal response of the Schottky junction while effectively suppressing thermal background signals. In addition, no significant change in the time-resolved current was observed when the tip contact force was varied, confirming that the time-resolved response of the van der Waals Schottky junction is measured reproducibly under the present experimental conditions (Supplementary Fig. S3).

Representative OPP-TR-AFM measurements are shown in Fig. 2a, b. The time-resolved current exhibits pronounced dependence on both sample bias and excitation fluence. Figure 2a shows the bias dependence measured at an excitation fluence of $3.4 \mu\text{J cm}^{-2}$. At low bias ($V_s = +0.3 \text{ V}$), only a negative response is observed. As the bias increases, a positive component emerges, and at $V_s = +0.6 \text{ V}$, both positive and negative components coexist. At higher biases (+1.0 and +1.5 V), the positive component becomes dominant. To further characterize this behavior, we also analyzed the bias dependences of the illuminated DC current (average current), measured without delay-time modulation, and the time-resolved current at zero delay, $\Delta I_L(t_d = 0)$ (Supplementary Fig. S4).

Figure 2b shows the excitation-fluence dependence measured at $V_s = +0.3 \text{ V}$. At the maximum excitation fluence (100%), a large negative response is observed. As the excitation fluence decreases, however, the negative component is rapidly suppressed and a positive component appears instead. The positive component reaches its maximum at 50% excitation fluence and decreases again at 10%. These observations indicate that the measured time-resolved current originates from the competition between two processes with distinct excitation-fluence dependences, and

that their relative contributions are strongly governed by the photoexcited carrier density. Furthermore, the excitation-fluence dependence of the photocurrent obtained from the light-modulated I – V measurements was also analyzed (Supplementary Fig. S5), providing complementary information on the excitation-density dependence of the photoresponse.

The amplitudes and lifetimes obtained by exponential fitting are summarized in Fig. 2c, d, respectively. As shown in Fig. 2c, the amplitude of the positive component increases with sample bias and becomes dominant at higher bias, particularly under the 100% excitation condition. In contrast, the negative component is observed only at the maximum excitation fluence (100%) in the low-bias region and gradually decreases with increasing bias.

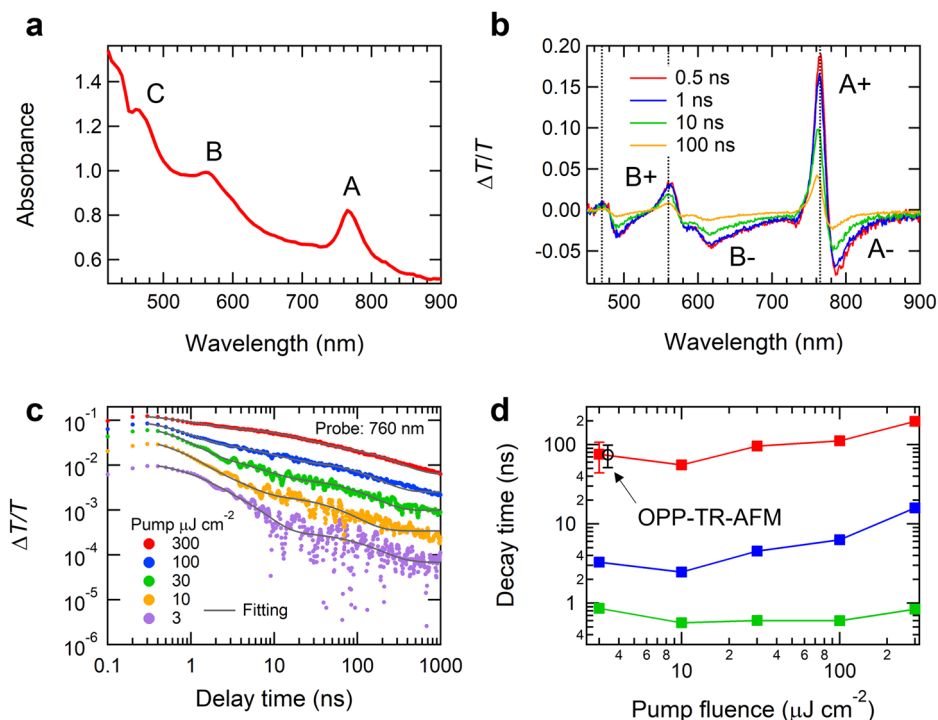
As shown in Fig. 2d, the lifetimes also exhibit distinct behaviors for the positive and negative components. The lifetime of the negative component decreases from $\sim 40 \text{ ns}$ at $+0.3 \text{ V}$ to $\sim 20 \text{ ns}$ at $+0.6 \text{ V}$ under the 100% excitation condition. In contrast, the positive component exhibits a lifetime of ~ 50 – 90 ns , and shows weak bias dependence particularly under the 10% and 50% excitation conditions.

These results indicate that the time-resolved current consists of two components with opposite polarities, each exhibiting distinct bias and excitation-fluence dependences. In the following section, we discuss the physical origins of these two time-resolved current components and how their competition determines the observed temporal response.

Nanosecond carrier dynamics in bulk WSe₂

To characterize the optical response of bulk WSe₂, TAS covering the picosecond-to-microsecond time range was performed using the Randomly Interleaved Pulse Train (RIPT) method¹⁷ (see Methods). Figure 3a shows the steady-state absorption spectrum of bulk WSe₂. The absorption features labeled A and B correspond to the A ($\sim 1.65 \text{ eV}$) and B ($\sim 2.1 \text{ eV}$) excitonic transitions at the K point, respectively, while the broad C band originates from higher-energy transitions in the band-nesting region²⁶ (Supplementary Fig. 6). The 488 nm pump pulse (2.54 eV) used in this study predominantly excites carriers through the C-band transition. Because the exciton binding energy in bulk WSe₂ is $\sim 30 \text{ meV}$, comparable to the thermal energy at room temperature, photoexcited excitons readily dissociate into

Fig. 3 | Photocarrier dynamics in bulk WSe₂ revealed by transient absorption spectroscopy. **a** Steady-state absorption spectrum of bulk WSe₂. The A and B peaks correspond to the excitonic transitions at the K point, while the broad C band originates from higher-energy transitions in the band-nesting region. **b** Representative differential transmission spectra $\Delta T/T$ of bulk WSe₂ measured at selected delay times under 488 nm excitation (pump fluence: 300 $\mu\text{J cm}^{-2}$). **c** Delay-time dependence of $\Delta T/T$ measured at 760 nm (A') for different pump fluences. Solid lines represent fits using a phenomenological triple-exponential function. **d** Pump-fluence dependence of the decay times extracted from the triple-exponential fitting. Open symbols indicate the lifetime of the positive component obtained from the OPP-TR-AFM measurements at 3.4 $\mu\text{J cm}^{-2}$ (Fig. 2d).



free carriers^{27,28}. Consequently, the nanosecond dynamics discussed in this study are expected to be governed predominantly by free carriers.

Representative differential transmission $\Delta T/T$ spectra are shown in Fig. 3b. Positive transient features (A' and B') are observed near the A and B excitonic transitions, accompanied by corresponding negative sidebands (A- and B-) on their lower-energy sides. Similar spectral features have previously been reported in transient absorption studies of bulk TMDCs, primarily over the sub-picosecond to nanosecond time range²⁹. By employing the RIPT method, the present measurements extend these observations to the previously unexplored nanosecond-to-microsecond regime. The A' and B' features, together with their corresponding sidebands, evolve continuously with delay time. Such spectral evolution has been attributed to energy shifts and linewidth changes of the excitonic absorption arising from interactions between photoexcited free carriers and excitonic resonances³⁰.

At longer delay times, however, the transient spectra may also contain contributions from photoinduced heating. To evaluate this effect, we analyzed the energy shift and linewidth broadening of the excitonic resonance based on the transient absorption spectra (Supplementary Fig. 7). The analysis suggests that nonthermal interactions between the remaining photoexcited carriers and the excitonic resonances play a dominant role in the transient spectral evolution under the low-excitation conditions corresponding most closely to those employed in the OPP-TR-AFM measurements. Accordingly, the relaxation dynamics obtained by TAS were used as a reference for the temporal evolution of bulk photocarriers when compared with the time-resolved current measured by OPP-TR-AFM.

Figure 3c shows the delay-time dependence of $\Delta T/T$ at 760 nm (A'). The transient signal persists over several tens to hundreds of nanoseconds. To quantify the observed dynamics, the data were fitted using a phenomenological triple-exponential function. The decay times extracted from the fitting are summarized in Fig. 3d for each excitation fluence. At the lowest excitation fluence of 3 $\mu\text{J cm}^{-2}$, comparable to that used in the OPP-TR-AFM measurements, the extracted decay times were $\tau_1 \approx 0.9$ ns, $\tau_2 \approx 3.3$ ns, and $\tau_3 \approx 75.9 \pm 30$ ns (Supplementary Fig. 6). For comparison, the lifetimes obtained from the OPP-TR-AFM measurements are also plotted as open symbols. The lifetime of the positive time-resolved response observed by OPP-TR-AFM falls within the same tens-of-nanoseconds time range as the

longest-lived component obtained from TAS. This correspondence in timescales supports the interpretation that the positive OPP-TR-AFM response is closely associated with long-lived photocarriers generated in the bulk WSe₂ and subsequently diffusing toward the interface. It should also be noted that the $\Delta T/T$ signal is nearly zero at the excitation wavelength of 488 nm used for the OPP-TR-AFM measurements, indicating that the OPP-TR-AFM signal cannot be explained simply by changes in optical absorption. Taken together, these results support the interpretation that the OPP-TR-AFM signal reflects the coupled dynamics of long-lived carrier transport in the bulk and the response of the local Schottky junction.

Origin of the positive and negative time-resolved current responses

To understand the origin of the positive and negative time-resolved current responses observed in the OPP-TR-AFM measurements, we constructed the carrier transport model summarized in Fig. 4. We propose that the time-resolved response in the reverse-bias regime can be understood as the competition between the hole diffusion current I_{hole} , arising from photo-generated holes diffusing through the bulk, and the thermionic emission current I_{th} , modulated by photoinduced changes in the interfacial potential.

We first consider I_{hole} . Based on the light-modulated I - V measurements, the WSe₂ surface is treated as an effectively n -type semiconductor with a dark electron density of N_{dark} . Under backside illumination, photo-excited carriers are generated predominantly within the bulk WSe₂ region and subsequently diffuse toward the WSe₂/PtIr interface. As shown in Fig. 3d, bulk WSe₂ exhibits a long-lived optical response extending over several tens of nanoseconds. To describe the relaxation dynamics of bulk photocarriers, we employ the Shockley-Read-Hall (SRH) recombination model^{31,32}.

As illustrated in Fig. 4a, b, the effective hole lifetime is strongly influenced by the occupancy of trap states in the bulk. When the photocarrier densities (ΔN_e , ΔN_p) are lower than N_{dark} (Fig. 4a), most trap states remain occupied by electrons, allowing trap-assisted hole recombination to proceed efficiently and resulting in a relatively short effective hole lifetime. In contrast, when ΔN_e and ΔN_p exceed N_{dark} (Fig. 4b), the electron occupancy of the trap states decreases, reducing the probability of hole capture and

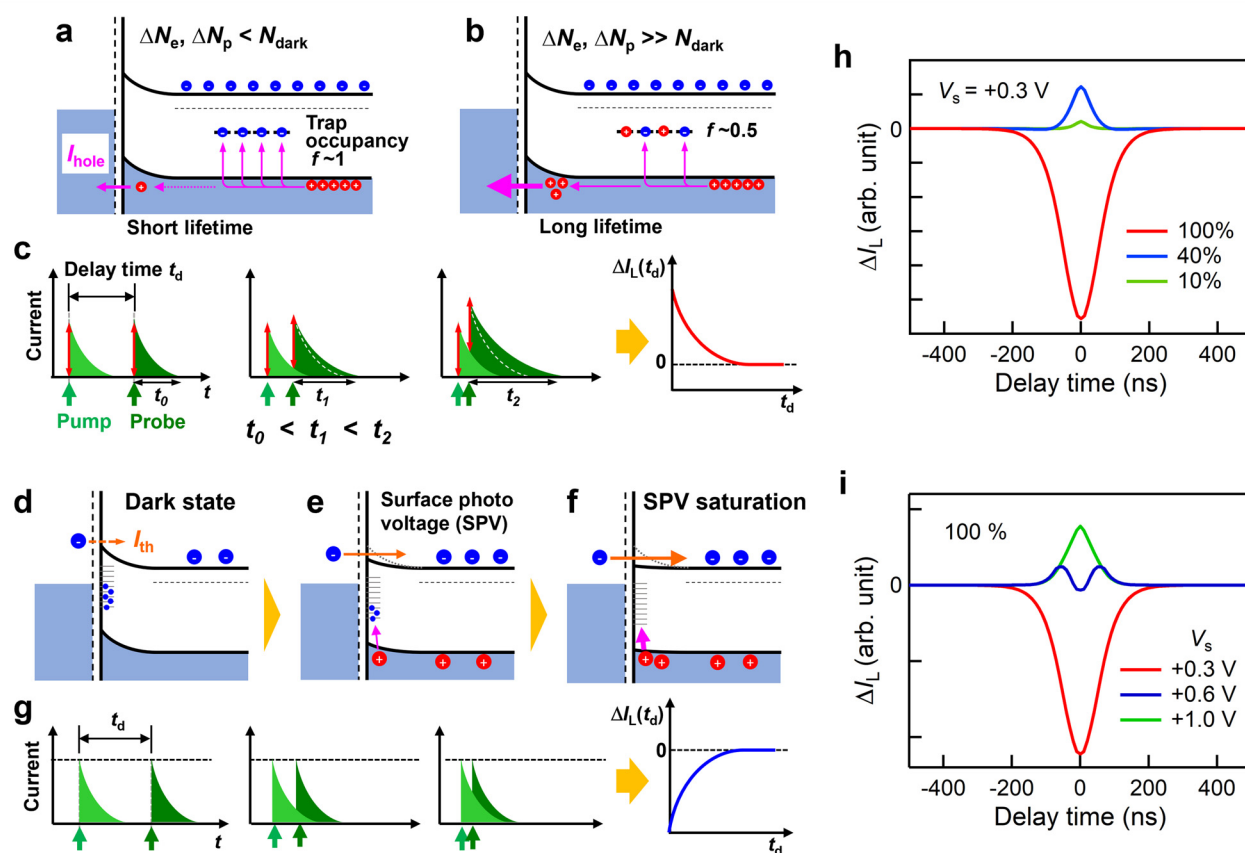


Fig. 4 | Physical model and numerical simulations of the time-resolved current responses in the WSe₂/PtIr junction. **a, b** Schematic illustration of the mechanism responsible for I_{hole} . Changes in the occupancy of bulk trap states modify the effective hole lifetime, resulting in an increase or decrease in I_{hole} . Here, ΔN_e and ΔN_p denote the photoexcited electron and hole densities, respectively, and N_{dark} is the equilibrium electron density in the dark. **c** Schematic pump–probe response corresponding to I_{hole} , giving rise to the positive time-resolved signal. **d–f** Schematic illustration of the photoinduced modulation mechanism of I_{th} . Charge

accumulation near the interface gives rise to SPV, thereby modulating the effective Schottky barrier. The simulated time evolution of the photoinduced interface potential is shown in Supplementary Fig. S10. **g** Schematic pump–probe response corresponding to the photoinduced modulation of I_{th} , giving rise to the negative time-resolved signal. **h** Simulated excitation-fluence dependence of the time-resolved current at $V_s = +0.3$ V. **i** Simulated bias-voltage dependence of the time-resolved current at a fixed excitation fluence.

thereby increasing the effective hole lifetime. Consequently, a larger fraction of the photogenerated holes reaches the interface and contributes to I_{hole} . To further support this picture, we calculated the hole lifetime based on the SRH model by explicitly considering the occupancy of trap states³³ (Supplementary Fig. S8). The calculated effective hole lifetime increases with increasing photocarrier density ($\Delta N_p = \Delta N_e$), consistent with the proposed SRH-based description of the hole diffusion current.

The corresponding pump–probe response is illustrated in Fig. 4c. At short delay times, the pump pulse generates relatively high photocarrier densities ($\Delta N_e, \Delta N_p$), corresponding to the situation shown in Fig. 4b, resulting in a large hole diffusion current induced by the probe pulse. As the delay time increases, the photocarrier density gradually decreases, and the system evolves toward the condition shown in Fig. 4a. Consequently, the effective hole lifetime becomes shorter, and the hole diffusion current decays with time. This behavior appears experimentally as the positive time-resolved component.

We next consider I_{th} , arising from photoinduced modulation of the effective Schottky barrier. In the dark, the reverse-bias current is dominated by thermionic emission across the WSe₂/PtIr Schottky junction (Fig. 4d). Upon photoexcitation, photogenerated charges are transiently accumulated in interfacial localized states associated with the local WSe₂/PtIr contact. The accumulated charges partially screen the built-in electric field, thereby generating an SPV (Fig. 4e). This modifies the effective Schottky barrier and

thereby changes I_{th} . At sufficiently high excitation densities, the accumulation of interfacial charge approaches saturation (Fig. 4f).

The corresponding pump–probe response is illustrated in Fig. 4g. At short delay times, a large population of pump-generated carriers remains near the interface, resulting in a high occupancy of the interfacial states. Consequently, only a small amount of additional interfacial charge is accumulated by the probe pulse, leading to a small photoinduced modulation of I_{th} . As the delay time increases, the pump-generated carriers gradually relax, allowing the probe pulse to induce a larger accumulation of interfacial charge and, consequently, a larger photoinduced modulation of I_{th} . Therefore, the time-resolved current measured at short delay times is smaller than that at the reference delay time t_{max} , giving rise to a negative $\Delta I_L(t_D)$. As the delay time increases, $I_L(t_d = t_D)$ gradually approaches the reference value $I_L(t_d = t_{\text{max}})$, and $\Delta I_L(t_D)$ correspondingly recovers toward zero.

Numerical simulations based on these two competing processes successfully reproduce the principal experimental observations (see the Supplementary Information for details). Figure 4h shows the excitation-fluence dependence at $V_s = +0.3$ V. At high excitation fluences, the negative time-resolved response dominates. As the excitation fluence decreases, however, the negative component is progressively suppressed, while the positive component emerges, in good agreement with the experimental results shown in Fig. 2b.

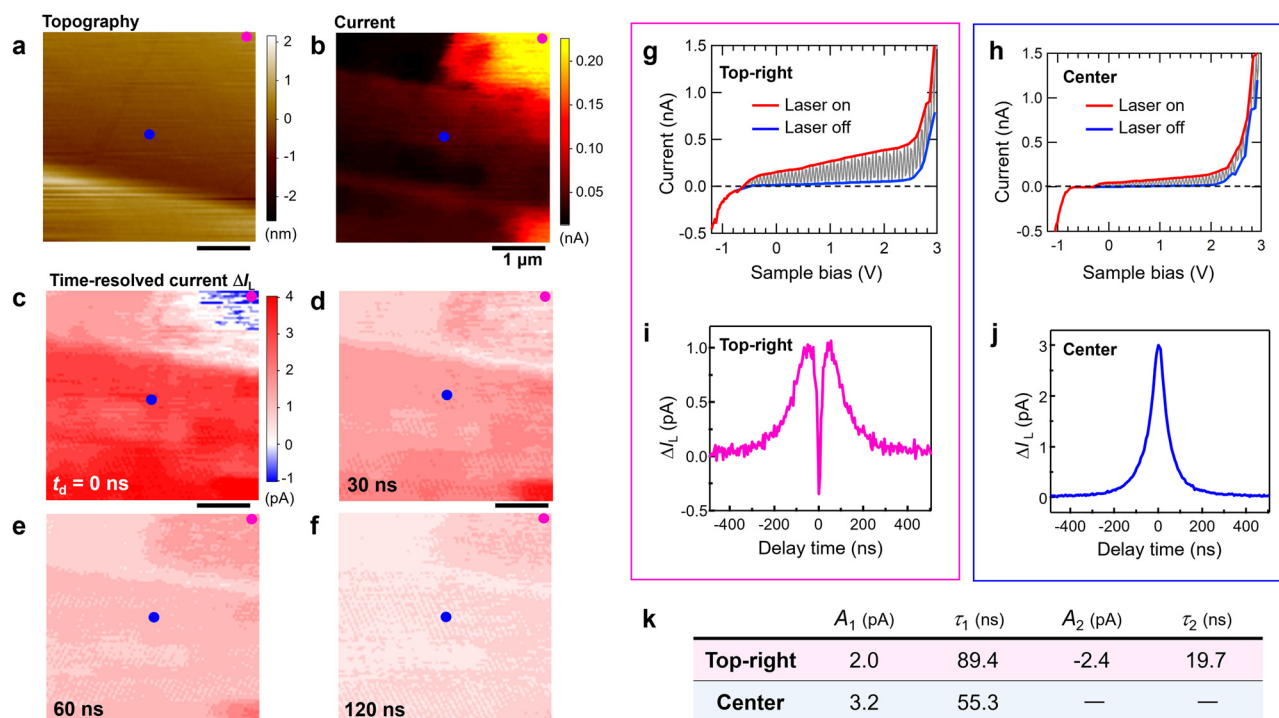


Fig. 5 | Spatially resolved time-resolved currents and local photocurrent dynamics in the WSe₂/PtIr junction. **a** Contact-mode AFM image (4 $\mu\text{m} \times 4 \mu\text{m}$, setpoint force: 16.8 nN). **b** Simultaneously acquired DC current map at $V_s = +0.6$ V. The circles indicate the locations where the light-modulated I - V characteristics and time-resolved currents were measured. **c–f** Spatial maps of the time-resolved current acquired at delay times of $t_d = 0, 30, 60$, and 120 ns ($V_s = +0.6$ V). **g, h** Light-modulated I - V characteristics measured at the blue and magenta locations,

respectively (excitation fluence: $6.7 \mu\text{J cm}^{-2}$). **i, j** Delay-time dependences of the time-resolved current measured at the magenta and blue locations, respectively ($V_s = +0.6$ V, setpoint force: 14.4 nN). **k** Amplitudes A_1, A_2 and lifetimes τ_1, τ_2 obtained by exponential fitting of the time-resolved currents shown in (**i**) and (**j**). The response at the magenta location was fitted with two components of opposite polarity, whereas that at the blue location was fitted with a single positive component.

Figure 4i shows the bias dependence at a fixed excitation fluence. This behavior can be understood in terms of the relaxation dynamics of the interfacial charge population. In the model, increasing the reverse bias shortens the relaxation time of the photoinduced interfacial potential (Supplementary Eq. (S10) in the Supplementary Information), leading to more rapid recovery of the effective Schottky barrier. Consequently, the negative time-resolved response originating from photoinduced modulation of I_{th} is reduced, whereas the relative contribution of the positive response arising from I_{hole} increases. As a result, the simulated response gradually changes from a negative signal at low bias to a positive response at high bias, reproducing the experimentally observed behavior.

In this model, I_{hole} is the primary origin of the positive time-resolved response, whereas I_{th} is primarily responsible for the negative response. However, because I_{th} is modulated by the SPV dynamics associated with hole trapping and detrapping near the interface, it also contains a small component originating from the hole diffusion process (Supplementary Fig. S9).

These results demonstrate that the characteristic bias and excitation-fluence dependences observed by OPP-TR-AFM arise from the competition between I_{hole} , driven by long-lived bulk photocarriers, and I_{th} , modulated by the interfacial potential. The present model provides a physical framework linking bulk photocarrier transport with local interfacial barrier dynamics in van der Waals Schottky junctions.

Spatial mapping of the time-resolved current

A key advantage of OPP-TR-AFM is its ability to simultaneously achieve nanometer-scale spatial resolution and nanosecond temporal resolution. To demonstrate this capability, we performed spatial mapping of the time-resolved current over a $4 \mu\text{m} \times 4 \mu\text{m}$ region on the WSe₂ surface.

Figure 5a, b shows the contact-mode AFM image and the simultaneously acquired DC current map at a sample bias of $V_s = +0.6$ V,

respectively. While the surface topography is relatively uniform over the entire measurement area, the DC current exhibits pronounced spatial inhomogeneity, indicating that the local junction characteristics vary across the sample.

Figure 5c–f presents spatial maps of the time-resolved current acquired at delay times of 0, 30, 60, and 120 ns. At $t_d = 0$ ns, both positive and negative time-resolved responses coexist within the same field of view. This observation indicates that the relative contributions of I_{hole} and the photoinduced modulation of I_{th} vary locally depending on the junction characteristics.

To investigate the origin of this spatial inhomogeneity, light-modulated I - V measurements were performed at the representative locations indicated by the magenta and blue circles in Fig. 5a. The corresponding results are shown in Fig. 5g, h. The illuminated I - V characteristics differ substantially between the two locations. In particular, the magenta position exhibits a larger photoinduced shift of the I - V characteristics than the blue position, together with a shift of the forward current onset toward lower bias. These observations suggest that photoinduced Schottky barrier modulation contributes more strongly at the magenta position.

The corresponding time-resolved current responses are shown in Fig. 5i, j. At the magenta position, both positive and negative components are observed, whereas only a positive time-resolved response appears at the blue position. These results suggest that the photoinduced modulation of I_{th} makes a larger contribution at the magenta position, while I_{hole} dominates at the blue position. The fitting analysis (Fig. 5k) yields long-lived components with lifetimes of ~ 55 – 90 ns, in good agreement with the longest-lived optical response observed by TAS. This correspondence further supports the interpretation presented in the previous section that the positive time-resolved response originates from long-lived bulk photocarriers.

Importantly, the spatial inhomogeneity observed in this study reflects the coupled dynamics of bulk photocarriers and the local interfacial response. Photocarriers generated within the bulk WSe₂ by backside

illumination subsequently diffuse toward the WSe₂/PtIr interface, where they interact with the local Schottky junction. Therefore, the observed spatial variations are likely to arise from multiple factors, including spatial variations in the effective Schottky barrier height, the density of interfacial localized states, local variations in bulk carrier transport associated with trap-assisted recombination. Further investigation will be required to distinguish the relative contributions of these factors. These results directly reveal that the coupling between bulk photocarrier transport and the local interfacial response varies substantially across the junction on the nanoscale.

Using OPP-TR-AFM in combination with light-modulated *I*–*V* measurements, transient absorption spectroscopy, and numerical simulations, we elucidated the mechanism underlying the photoinduced time-resolved current in a WSe₂/PtIr point-contact van der Waals Schottky junction. The observed response is explained by the competition between the diffusion of long-lived bulk photocarriers toward the interface and the photoinduced modulation of thermionic emission through transient changes in the effective Schottky barrier. Furthermore, spatially resolved measurements revealed pronounced nanoscale inhomogeneity in the time-resolved current response, revealing that the interplay between bulk photocarrier transport and local interfacial dynamics varies locally across the junction.

These findings demonstrate that OPP-TR-AFM provides a unique approach for investigating the interplay between bulk carrier transport and local interfacial dynamics with simultaneous nanometer-scale spatial resolution and nanosecond temporal resolution. Beyond TMDC-based optoelectronic devices, this technique is expected to be broadly applicable to a wide range of van der Waals heterostructures and low-dimensional semiconductor junctions, providing new opportunities to elucidate nanoscale carrier transport and interfacial dynamics and to guide the design and optimization of next-generation optoelectronic and energy-harvesting devices.

Methods

Optical pump-probe time-resolved current

The optical pump-probe delay time is denoted by t_d . During the measurement, a specific delay time $t_d = t_D$ is selected, and the corresponding photoinduced transient current $I_L^*(t_D)$ persists until the pump-probe excited state has completely relaxed. Accordingly, the measured time-resolved signal $I_L(t_D)$ is given by the cycle-averaged transient current over one laser repetition period τ_R ,

$$I_L(t_D) = \frac{1}{\tau_R} \int_0^{\tau_R} I_L^*(t_d = t_D) dt \quad (1)$$

To detect the small current variation with high sensitivity, lock-in detection was employed. Furthermore, to minimize the influence of photothermal expansion of both the tip and the sample, the delay-time modulation method was used²⁵. In this approach, instead of modulating the pump or probe pulses themselves as the lock-in reference, the pump-probe delay is alternately switched between the delay of interest $t_d = t_D$ and a sufficiently long delay $t_d = t_{\max}$. The lock-in signal is then obtained as the differential current between these two delay conditions,

$$\Delta I_L(t_D) = I_L(t_d = t_D) - I_L(t_d = t_{\max}) \quad (2)$$

This method enables highly sensitive extraction of the temporal response of the Schottky junction while minimizing thermal artifacts arising from laser irradiation.

Optical system for OPP-TR-AFM

The laser pulses were precisely synchronized using electronic triggering, enabling a simple configuration that does not require a complex optical setup (OPP-NS, Unisoku)^{34,35}. In this study, we focused on nanosecond dynamics associated with the Schottky barrier and localized interfacial states.

Accordingly, two nanosecond pulsed lasers (NPL49B, Thorlabs) were used as the pump and probe sources. Both lasers operated at a wavelength of 488 nm with a pulse duration of ~6 ns and a repetition rate of 1 MHz. The timing jitter between the two lasers relative to the electronic trigger was ~20 ps, resulting in an estimated overall temporal resolution of ~8.5 ns. The system was integrated with a commercial AFM operated in air (Dimension XR Icon, Bruker) to enable its application to a wide range of materials and devices. All measurements were performed at room temperature in air using a PtIr-coated conductive cantilever (PF-TUNA, Bruker). For the light-modulated current-voltage measurements, the illumination was periodically switched on and off using an optical chopper.

Sample preparation

To enable backside optical excitation, a bulk WSe₂ single crystal (HQ Graphene) was mounted on a conductive ITO-coated glass substrate using a transparent insulating epoxy adhesive (FH5313PK, Thorlabs) (Fig. 1a). The WSe₂ surface was prepared by mechanical exfoliation, and its atomically flat surface was confirmed by STM (Supplementary Fig. 1). To establish an electrical connection between the WSe₂ crystal and the ITO electrode, conductive silver epoxy (EPO-TEK H20E) was applied to the edge of the crystal.

During the measurements, the sample was placed on the glass plate of the AFM stage and electrically contacted through a magnetic contact pin, which also served as the sample-bias electrode (Fig. 1a). In this configuration, the illuminated region of the WSe₂ crystal was electrically isolated from the ITO substrate, and the WSe₂ crystal was connected to the ITO electrode only through the conductive silver epoxy located outside the illuminated region.

The measurement geometry contained two metal–WSe₂ junctions: the PtIr-coated AFM cantilever–WSe₂ junction and the silver-epoxy–WSe₂ junction. However, as demonstrated in previous studies¹⁹, the measured current is dominated by the local junction directly beneath the AFM tip, owing to the orders-of-magnitude smaller contact area of the PtIr/WSe₂ junction compared with the silver-epoxy/WSe₂ contact.

Picosecond-to-microsecond transient absorption spectroscopy (ps-TAS)

In conventional transient absorption spectroscopy, the time window from ~1 ns to several tens of nanoseconds has been a “blind gap,” largely because it is difficult to continuously vary the pump-probe delay within this regime using standard delay-line optics. To overcome this limitation, we utilized a commercially available picosecond transient absorption system, picoTAS (Unisoku), based on the RIPT method¹⁷. This technique, which we previously developed, enables asynchronous and randomized sampling of pump-probe delays using a picosecond supercontinuum laser as the probe light, thereby providing access to this otherwise inaccessible temporal region. For the pump pulses, we used a 1 kHz, 25 ps mode-locked picosecond laser equipped with a tunable wavelength unit (PL2210A and PG403, EKSPLA). The pump wavelength was set to 488 nm to allow direct comparison with the OPP-TR-AFM measurements. As the probe source, we used a 20 MHz, 50–100 ps supercontinuum laser (SMHP-20.2-A, Leukos), spectrally selected by a diffraction-grating monochromator to cover the wavelength range from 450 to 900 nm. Temporal resolution of the system was better than 100 ps and was primarily determined by the pulse width of the probe light. All ps-TAS measurements were performed at room temperature under ambient conditions.

Data availability

The datasets generated and/or analyzed during the current study are not publicly available because they are too large to be deposited in a public repository but are available from the corresponding author upon reasonable request.

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References

- Xia, F. et al. Two-dimensional material nanophotonics. *Nat. Photon* **8**, 899–907 (2014).
- Koppens, F. H. L. et al. Photodetectors based on graphene, other two-dimensional materials and hybrid systems. *Nat. Nanotechnol.* **9**, 780–793 (2014).
- Fiori, G. et al. Electronics based on two-dimensional materials. *Nat. Nanotechnol.* **9**, 768–779 (2014).
- Lopez-Sanchez, O. et al. Ultrasensitive photodetectors based on monolayer MoS₂. *Nat. Nanotechnol.* **8**, 497–501 (2013).
- Liu, X. et al. Fermi Level Pinning Dependent 2D Semiconductor Devices: Challenges and Prospects. *Adv. Mater.* **34**, 2108425 (2022).
- Zhang, Y. et al. High-Speed Transition-Metal Dichalcogenides Based Schottky Photodiodes for Visible and Infrared Light Communication. *ACS Nano* **16**, 19187–19198 (2022).
- Strauß, F. et al. A simple 230 MHz photodetector based on exfoliated WSe₂ multilayers. *RSC Appl. Interfaces* **1**, 728–733 (2024).
- Zhang, M. et al. Schottky-Contacted WSe₂ Hot-Electron Photodetectors with Fast Response and High Sensitivity. *ACS Photonics* **9**, 132–137 (2022).
- Kong, L. et al. Doping-free complementary WSe₂ circuit via van der Waals metal integration. *Nat. Commun.* **11**, 1866 (2020).
- Keller, K. R. et al. Ultrafast Thermionic Electron Injection Effects on Exciton Formation Dynamics at a van der Waals Semiconductor/Metal Interface. *ACS Photonics* **9**, 26830–2690 (2022).
- Wang, Y. et al. Van der Waals contacts between three-dimensional metals and two-dimensional semiconductors. *Nature* **568**, 70–74 (2019).
- Allain, A., Kang, J., Banerjee, K. & Kis, A. Electrical contacts to two-dimensional semiconductors. *Nat. Mater.* **14**, 1195–1205 (2015).
- Chen, D. et al. Optical spectroscopic detection of Schottky barrier height at a two-dimensional transition-metal dichalcogenide/metal interface. *Nanoscale* **16**, 5169–5176 (2024).
- Tao, Y., Hong, C. & Kim, J. Ultrafast reversible photoconductivity in 2D MoTe₂/Pt van der Waals heterostructure. *Sci. Adv.* **11**, eady1321 (2025).
- Hong, C. et al. Ultrafast hot carrier extraction and diffusion at the MoS₂/Au van der Waals electrode interface. *Sci. Adv.* **11**, eadr1534 (2025).
- Gurarslan, A. et al. Van der Waals Force Isolation of Monolayer MoS₂. *Adv. Mater.* **28**, 10055–10060 (2016).
- Nakagawa, T., Okamoto, K., Hanada, H. & Katoh, R. Probing with randomly interleaved pulse train bridges the gap between ultrafast pump-probe and nanosecond flash photolysis. *Opt. Lett.* **41**, 1498–1501 (2016).
- Takeuchi, O., Yoshida, S. & Shigekawa, H. Light-modulated scanning tunneling spectroscopy for nanoscale imaging of surface photovoltage. *Appl. Phys. Lett.* **84**, 3645–3647 (2004).
- Giannazzo, F. et al. Nanoscale inhomogeneity of the Schottky barrier and resistivity in MoS₂ multilayers. *Phys. Rev. B* **92**, 081307R (2015).
- Bampoulis, P. et al. Defect Dominated Charge Transport and Fermi Level Pinning in MoS₂/Metal Contacts. *ACS Appl. Mater. Interfaces* **9**, 19278 (2017).
- Sothawes, K. et al. Universal Fermi-Level Pinning in Transition-Metal Dichalcogenides. *J. Phys. Chem. C* **123**, 5411 (2019).
- Fan, Y. et al. Photoinduced Schottky Barrier Lowering in 2D Monolayer WS₂ Photodetectors. *Adv. Opt. Mater.* **4**, 1573–1581 (2016).
- Hamers, R. J. & Cahill, D. G. Ultrafast time resolution in scanned probe microscopies: Surface photovoltage on Si(111)-(7×7). *J. Vac. Sci. Technol. B* **9**, 514–518 (1991).
- Kumar, J. & Shrivastava, M. Role of Chalcogen Defect Introducing Metal-Induced Gap States and Its Implications for Metal-TMDs' Interface Chemistry. *ACS Omega* **8**, 10176–10184 (2023).
- Terada, Y., Yoshida, S., Takeuchi, O. & Shigekawa, H. Real-space imaging of transient carrier dynamics by nanoscale pump-probe microscopy. *Nat. Photon* **4**, 869–874 (2010).
- Mishra, H., Bose, A., Dhar, A. & Bhattacharya, S. Exciton-phonon coupling and band-gap renormalization in monolayer WSe₂. *Phys. Rev. B* **98**, 045143 (2018).
- Schuster, R., Wan, Y., Knupfer, M. & Büchner, B. Nongeneric dispersion of excitons in the bulk of WSe₂. *Phys. Rev. B* **94**, 085201 (2016).
- Völzer, T., Lütgens, M., Fennel, F. & Lochbrunner, S. Recombination Dynamics of Optically Excited Charge Carriers in Bulk MoS₂. *J. Phys. B At. Mol. Opt. Phys.* **50**, 194003 (2017).
- Chen, X. et al. Investigating the dynamics of excitons in monolayer WSe₂ before and after organic super acid treatment. *Nanoscale* **10**, 9346–9352 (2018).
- Fluence-dependent dynamics of localized excited species in monolayer versus bulk MoS₂. *Phys. Rev. B* **103**, 045423 (2021).
- Shockley, W. & Read, W. T. Jr Statistics of the Recombinations of Holes and Electrons. *Phys. Rev.* **87**, 835 (1952).
- Hall, R. N. Germanium rectifier characteristics. *Phys. Rev.* **83**, 228 (1951).
- Liraz, D., Cheng, P., Yang, Y. & Tessler, N. Light-induced trap emptying revealed by intensity-dependent quantum efficiency of organic solar cells. *J. Appl. Phys.* **131**, 135501 (2022).
- Mogi, H. et al. Externally triggerable optical pump-probe scanning tunneling microscopy. *Appl. Phys. Express* **12**, 025005 (2019).
- Iwaya, K. et al. Externally-triggerable optical pump-probe scanning tunneling microscopy with a time resolution of tens-picosecond. *Sci. Rep.* **13**, 818 (2023).

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Author contributions

OPP-TR-AFM experiments were performed by M.Y., K.I., and T.M., while the ps-TAS measurements were carried out by T.N. S.Y. and O.T. provided technical advice on sample preparation and experimental methodologies. The mechanism of photoinduced dynamics at the Schottky junction was primarily conceived by H.M., Y.M., M.Y., and H.S., and was further established through combined investigations of time-resolved signal simulations by Y.M. and analyses of I–V characteristics by H.M. Based on these experimental and theoretical results, all authors discussed the details and jointly prepared the manuscript. The project was organized and supervised by K.I. and H.S., who coordinated the overall research effort.

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Competing interests

The authors declare no competing interests.

Additional information

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