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Low-threshold interlayer exciton multiplication in twisted transition metal dichalcogenides heterobilayers

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Abstract

Multiple exciton generation (MEG) from single-photon absorption can enhance quantum efficiencies in light emission and energy conversion. However, its practical application is limited by high photon-energy thresholds, large material bandgaps, and rapid exciton recombination. Here, we report the observation of interlayer exciton multiplication (IXM) in twisted van der Waals heterobilayers, where the process threshold is as low as twice the type-II bandgap and the resulting multiple interlayer excitons (IXs) exhibit nanosecond-scale lifetimes. These properties directly address the core challenges of MEG, leading to a significant boost in both interlayer exciton emission and photocurrent quantum efficiency. Theoretical calculations confirm the experimental results, revealing that low-threshold IXM is facilitated by interlayer hot-carrier scattering and is accompanied by attractive IX interactions, even at larger twist angles where momentum mismatch is significant. Twist-angle-induced momentum mismatches have a minimal impact on the IXM threshold due to strong hot-carrier interlayer transfer and efficient intervalley impact ionization. The IXM efficiency reaches ~90% in small-twist-angle heterobilayers but decreases at larger angles and higher pump photon energies, a trend attributed to reduced interlayer hot-carrier Coulomb scattering. Furthermore, we demonstrate photocurrent multiplication driven by IXM. Applied electric fields further lower the IXM threshold and increase the IX yield, enabling a twofold enhancement in quantum efficiency and a fourfold increase in responsivity in a self-powered heterobilayer photodiode. Our results establish IXM as a promising mechanism for high-efficiency carrier-multiplication optoelectronics and provide insights into the many-body physics of stable multiple excitons.

Introduction

Single-photon-driven multiple exciton generation (MEG) is a process in which the absorption of a single high-energy photon generates two or more electron-hole pairs. Instead of dissipating its excess kinetic energy as heat, the initially excited “hot” carrier can promote an additional electron across the bandgap via impact ionization. MEG holds great promise for breaking the Shockley-Queisser limit by enhancing the quantum efficiency of optoelectronic devices^{1–4}. Ideally, the

threshold photon energy ($h\nu_{\text{th}}$) for MEG is twice the material bandgap (E_g). However, in practice, energy and momentum conservation constraints lead to low efficiencies and high thresholds ($h\nu_{\text{th}} \geq 4E_g$) for carrier multiplication in bulk semiconductors like Si and Ge^{5–7}. While enhanced MEG has been demonstrated in quantum-confined systems such as PbSe, PbS^{8–11}, Si¹², Ge¹³ nanocrystals (NCs), as well as in few-layered 2D MoTe₂^{14,15}, their utility is hampered by the ultrafast Auger recombination of multiple excitons (e.g., within around 5 ps in few-layer MoTe₂^{14,15}) and large E_g (e.g., halide perovskite NCs¹⁶). Consequently, the development of efficient MEG materials that combine a low threshold, a tunable narrow bandgap, and slow multiple exciton recombination remains an outstanding challenge.

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Two-dimensional van der Waals (vdW) heterobilayers, composed of different transition metal dichalcogenide (TMDC) monolayers, offer strong light-matter interactions and electrical and twist-angle tunability, enabling diverse phenomena in optoelectronics and quantum photonics^{17–21}. A key feature of type-II heterobilayers is the formation of interlayer excitons (IXs), where electrons and holes are spatially separated across the two layers and bound across the staggered band structure^{22–24}. This type-II alignment allows the interlayer bandgap ($E_{g(\text{type-II})}$) to be tuned from the visible range of the constituent monolayers into the near-infrared, with further electrical tunability^{25,26}. Despite this potential, single-photon-driven generation of multiple interlayer excitons and their subsequent interactions have not been explored in twisted 2D vdW heterobilayers.

In this work, we report the observation of low-threshold interlayer exciton multiplication (IXM) in twisted type-II $\text{MoS}_2/\text{WSe}_2$ heterobilayers, yielding long-lived multiple IXs. This process leads to enhanced interlayer exciton emission and photocurrent quantum efficiency at room temperature. We find an IXM threshold energy as low as twice $E_{g(\text{type-II})}$, approaching the fundamental energy conservation limit, which persists across all twist angles despite momentum mismatch. This is enabled by efficient hot-carrier interlayer transfer. Notably, the IXM efficiency reaches $\sim 90\%$ in small-twist-angle heterobilayers, where shorter attractive IX interaction distances facilitate the process, and gradually decreases with increasing twist angle due to reduced Coulomb interactions. Our experimental findings are well supported by theoretical modeling based on a tight-binding approach. Finally, we demonstrate IXM-driven photocurrent multiplication in a self-powered $\text{MoS}_2/\text{WSe}_2$ heterobilayer photodiode, achieving a threshold as low as $1.8E_{g(\text{type-II})}$.

Results

Interlayer exciton emission multiplication

Figure 1a illustrates a twisted $\text{MoS}_2/\text{WSe}_2$ heterobilayer, where IXs form with electrons in MoS_2 and holes in WSe_2 . The twist angle of each heterobilayer is determined using second-harmonic generation microscopy (see Supplementary Fig. 1 and “Methods” for details)^{27–29}. Raman spectroscopy confirms strong interlayer coupling (Supplementary Note 2 and Supplementary Fig. 2a), and significant photoluminescence (PL) quenching of the constituent monolayers indicates efficient charge separation driven by the type-II band alignment (Supplementary Fig. 2b).

The calculated momentum-space structure of the heterobilayer is shown in Fig. 1b. The electron and hole states at the K valleys are localized within their respective layers (left panel), forming the dominant K-K IXs. In

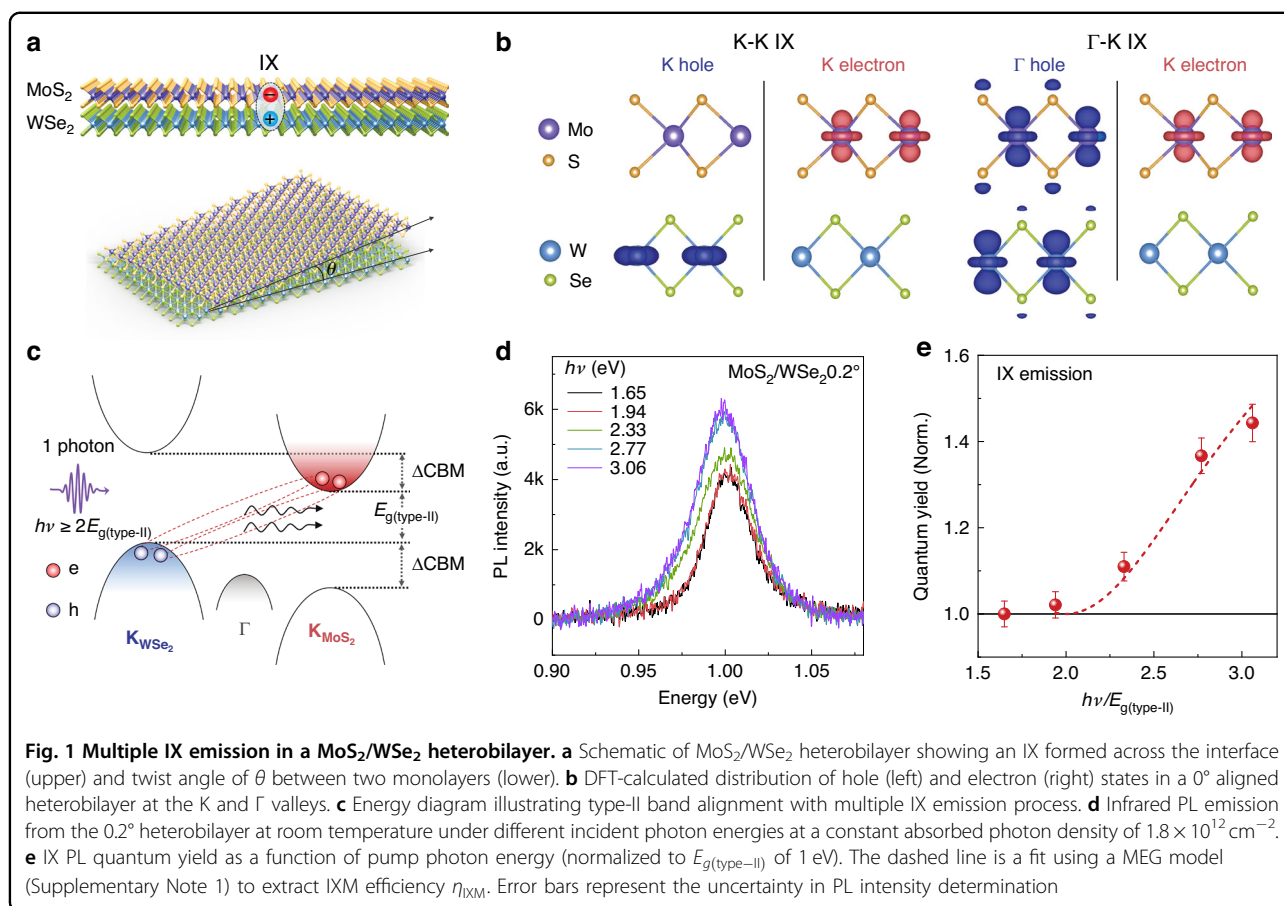
contrast, the hybrid hole state at the Γ point is distributed across both layers (right panel), giving rise to Γ -K IXs, consistent with previous reports^{30,31}. The twist angle has only a minor effect on these charge-state distributions (Supplementary Fig. 3 and Supplementary Table 1). Figure 1c shows the schematic band alignment with conduction band minimum (CBM) and valence band maximum (VBM) offsets of $\Delta\text{CBM} = 0.76$ eV and $\Delta\text{VBM} = 0.83$ eV (ref. ³²). From an energy conservation perspective, if the band-edge offset energy can be utilized as excess energy for impact ionization, it becomes possible to drive IXM when this energy exceeds $E_{g(\text{type-II})}$.

To study IXM, we first calibrated the absorbed photon density using steady-state absorption spectra of the monolayers and heterobilayers (Supplementary Note 2 and Supplementary Fig. 4). In the near-infrared region, we observe prominent emission around 1.0 eV from K-K IXs at room temperature in the 0.2° heterobilayer (Fig. 1d). This confirms a type-II bandgap of $E_{g(\text{type-II})} = 1.0 \pm 0.1$ eV, consistent with calculations based on monolayer bandgaps and the band offset. The PL intensity increases with pump photon energy. The PL quantum yield (QY) of the IX emission remains constant for $h\nu < 2E_{g(\text{type-II})}$ but shows a significant increase once this threshold is exceeded (Fig. 1e). This clear threshold behavior signals the onset of IXM, with a fitted IXM efficiency (η_{IXM}) of $90 \pm 5\%$ (see the dashed-line fitting model in Supplementary Note 1). In heterobilayers with near-zero twist angles, the close alignment of the monolayer Brillouin zones³³ allows IXs near the K-valley to exhibit nearly direct optical transitions, making them highly optically active.

Long-lived multiple IXs

To investigate the dynamics of multiple IXs and probe the IXM process, we used pump-probe transient absorption (TA) microscopy to monitor the population kinetics (Fig. 2a and Supplementary Fig. 5). The differential transmittance, $\Delta T/T_0$, is proportional to the total occupation of the electron and hole states involved in the probe beam transitions, $[\sum_i (n_i^e + n_i^h)]$. Figure 2b presents the 2D TA spectra for the 0.2° heterobilayer, revealing two distinct photobleaching (PB) bands at 1.81 eV and 1.61 eV, which correspond to the band-edge transitions of MoS_2 and WSe_2 , respectively (see Supplementary Fig. 6 for other twist angles). The persistence of these PB signals—with recombination lifetimes two orders of magnitude longer than those of intralayer excitons—confirms efficient charge separation and the formation of stable IXs^{23,34} (Fig. 2c; see Supplementary Note 3 and Supplementary Fig. 7). As illustrated in Fig. 2a, after photoexcitation and hot-carriers transfer, electrons reside in the K valley of MoS_2 for both K-K and K- Γ IX transitions, while holes occupy distinct valleys for these two types of





IXs. Consequently, probed at the WSe₂ band-edge transition specifically reflect the hole population of K–K IXs, and the dynamics probed at the MoS₂ reflect the electron population of K–K and K– Γ IXs.

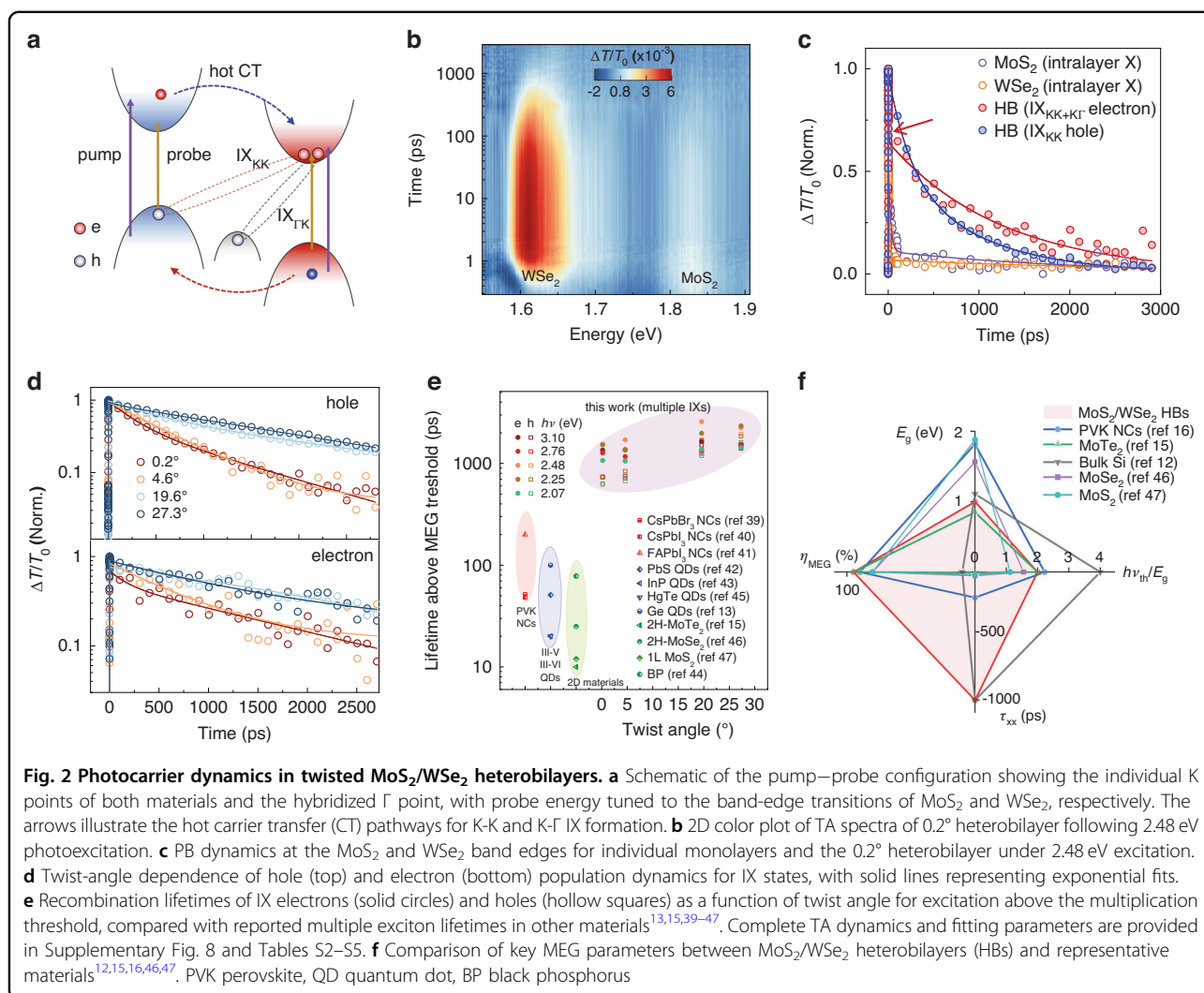
An additional fast decay component (<5 ps) is observed for IX electrons (Fig. 2c, indicated by the red arrow; see Supplementary Fig. 7c, d for sub-100 ps dynamics). This fast decay resembles intralayer exciton formation dynamics^{35,36} and is attributed to Coulomb interactions between hybrid holes at the Γ point and electrons at the K point of MoS₂ (Fig. 1b). The amplitude of this fast decay decreases with increasing twist angle (~47% at 0.2° vs. 37% at 27.3°), consistent with the calculated reduction in Γ -hole distributions at larger angles (Supplementary Table 1). The longer decay lifetime of electrons compared to holes arises from the mixed decay dynamics involving momentum-indirect K– Γ transitions. Furthermore, while the hole lifetimes of K–K IXs increase significantly with twist angle due to enhanced momentum mismatch, the electron lifetimes are less affected (Fig. 2d, e; Supplementary Note 3, Supplementary Fig. 8, and Supplementary Tables 2–5). Similar asymmetric electron and hole population dynamics have been observed in WS₂–WSe₂ heterobilayers, associated with K–K and K–Q IXs³⁷. The

clear trend of increasing IX recombination lifetimes with larger twist angles, which we attribute to greater parallel momentum mismatch³³ and expanded interlayer spacing³⁸.

Figure 2e summarizes the lifetimes of interlayer electrons and holes across various pump energies above IXM threshold. In all twisted heterobilayers, excitation above the $2E_{g(\text{type-II})}$ threshold produces long-lived multiple IXs with nanosecond-scale lifetimes. These lifetimes are one to two orders of magnitude longer than those of multiple intradot or intralayer excitons in previous MEG materials, such as perovskite NCs and other 2D systems^{13,15,39–47}. This enhanced lifetime stems from the significantly reduced electron-hole wavefunction overlap in interlayer excitons compared to their intralayer counterparts, as well as from long-range (nanometer-scale) dipole-dipole interactions among them, which will be elaborated in following sections.

Figure 2f compares key performance parameters across various MEG materials, including multiple exciton lifetimes above the MEG threshold, MEG efficiency (η_{MEG} , equivalent to η_{IXM} for our samples), bandgap (E_g , equivalent to $E_{g(\text{type-II})}$ for our samples), and the normalized threshold energy ($h\nu_{\text{th}}/E_g$). The theoretical





maximum η_{MEG} is 100%, corresponding to a step-like QY described by $\text{QY}_{\text{max}} = \lfloor (h\nu/E_g - 1) * \eta_{\text{MEG}} \rfloor + 1$ (ref. 48), where the floor operator ($\lfloor \rfloor$) denotes rounding down. A 100% η_{MEG} represents the ideal limit of no energy loss during multiple electron-hole pair generation. Bandgap values are included for context, as optimal values are application-dependent; for photovoltaics, MEG is most effective for bandgaps of $\sim 0.7\text{--}1.0$ eV (Supplementary Fig. 9 and Supplementary Note 4). Our MoS₂/WSe₂ heterobilayers uniquely combine high η_{IXM} , a tunable type-II bandgap, a low threshold, and—most notably—exceptionally long-lived multiple interlayer excitons. This combination of properties represents a significant advance in MEG performance and establishes a new benchmark for carrier multiplication materials.

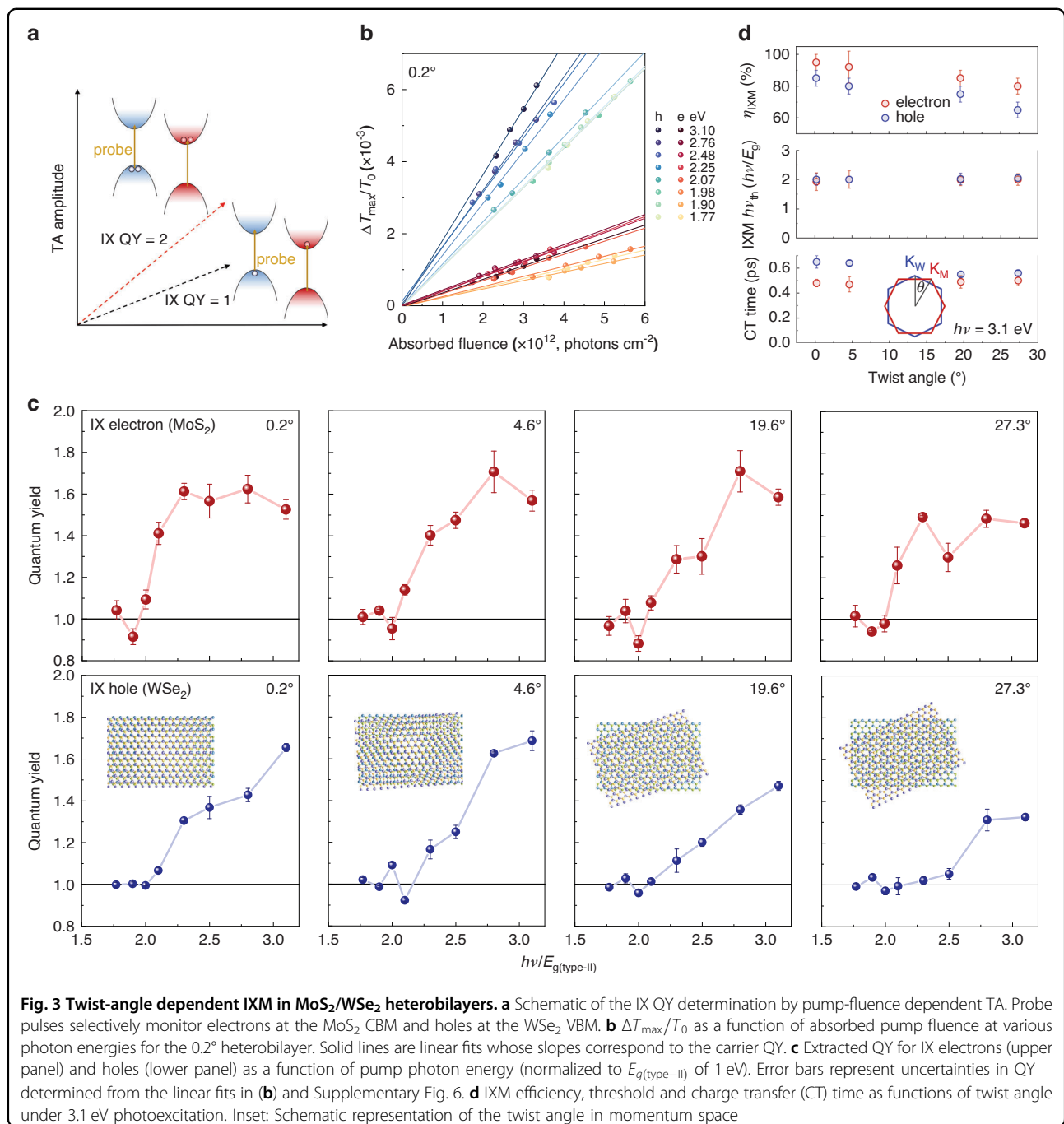
Evidence of IXM in twisted heterobilayers

Beyond emission multiplication, TA measurements provide further evidence and deeper insights into IXM

and its twist-angle dependence (Fig. 3a). To accurately determine the interlayer exciton quantum yield (IX QY), we performed pump-fluence-dependent TA. The peak PB signals ($\Delta T_{\text{max}}/T_0$) increase linearly with low excitation fluence across various photon energies (Fig. 3b; see Supplementary Figs. 6d–f and 10–17). This linearity confirms one-photon excitation without contributions from non-linear processes. The peak PB signal is given by $\Delta T_{\text{max}}/T_0 = \Phi \sigma_{\text{PB}} F_{\text{abs}}$, where Φ is IX generation QY per absorbed photon, σ_{PB} is the probe absorption cross-section (constant at a fixed energy), and F_{abs} is the absorbed pump photon fluence, calibrated from microscope absorption spectra (Supplementary Fig. 4). The IX QY is thus extracted from the slope of $\Delta T_{\text{max}}/T_0$ versus F_{abs} (Fig. 3a).

Analysis across multiple twist angles confirms IXM. As shown for the 0.2° sample in Fig. 3b, the slopes for both electron and hole states remain constant for $h\nu < 2E_{\text{g(type-II)}}$, consistent with an IX QY of 1 below the





MEG threshold. Above this energy, the slopes increase, indicating enhanced IX QY. Figure 3c presents the extracted QYs as a function of $h\nu/E_{g(\text{type-II})}$ for different twist angles, probed at the MoS₂ (electrons of K–K and K–Γ IXs) and WSe₂ (holes of K–K IXs), respectively. The QYs exceed unity at higher photon energies, confirming the generation of more than one carrier per absorbed photon in IX. The simultaneous observation of interlayer electron and hole multiplication (IEM and IHM) with a threshold near $2E_{g(\text{type-II})}$, combined with the long IX

lifetimes (Fig. 2e), provides definitive evidence for IXM. The extracted η_{IXM} values remain valid even at long delay times (e.g., 100 ps), as the subsequent decay dynamics are independent of the initial pump energy due to the long-lived nature of the IXs (Supplementary Fig. 8).

Figure 3d summarizes the averaged IEM and IHM efficiencies $\left(\bar{\eta}_{\text{IEM/IHM}} = \frac{\sum \eta(h\nu)}{n}\right)$ for different twist angles. The IEM efficiencies are $95 \pm 5\%$, $92 \pm 10\%$, $85 \pm 5\%$, and $80 \pm 5\%$, while the IHM efficiencies are $85 \pm 5\%$, $80 \pm 5\%$,



$75 \pm 5\%$, and $65 \pm 5\%$ for the 0.2° , 4.6° , 19.6° , and 27.3° heterobilayers, respectively. This yields overall IXM efficiencies of $\sim 90 \pm 5\%$, $86 \pm 8\%$, $80 \pm 5\%$, and $73 \pm 5\%$. The slightly higher η_{IEM} is attributed to the electron population comprising both K–K and K– Γ excitons, where the additional K– Γ IXs contribute to a higher IEM efficiency compared to IHM. Substantial momentum mismatch at larger twist angles⁴⁹ would intuitively be expected to significantly suppress interlayer hot-carrier Coulomb scattering. The IXM process involves two key steps: (i) intralayer optical excitation of high-energy hot carriers, and (ii) interlayer scattering to generate additional electron-hole pairs, with efficient phonon scattering facilitating momentum conservation on a tens-of-femtoseconds timescale⁵⁰. Thus, despite increased momentum mismatch at larger angles, the involvement of carrier-phonon scattering mitigates a more pronounced reduction in IXM efficiency. The QY versus $h\nu$ data further reveal that η_{IXM} decreases with increasing pump energy, a trend more pronounced at larger twist angles. This results from an energy-dependent competition between IXM and hot-carrier relaxation, stemming from reduced interlayer carrier-carrier scattering, potentially due to a lower density of states at higher energies (Supplementary Fig. 18 and Supplementary Note 5). A deeper understanding is provided by subsequent theoretical calculations that account for all scattering pathways.

Notably, the threshold photon energy ($h\nu_{\text{th}}$) and hot carrier transfer (HCT) appear unaffected by twist angle (Fig. 3d). The TA signal rise time under above-threshold excitation is associated with HCT (Supplementary Figs. 19, 20), which can overcome momentum mismatch even at large angles⁵¹. The transfer of hot electrons (holes) allows for broader exploration of $K_{\parallel}$ space above the CBM (VBM), encompassing the M point where it intersects the K valley in WSe₂ (MoS₂) at large twist angles⁵². These energetic hot carriers then relax to the band edge, releasing their excess energy to excite additional carriers via scattering pathways that persist across all twist angles, leaving the threshold largely unaffected by momentum mismatch.

Importantly, to validate the universality of IXM across different material systems, we fabricated another type-II heterobilayer (MoS₂/MoTe₂) with an interlayer bandgap of 0.8 eV (Supplementary Figs. 21–23). Efficient interlayer charge separation and long-lived IX formation are similarly observed in this system. By selectively probing the CBM of MoS₂, we quantitatively monitor the electron population dynamics of IX. The interlayer IEM QY exhibits significant jumps above 1 at the threshold energy of 1.77 eV (Supplementary Fig. 22c), confirming the presence of robust IXM in type-II heterobilayers (Supplementary Fig. 22d).

Theoretical modeling of low-threshold IXM

To provide a theoretical foundation for the observed low-threshold IXM, we performed detailed calculations to

elucidate the underlying mechanisms. Our model confirms the existence of IX multiplication channels with a threshold energy of $2E_{\text{g(type-II)}}$ across various twisted heterobilayers, mediated by interlayer carrier-carrier scattering. The band structure of the heterobilayer is described using an ab-initio-based tight-binding model⁵³, with band offsets adjusted to align with experimental observations.

The IXM process can be classified into hot-electron-induced IEM and hot-hole-induced IHM, each involving two primary steps (Fig. 4a). In the IEM process: (1) A valence electron in WSe₂ is excited to a high-energy state within its conduction band. (2) This hot electron transfers to the MoS₂ layer. If its excess energy exceeds $E_{\text{g(type-II)}}$, it can undergo impact scattering, exciting a second valence electron from WSe₂ to the CBM of MoS₂ and thereby generating an additional electron-hole pair via impact ionization. A analogous process occurs for IHM (Fig. 4a, lower panel). Figure 4b displays the calculated band structure of the 0.2° heterobilayer, illustrating how IXM can occur as electrons scatter from the K/K' valley of WSe₂ to the K/K' valley of MoS₂, conserving both energy and momentum.

Within this framework, the QY for the interlayer multiplication process is calculated by⁵⁴

$$\text{QY}(h\nu) = \frac{1}{N_k^2} \sum_X D_X(h\nu) \frac{2W_X + \gamma_{\text{cool}}}{W_X + \gamma_{\text{cool}}} \quad (1)$$

where $N_k^2 = 10^4$ is the number of k -point samplings in the 2D Brillouin zone, and γ_{cool} is the intralayer hot-carrier cooling rate. For a transition $X = (W; i, m; \mathbf{k})$, the term $D_X(h\nu) = |d_{\mathbf{k}}^{im}|^2 \delta(E_{m\mathbf{k}} - E_{i\mathbf{k}} - h\nu)$ denotes the absorption rate for an electron excited from the initial state $|W, i, \mathbf{k}\rangle$ to the final state $|W, m, \mathbf{k}\rangle$ by a photon of energy $h\nu$. The term W_X represents the scattering rate between two hot carriers, given by:

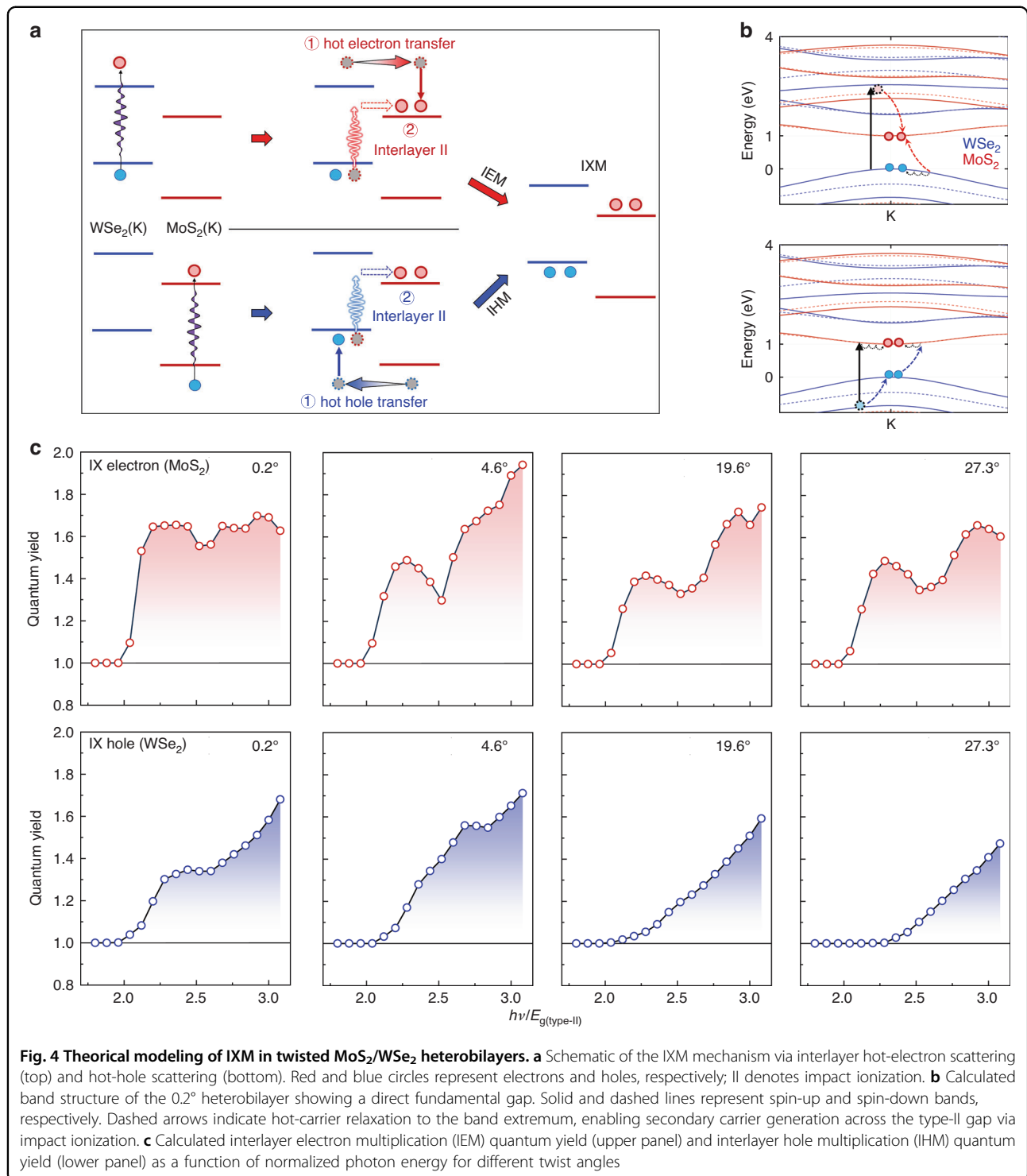
$$W_X = \sum_{m' n'} \sum_{\mathbf{k}' \mathbf{q}'} \sum_{\mathbf{G}} V(\mathbf{k}' - \mathbf{k} - \mathbf{G}) I_{m\mathbf{k}, n\mathbf{q}}^{m' \mathbf{k}', n' \mathbf{q}'} \delta_{\mathbf{k} + \mathbf{q} + \mathbf{G}}^{\mathbf{k}' + \mathbf{q}'} \delta(E_{m\mathbf{k}} + E_{n\mathbf{q}} - E_{m' \mathbf{k}'} - E_{n' \mathbf{q}'}) \quad (2)$$

This formulation includes all scattering pathways that ensure energy and momentum conservation in twisted heterobilayers. Here, $\mathbf{G}$ is a reciprocal lattice vector. The screened Coulomb interaction in k space is:

$$V(\mathbf{k}) = \frac{e^2}{2\epsilon_0 |\mathbf{k}|} \epsilon_{\text{eff}}^{-1}(\mathbf{k}) \quad (3)$$

where ϵ_0 is the vacuum permittivity and ϵ_{eff} is the effective dielectric function obtained by solving Poisson's equation⁵⁵. The interaction strength decreases with increasing momentum difference $|\mathbf{k}|$, which accounts for the





reduced IXM efficiency at larger twist angles. The overlap matrix elements $I_{mk,nq}^{m'k',n'q'}$ are approximated as a constant $I_0(\theta)$ for a given twist angle θ . In our simulations, the ratio $I_0(\theta)/\gamma_{\text{cool}}$ in Eq. (1) is adjusted to account for variations in the cooling rate and interlayer wavefunction overlap at different twist angles. We also define the density of trion

states (DOTS) as:

$$\rho_T = \sum_X \sum_{m'n'm'} \sum_{k'q'q'} \sum_G \delta_{\mathbf{k}+\mathbf{q}+\mathbf{G}}^{\mathbf{k}+\mathbf{q}'} \delta(E_{mk} + E_{nq} - E_{m'k'} - E_{n'q'}) \quad (4)$$

to characterize the number of scattering pathways.



The results in Fig. 4c clearly demonstrate both IEM and IHM when $h\nu \geq 2E_{g(\text{type-II})}$, aligning well with experimental measurements. The consistent low threshold indicates that energy conservation is achieved by leveraging hot-carrier excess energies from band offset energies. Additionally, twist-angle-induced momentum mismatches have an unexpected small effect on the threshold of IXM, indicating the strong hot-carrier interlayer transfer and intervalley phonon-assisted impact ionization⁵⁶. The higher IXM QY at small twist angles results from better interlayer alignment in k space, resulting from the stronger Coulomb scattering. The characteristic peaks in IEM (Figs. 3c and 4c upper panels) and the subtle peaks in IHM (Figs. 3c and 4c lower panels) reflect the features of DOTs as a function of photon energy (Supplementary Fig. 24).

Long-range attractive interaction of multiple interlayer excitons

Beyond the quantitative correlation between TA amplitude and IX density, distinct spectral features provide deeper insights into IXM and the interactions among the generated multiple IXs. Figure 5a reveals clear signatures of enhanced IX populations and interactions. The PB amplitudes probed at the MoS₂ CBM and WSe₂ VBM for excitation above the $h\nu_{\text{th}}$ threshold are significantly higher than those below it, confirming an increased IX population for a fixed absorbed photon density. Furthermore, the electron and hole states above the threshold exhibit a pronounced redshift (Fig. 5a), a phenomenon absent in individual WSe₂ monolayers across the same pump energy range (Fig. 5b). We can rule out laser-induced thermal effects or hot-carrier dynamics as the origin of this shift. This is evidenced by the stable position of the WSe₂ intralayer exciton resonance, which shows no measurable energy shift under identical pump photon energies (Fig. 5b) and at high pump fluence (Supplementary Fig. 25).

Figure 5c summarizes the energy positions of the IX PB peaks for electrons and holes in the 0.2° heterobilayer, compared to the monolayer WSe₂ peak. This redshift is only observed under high pump fluence ($>20 \times 10^{12}$ photons cm⁻²) when exciting below the IXM threshold at 1.9 eV, whereas it appears even at low fluence for above-threshold excitation (Supplementary Fig. 26 and Supplementary Note 6). This fluence dependence further validates that IXM occurs efficiently under low fluence once the photon energy threshold is exceeded. We attribute the redshift in the heterobilayer to attractive interactions arising from quantum-exchange correlation effects⁵⁷ between the single-photon-generated multiple IXs. We define the redshift of the transition energy under above-threshold excitation as $\Delta E(h\nu) = E(2E_g) - E(h\nu)$. The experimental data (Fig. 5d, dots and squares) are well-

described by a Lennard-Jones potential model (solid curve)^{57,58}:

$$\Delta E(h\nu) = \varepsilon \left\{ \left[\frac{r_s}{r} \right]^m - \left[\frac{r_s}{r} \right]^k \right\}, r = [n_0 \pi \text{QY}(h\nu)]^{-\frac{1}{2}} \quad (5)$$

Here, the first term represents short-range Pauli repulsion and the second term long-range vdW attraction (Fig. 5d, solid lines). The variable r is the average distance between electrons or holes in the multiple IX ensemble, related to the charge carrier density $n(h\nu)\pi r^2 = 1$. Given that $n(h\nu) \propto n_0 \cdot \text{QY}(h\nu)$ due to IXM, where n_0 is the initial concentration of photoexcited charge carriers (on the order of 10^{12} cm⁻²), the distance r decreases as the QY increases. The parameter r_s ($s = h$ or e) represents the effective Bohr radius of the exciton.

This redshift is also observed in heterobilayers with other twist angles (Supplementary Figs. 27, 28 and Supplementary Table 6). From the fits, we determine an interaction radius r_h of ~6 nm for holes and r_e of ~4 nm for electrons in the 0.2° heterobilayer. These large distances, significantly exceeding the heterobilayer thickness, signify long-range attractive dipole-dipole interactions between the stable multiple IXs⁵⁷. Furthermore, r_h reaches a minimum at 4.6° before increasing, while r_e rises slightly with twist angle (Supplementary Table 6). Correlating the behavior of r_s with the carrier multiplication QYs from Fig. 3c reveals a general trend: higher carrier multiplication QY correspond to shorter interlayer attractive interaction distances, enhancing the attractive interactions.

IXM-induced photocurrent multiplication

Having established that multiple IXs generated by high-energy photons exhibit long lifetimes—a critical property for efficient carrier extraction—we next demonstrate photocurrent multiplication induced by IXM in a functional device. We fabricated a 3°-twisted MoS₂/WSe₂ photodiode and evaluated its photocurrent QY under varying photon energies (Fig. 6a). The application of a voltage to the WSe₂ layer creates a lateral electric field that separates and collects the electron-hole pairs generated in the heterobilayer. Notably, under a negative bias, photoexcited hot electrons in the WSe₂ conduction band gain additional energy (ΔV) due to the shift of the conduction band, effectively lowering the energy required for impact ionization (Fig. 6a, lower panel). Current-voltage (I - V) characterization shows typical diode behavior in the dark and a clear photovoltaic response under illumination, with an open-circuit voltage in the millivolt range (Fig. 6b; Supplementary Figs. 29, 30). The photocurrent increases linearly with absorbed photon flux across all excitation energies (Fig. 6c), confirming that the response



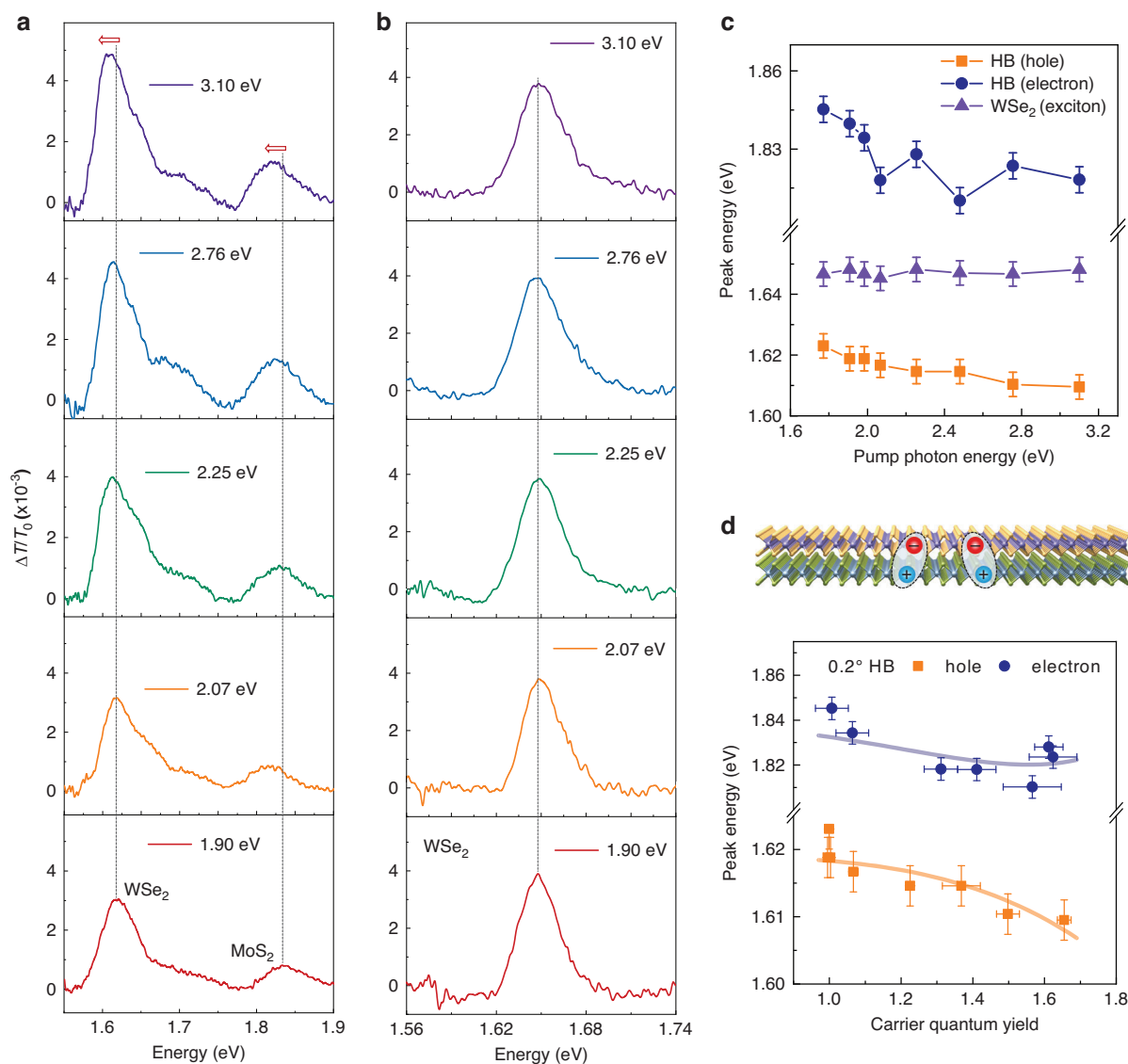


Fig. 5 Long-range attractive interactions between multiple IXs. **a** TA spectra of the 0.2° MoS₂/WSe₂ heterobilayer (HB) at a 1.5 ps delay, measured at a constant absorbed photon density of $3.0 \times 10^{12} \text{ cm}^{-2}$. Dashed lines indicate PB peak positions. **b** Corresponding TA spectra of monolayer WSe₂ under identical conditions for comparison. **c** Transition energies for IX electrons and holes as functions of pump photon energy, and **d** IX QY with theoretical fits (solid curves; see main text). Error bars represent uncertainties in transition energy (vertical) and QY (horizontal). Inset: Schematic of two IXs exhibiting long-range dipole-dipole interactions

is governed by single-photon processes. The slope of these curves corresponds directly to the photocurrent QY at each photon energy. The internal quantum efficiency (IQE) nearly doubles, increasing to ~42% at $V = -1 \text{ V}$ as the photon energy rises from 2 eV to 3.1 eV (Supplementary Fig. 31a).

Figure 6d displays the normalized internal photocurrent QY at $V = 0 \text{ V}$ (self-powered mode), revealing a clear threshold behavior where the QY exceeds unity for photon energies above $2E_{g(\text{type-II})}$. As the electric field is constant for these measurements, any field-induced

effects on exciton dynamics are consistent and accounted for in the normalization. This result directly correlates with the optical measurements in Fig. 3, confirming that the observed IXM translates into a measurable photocurrent gain. Applying a negative bias of -1 V further enhances the effect, yielding both a lower experimental IXM threshold of 1.8 eV and a higher photocurrent QY (Fig. 6d). This is attributed to the additional potential energy imparted to the carriers, which reduces the net energy required for impact ionization. Our theoretical calculations corroborate this, showing enhanced IXM



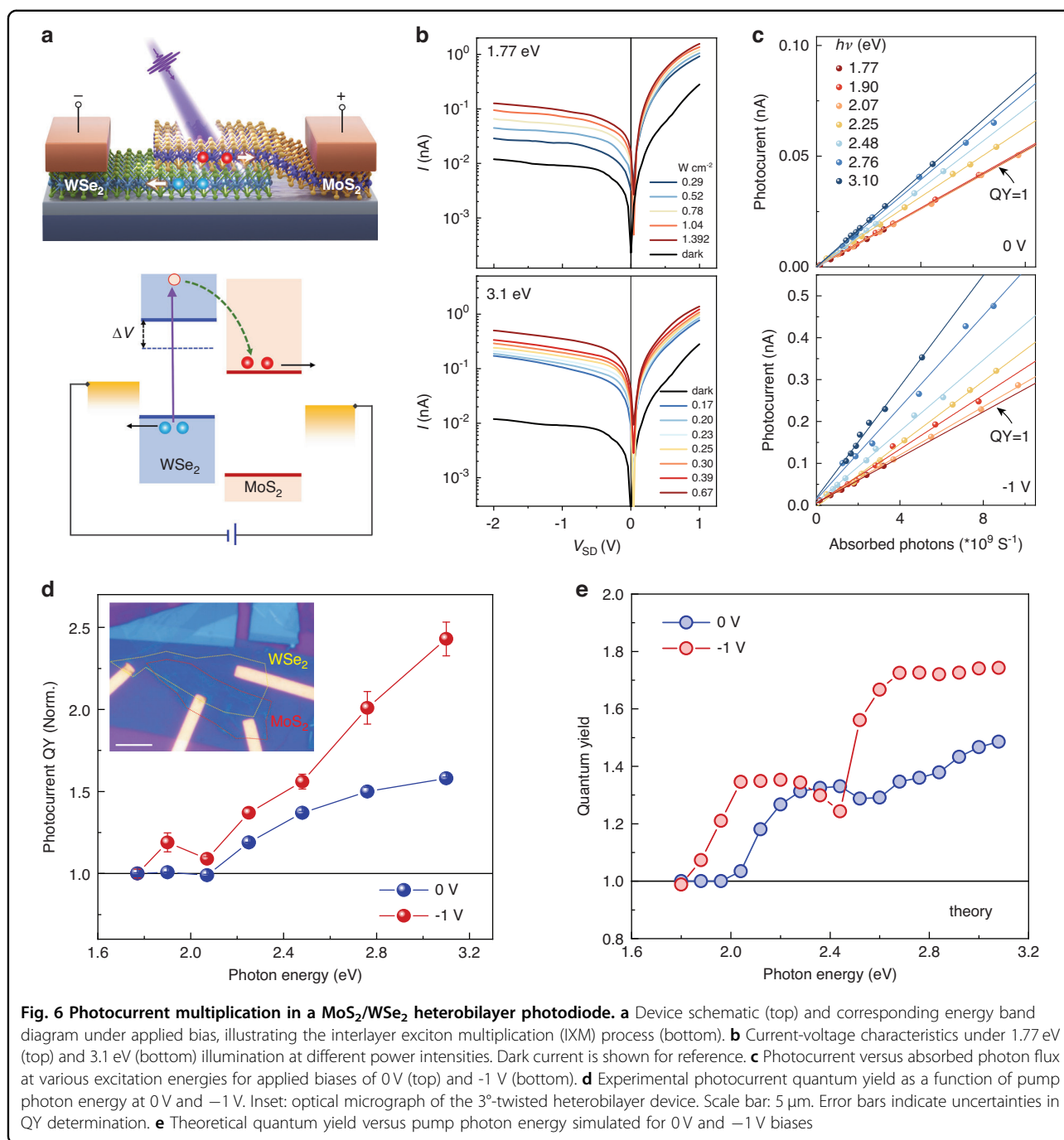


Fig. 6 Photocurrent multiplication in a $\text{MoS}_2/\text{WSe}_2$ heterobilayer photodiode. **a** Device schematic (top) and corresponding energy band diagram under applied bias, illustrating the interlayer exciton multiplication (IXM) process (bottom). **b** Current-voltage characteristics under 1.77 eV (top) and 3.1 eV (bottom) illumination at different power intensities. Dark current is shown for reference. **c** Photocurrent versus absorbed photon flux at various excitation energies for applied biases of 0 V (top) and -1 V (bottom). **d** Experimental photocurrent quantum yield as a function of pump photon energy at 0 V and -1 V. Inset: optical micrograph of the 3° -twisted heterobilayer device. Scale bar: 5 μm . Error bars indicate uncertainties in QY determination. **e** Theoretical quantum yield versus pump photon energy simulated for 0 V and -1 V biases

efficiency and a lower threshold under negative bias on WSe_2 (Fig. 6e). At a given pump energy, carriers under negative bias possess greater excess energy, opening more scattering pathways and increasing the QY. This IXM-driven enhancement directly improves device performance. Due to the multiplication process, the external quantum efficiency (EQE) and responsivity increase from 0.9% and 4.9 mA W^{-1} at 2 eV to 4.7% and 15.4 mA W^{-1} at 3.1 eV under a -1 V bias (Supplementary Fig. 31b, c).

Discussion

In summary, we have demonstrated highly efficient, low-threshold IXM in twisted type-II TMDC heterobilayers. The IXM process features a threshold as low as twice the interlayer bandgap, approaching the fundamental energy conservation limit. Our combined experimental and theoretical work reveals that this low threshold is robust against twist-angle-induced momentum mismatch, enabled by efficient hot-carrier interlayer transfer and phonon-assisted scattering. We achieved a



peak IXM efficiency of $\sim 90\%$ in a 0.2° heterobilayer, where shorter inter-carrier distances enhance attractive interactions. This efficiency gradually decreases with larger twist angles and higher photon energies due to reduced interlayer Coulomb scattering. Furthermore, we successfully translated this optical multiplication into a functional device, demonstrating IXM-induced photocurrent gain. The photocurrent QY exhibits a clear multiplication threshold at $2E_{g(\text{type-II})}$ in a self-powered photodiode, which can be further reduced to $1.8E_{g(\text{type-II})}$ under a modest reverse bias. Our findings establish a new paradigm for carrier multiplication and pave the way for developing highly efficient optoelectronic devices, such as photodetectors and solar cells, that surpass conventional efficiency limits. Additionally, the observed long-range interactions between stable multiple IXs highlight the potential of these systems for exploring novel quantum optical phenomena.

Materials and methods

Sample fabrication

Monolayers of MoS_2 , WSe_2 , and MoTe_2 , along with few-layer h-BN flakes, were mechanically exfoliated from their bulk crystals using adhesive tape. The exfoliated flakes were transferred onto a polydimethylsiloxane (PDMS) stamp, and their thicknesses were identified based on optical contrast under an optical microscope. Twisted $\text{MoS}_2/\text{WSe}_2$ and $\text{MoS}_2/\text{MoTe}_2$ heterobilayers were fabricated using a precision dry-transfer technique under optical microscopy guidance. The heterostructures were fully encapsulated by top and bottom h-BN layers. Specifically, a selected h-BN flake was first transferred onto a quartz substrate. Subsequently, MoS_2 , WSe_2 (or MoTe_2), and another h-BN flake were sequentially transferred with meticulous alignment to achieve the target twist angles. Following each transfer step, the sample was annealed in a vacuum at 473 K for 3 h to enhance interfacial cleanliness and contact quality.

Device fabrication and characterization

Monolayer flakes of MoS_2 , WSe_2 , and MoTe_2 were mechanically exfoliated onto PDMS stamps. Selected flakes were then precisely aligned and vertically stacked using a dry-transfer method onto a Si substrate with a 300 nm thermal oxide layer. The substrate was pre-cleaned by sequential ultrasonication in acetone, isopropyl alcohol, and deionized water. The heterostructure was subsequently annealed at 200°C for 2 h in vacuum to improve interfacial contact. Cr/Au electrodes (10/50 nm) were then fabricated on the $\text{MoS}_2/\text{WSe}_2$ or $\text{MoS}_2/\text{MoTe}_2$ heterobilayers via standard electron-beam lithography, thermal evaporation, and a lift-off process. Finally, a few-layer h-BN flake was transferred onto the active channel as a top-gate dielectric, followed by a second annealing

step at 200°C for 2 h. Electrical characterization of the devices was performed using a Keithley B1500A semiconductor parameter analyzer in a Lake Shore cryogenic probe station under a vacuum of 10^{-6} Torr. Measurements were conducted in both dark and illuminated conditions. The illumination source was a fiber-coupled laser system, and the laser power was calibrated before each measurement using a Newport 843-R optical power meter with a PD300-UV photodetector. All electrical measurements were performed at room temperature.

Angle-resolved second-harmonic-generation

The SHG measurements were conducted using a home-built inverted microscopy system. A linearly polarized femtosecond laser light with the following parameters: wavelength of 800 nm, repetition rate of 80 MHz, pulse duration of 100 fs, and power of 1 mW, was focused onto the monolayer region of either MoS_2 or WSe_2 using a $50\times$ objective. The reflected 400 nm light was collected by the same objective and directed through a linear polarizer and a 400 nm narrow band filter before reaching a Spectra Pro 2300i Acton equipped with a CCD camera (Pixis 256BR, Princeton Instruments). During the measurements, the polarization direction of both the excitation light and the collected light was fixed, while the heterobilayer sample was rotated. The absolute rotational angle is given in arbitrary units. The angular-dependent SHG intensity (I) was fitted using the equation $I = I_{\max} \cos^2[3(\theta + \varphi)]$, where φ is between 0° and 30° . Since SHG intensity measurements are not sensitive to phase, they produce a 6-fold symmetry pattern. The extracted twist angle $\Delta\phi = |\varphi_{\text{W}} - \varphi_{\text{M}}|$ for all heterobilayers ranged between 0° and 30° . It is important to note that all monolayer samples exhibited a near-perfect 6-fold symmetry, which confirms their single crystal nature. All optical measurements were performed at room temperature.

Steady-state absorption microscopy

The absorption spectra of the monolayers and heterobilayers were measured using a laser-driven broadband light source (EQ-77-fc, Energetiq). The light was focused to a spot size of $\sim 2\mu\text{m}$ at the sample plane. The transmitted light was collected by a $50\times$ microscope objective (NA = 0.5) and directed to a spectrometer (Spectra Pro 2300i, Acton) coupled with a CCD camera (Pixis 256BR, Princeton Instruments). Absorbance (A) was calculated using the formula $A = \log_{10}(I_0/I)$, where I is the transmitted intensity through the sample (TMD flake and substrate) and I_0 is the reference intensity transmitted through the bare sapphire substrate.

Femtosecond transient absorption microscopy

Carrier dynamics were investigated using a femtosecond TA microscope in a transmission geometry. The system



was driven by a 1 kHz Ti:sapphire amplified laser system, producing 50 fs pulses centered at 800 nm. The output was split into two paths: the majority of the pulse energy pumped an optical parametric amplifier to generate a wavelength-tunable pump beam, which was mechanically chopped at 500 Hz. The remaining 800 nm fundamental beam was focused onto a sapphire crystal to generate a white-light continuum, serving as the broadband probe. The pump and probe pulses were spatially combined using a beam splitter and temporally and spatially overlapped at the sample position using a microscope objective. The time delay between them was controlled by a motorized linear stage with a retroreflector in the probe path. After transmitting through the sample, the probe beam was dispersed by a spectrometer and detected by a CMOS sensor, covering the visible spectrum (400–800 nm). This configuration allowed for the measurement of the differential transmittance ($\Delta T/T_0$), which reflects pump-induced changes in the sample's complex refractive index and correlates with the photocarrier density. All measurements were conducted at room temperature under ambient conditions.

First-principles calculations

First-principles calculations based on density functional theory (DFT) were performed using the *Quantum ESPRESSO* package to investigate the electronic structure of twisted MoS₂/WSe₂ heterobilayers. Commensurate supercells for various twist angles were generated using the *Twister* tool⁵⁹. A vacuum spacing of 25 Å was introduced along the out-of-plane direction to eliminate spurious interactions between periodic images. The Perdew–Burke–Ernzerhof exchange–correlation functional within the generalized gradient approximation was employed⁶⁰. Norm-conserving pseudopotentials were used for Mo and W atoms, while ultrasoft pseudopotentials were used for S and Se atoms. The kinetic energy cutoffs were set to 40 Ry for the wavefunctions and 200 Ry for the charge density. For each supercell, self-consistent field calculations were performed using a $9 \times 9 \times 1$ Monkhorst–Pack k-point mesh, with a Gaussian smearing of 0.01 Ry. The total energy was converged to within 10^{-8} Ry. To analyze the layer hybridization, the partial charge densities of states at high-symmetry k-points (e.g., K and Γ) were computed. The spatial distribution of these states across the MoS₂ and WSe₂ layers was quantified by integrating the partial charge density within each monolayer's region³¹. The boundary between the two layers was defined by the minimum in the plane-averaged charge density along the out-of-plane direction.

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Author contributions

These authors contributed equally: Pengzhi Wang, Gan Wang, Chenhao Wang. P.W. prepared samples and performed optical characterizations. P.W. performed ultrafast spectroscopy measurements. T.D. guided G.W. and Y.C. in performing theoretical simulations. C.W. and P.W. prepared devices and electrical characterizations. Q.W., Q.L., L.Z., H.R., J.P., L.Z., and X.Z. helped with the sample fabrication and characterizations. All authors agreed with the final version of the manuscript. M.L. led the project.

Data availability

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

Conflict of interest

The authors declare no competing interests.

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