

Twist-angle tunable bandgap renormalization and exciton binding energy in WS₂ bilayers

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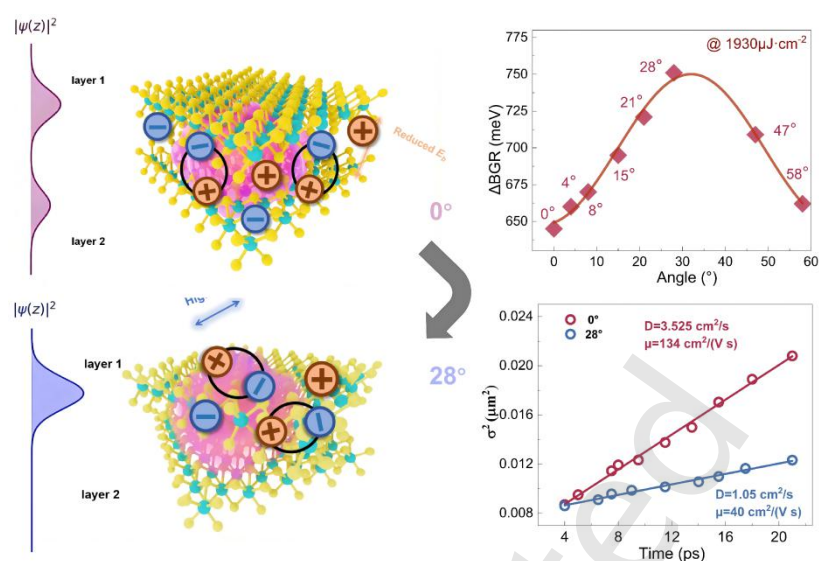
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Twist-angle engineering in bilayer WS₂ enables simultaneous control of exciton binding, photoinduced bandgap renormalization, and exciton transport. As the twist angle increases from 0° to 28°, weakened interlayer coupling enhances exciton localization and increases Eb while ΔBGR reaches its maximum near 28° under strong photoexcitation. Meanwhile, the 0° bilayer maintains superior exciton diffusion and mobility owing to stronger interlayer coupling.

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ABSTRACT: The twist angle at van der Waals interfaces has emerged as a powerful degree of freedom for tailoring excitonic energy landscapes in two-dimensional semiconductors, yet how interlayer coupling and photoinduced dynamic dipole screening cooperatively govern excitonic responses remains elusive. Here, we directly investigate the twist-angle-dependent Coulomb-dominated excitonic effects in bilayer WS₂, including bandgap renormalization (BGR), exciton binding energy, recombination lifetime, and mobility. Femtosecond transient absorption spectroscopy reveals a twist-angle-tunable exciton binding energy from 265 to 336 meV; upon approaching the Mott transition threshold, BGR varies by over 100 meV and is most significant near-30°, where weakened interlayer coupling makes Coulomb interactions dominant. Moreover, the exciton lifetime and mobility are jointly dictated by the twist angle: strong interlayer coupling suppresses recombination while enhancing exciton mobility by threefold. This work integrates photoinduced screening with twist-angle engineering, establishing a unified picture of exciton many-body dynamics and offering guidance for high-performance excitonic devices.

KEYWORDS: bilayer WS₂, twist angle, interlayer coupling, bandgap renormalization, exciton binding energy, dynamic dipole screening

1 Introduction

Unlike conventional semiconductors that rely on surface dangling bonds to modify their optoelectronic properties, two-dimensional (2D) transition metal dichalcogenides (TMDs) provide an undisturbed quantum confinement environment for excitons, owing to their atomically flat and dangling-bond-free surfaces[1-4]. In such ideal interfaces, exciton formation, migration, and interaction are governed mainly by intrinsic Coulomb interactions and quantum confinement effects rather than disorder-induced scattering[5-7]. In the 2D limit, reduced dielectric screening and strong quantum confinement lead to tightly bound exciton states, which dominate the optical response of TMDs and provide a platform for exploring correlated phenomena, such as excitonic insulators[8], Mott transitions[9], and non-equilibrium many-body physics[10]. Meanwhile, stacking layers of TMDs with controlled orientations to construct van der Waals homo- or hetero-structures introduces a twist angle, which continuously tunes symmetry breaking in the 2D system[11-15]. The twist angle not only modifies the interlayer coupling strength but also reconstructs the topological structure of electronic bands and the quantum geometry of excitons through the periodic potential of moiré superlattices[16,17]. This geometric reconstruction alters exciton binding energies and spatial distributions, providing a versatile platform for the artificial design of excitonic physical phenomena[18-20].

Complementing twist engineering, the dynamic dipole screening effect induced by photogenerated carriers under intense optical excitation provides a powerful route for modulating excitonic many-body interactions in 2D TMDs[21-23]. High-density optical excitation weakens the electron-hole Coulomb attraction, leading to reduced exciton binding energy accompanied by quasiparticle bandgap renormalization (BGR), with tuning amplitudes reaching several hundred millielectronvolts[24,25]. Compared to conventional tuning methods, such as electrostatic gating[26], chemical doping[27], or strain[28], photoinduced bandgap renormalization offers unique advantages of all-optical and ultrafast response. It enables dynamic modulation of the material's band structure on femtosecond to picosecond timescales and allows rapid recovery upon excitation cessation, providing unprecedented flexibility for exploring nonequilibrium many-body physics and designing ultrafast optoelectronic devices. As the carrier density approaches the Mott transition threshold, the system undergoes a transition from an excitonic insulator to an electron-hole plasma[29]. This light-driven phase transition not only serves as an ideal platform for studying nonequilibrium many-body physics but also holds promise for realizing novel device functionalities including ultrafast switches, optical logic gates, and artificial neurons[30-32]. However, current studies have largely focused on monolayers or multilayers with fixed stacking configurations. A central challenge remains: how to integrate twist angle with photoinduced Coulomb

screening to achieve synergistic and continuously tunable control over BGR and exciton binding energy. Elucidating this mechanism is essential for understanding many-body correlations in 2D materials and advancing excitonic devices.

In this work, we demonstrate twist-angle control of bandgap renormalization and exciton binding energy in bilayer WS₂ by exploiting the strong angle dependence of interlayer coupling. Bilayers with controlled twist angles were fabricated to examine how interlayer coupling and many-body Coulomb correlations cooperate under high-density excitation. Raman and photoluminescence (PL) spectroscopy was used to track the evolution of interlayer coupling through characteristic peak shifts and changes in the trion-to-exciton emission ratio. Transient absorption (TA) spectroscopy shows that increasing the twist angle from 0° to 30° tunes the exciton binding energy from 265 meV to 336 meV. Under elevated excitation densities, enhanced exciton-exciton interactions strengthen photoinduced dynamic dipole screening, reducing the binding energy and producing a bandgap renormalization modulation exceeding 100 meV near the Mott transition. In addition, exciton dynamics display strong twist-angle dependence. The 0°-twisted bilayer WS₂, characterized by maximal interlayer coupling, exhibits significant electron-hole spatial separation and higher delocalization. This configuration suppresses recombination, yielding a lifetime of 103 ps and an effective exciton transport mobility of 134 cm²/(V·s), thrice that of the bilayer WS₂ with a twist angle of approximately 30°. In contrast, the near-30° twisted bilayer, where interlayer coupling is weakest, behaves akin to a monolayer. Collectively, these findings establish the twist angle as a critical degree of freedom for tailoring high-density many-body Coulomb interactions in 2D TMDs.

2 Results and discussion

Here, monolayer WS₂ flakes were grown on sapphire by chemical vapor deposition (CVD). Twisted bilayer WS₂ samples with well-defined twist angles were assembled on SiO₂/Si substrates using a polystyrene-assisted transfer method (see METHODS for more details). The corresponding atomic force microscopy (AFM) images of the monolayer sample and the bilayer sample are provided in Electronic Supplementary Material Figure S1. Figure 1a schematically illustrates the pump-probe investigation of photoinduced many-body effects in bilayer WS₂. Under photoexcitation, the elevated carrier density strongly screens the Coulomb interaction: as the attractive force between electrons and holes is attenuated, the exciton binding energy decreases substantially. Simultaneously, the screening of repulsive Coulomb interactions among like charges reduces the quasiparticle energy, leading to a pronounced bandgap renormalization (BGR). Figure 1b displays optical microscopy images of the samples at various twist angles; the inset shows polarization-resolved second-harmonic generation (SHG). The twist angle is determined from the armchair direction (indicated by the petal-like patterns)[33,34]. The optical microscopy images and polarization-resolved SHG characterizations of the remaining samples (0°, 8°, 21°, 47°, and 58°) are provided in Figure S2, confirming the reliability of the twist-angle determination. Figure 1c presents the twist-angle-dependent Raman spectra. At 0°, the characteristic E_{2g}¹ (357 cm⁻¹) and A_{1g} (419 cm⁻¹) are observed. When the twist angle deviates from 0° or 60°, E_{2g}¹ exhibits a slight blue shift, whereas A_{1g} red-shifts maximally near 30°. The evolution of the frequency differences

between E_{2g}¹ and A_{1g} modes as a function of twist angles is illustrated in Figure 1d. Meanwhile, Figure 1e reveals that the intensity of the A_{1g} mode at 0° is approximately 2.2 times that at near-30°, indicating stronger interlayer coupling at 0° that enhances Fröhlich-type phonon-exciton interactions. Near 0° or 60°, structural registry maximizes interlayer hybridization, resulting in a large frequency splitting; any deviation weakens interlayer coupling[35-37]. These observations confirm that the twist-angle dependence of Raman peaks directly reflects the modulation of interlayer coupling, underscoring the critical role of the twist angle in tuning the electronic structure and excitonic behaviors.

The PL spectra in Figure 1f reveal that variations in the twist angle significantly influence the optical properties of bilayer WS₂. The spectra exhibit two distinct peaks corresponding to the neutral A exciton (A) and the trion (A⁻). Monolayer WS₂ typically shows higher PL intensity and narrower linewidths compared to bilayer WS₂, which is attributed to interlayer charge transfer and changes in the band structure upon increasing the layer number[38]. As demonstrated in Figure 1g, the trion binding energy in bilayer WS₂ is derived from the energy difference between the A⁻ and A peaks[39]. This energy difference varies with the twist angle: the trion binding energy increases near 0° or 60°, decreases near 30°, and reaches a minimum of approximately 37 meV. Figure 1h presents the twist-angle dependence of the A⁻/A intensity ratio obtained from spectral fitting. Previous studies have established that this intensity ratio is proportional to the excess electron concentration (n_e)[40,41]. As the twist angle approaches 30°, n_e reaches its minimum, facilitating electron detachment from trions and yielding the smallest trion binding energy. Conversely, near 0° or 60°, n_e maximizes, hindering electron removal and resulting in the largest trion binding energy. These findings highlight the pivotal role of twist-angle-controlled interlayer coupling in governing many-body exciton-charge interactions in bilayer WS₂.

Under nonequilibrium conditions at room temperature, the exciton binding energy in twisted bilayer WS₂ is dynamically controlled by photoexcited carriers, which intrinsically couples it to bandgap renormalization. To investigate this coupling, spectrally resolved TA spectroscopy was employed to systematically measure the decay dynamics of the A-exciton ground-state bleach (GSB) signal in the twisted bilayer WS₂. Figure 2a illustrates the underlying mechanism by comparing Rydberg state configurations under low and high excitation densities. As the excitation density increases, the dynamic dipole field of photogenerated carriers efficiently screens the Coulomb interaction between electrons and holes, leading to a contraction of the single-particle bandgap and a reduction in the exciton binding energy. Beyond a critical carrier density, higher-lying Rydberg states become unstable and merge into the continuum, marking the onset of exciton dissociation and the gradual transition toward an electron-hole plasma. Figure 2b shows the ultrafast dynamics of a bilayer WS₂ with a twist angle of 0°, measured at various excitation wavelengths under a pump fluence of 64 μJ·cm⁻².

To quantitatively determine the exciton binding energy, we employed a micro-area TA spectroscopy with sub-picosecond resolution. This approach tracks the ultrafast decay kinetics of the GSB signal following photoexcitation to identify the

threshold for free-carrier generation[23,42]. By tuning the excitation energy near the threshold, the Rydberg-to-continuum transition can be resolved (see Electronic Supplementary Material Note 1 for threshold analysis). This method not only provides an estimate of the exciton binding energy in the excited state but also captures the influence of dynamic dipole screening. The probe wavelength was fixed at 626 nm (1.98 eV), that is the A-exciton GSB maximum, while the excitation wavelength was tuned from 590 nm (near the A-exciton resonance) to 505 nm (well above the bandgap). When the photon energy exceeds the electronic bandgap (E_{gap}), nonequilibrium free carriers are generated; these hot carriers cool and form excitons on a sub-picosecond timescale, introducing a fast decay component. Two critical time points, indicated by black arrows in the figure, were selected for quantitative analysis: I_{max} , the maximum value of the excitation signal at the instant of excitation, and $I_{(t = 3.5 \text{ ps})}$, the amplitude at 3.5 ps after excitation (by which time the sub-picosecond process has concluded). The ratio $I_{\text{max}}/I_{(t = 3.5 \text{ ps})}$ serves as a metric for the GSB decay rate. Plotting this ratio against the excess excitation energy (Figure 2c) reveals two distinct thresholds. The higher threshold corresponds to the continuum edge (i.e., the bandgap), where the excess energy directly equals the exciton binding energy. By analyzing the second threshold between the 1S state and the continuum, we extract the energy difference $\Delta E = h\nu - E_A$, where E_A is the A-exciton energy. This experimental strategy circumvents complications arising from excited-state overlap in steady-state spectroscopy and provides a direct optical means to monitor bandgap renormalization and exciton screening effects in real time.

As demonstrated in Figure 2d, the A-exciton binding energy exhibits a significant nonlinear decrease with increasing excitation density (from $64 \mu\text{J}\cdot\text{cm}^{-2}$ to $190 \mu\text{J}\cdot\text{cm}^{-2}$), dropping from 265 meV to 207 meV. This behavior directly confirms that the dynamic dipole screening effect induced by high-density excitons effectively weakens the Coulomb interactions within the system. However, due to measurement limitations, we could not observe a steep decline in binding energy at the highest excitation densities; this is likely related to the self-amplifying exciton ionization effect[43]. Figure S3 shows the excitation wavelength threshold as a function of pump power densities for the 0° -twisted sample. Figure 2e summarizes the twist-angle dependence of the A-exciton binding energy. The decay ratio of the GSB signal intensity as a function of excitation wavelength for samples with different twist angles is shown in Figure S4. The threshold drift induced by twist angle variations is closely linked to exciton wavefunction hybridization and electron delocalization arising from interlayer coupling[44,45]. As the twist angle increases from 0° to near- 30° , the exciton binding energy rises from 265 meV to 336 meV. The difference between the 0° and 58° samples is mainly attributed to stacking-dependent interlayer coupling and the resulting screening strength. At 0° , the 3R-like stacking promotes strong exciton wavefunction delocalization and efficient interlayer screening, which weakens the electron-hole Coulomb attraction and yields the lowest binding energy. As the twist angle approaches 30° , weakened interlayer coupling reduces screening and increases the binding energy. Near 58° , partial recovery of 2H-like interlayer coupling leads to intermediate screening and a binding energy of 285 meV.

The photoinduced dynamic dipole screening effect simultaneously reduces the exciton binding energy and the electronic bandgap, leading to bandgap renormalization. As illustrated in Figure 3a, under equilibrium conditions, the sample exhibits strong Coulomb interactions due to the pronounced quantum confinement and weak dielectric screening of TMDs, producing a substantial energy offset between the electronic and optical bandgaps. Upon low pump density, the photogenerated carrier concentration gradually increases, enhancing Coulomb screening. Since the reduction in the electronic bandgap (E_g) slightly exceeds that in the exciton binding energy (E_b), the optical bandgap undergoes a slight transient redshift. When the excitation density surpasses the Mott transition threshold, the binding energy diminishes sufficiently that the continuum merges with the A-exciton resonance, triggering an insulator-to-metal transition. The critical photoexcited carrier density at which the excitonic resonance vanishes is defined as the Mott density (n_{Mott}). Based on the fact that the exciton Bohr radius a_0 in the double-layer system is approximately 1 nm, this critical density is usually approximated as $n_{\text{Mott}} \approx 1/(\pi a_0^2)$. A rough estimation of the Mott density for this system yields $n_{\text{Mott}} \sim 1 \times 10^{14} \text{ cm}^{-2}$ [46,47].

For excitation densities above n_{Mott} , abundant free carriers efficiently screen the Coulomb potential and strongly renormalize the quasiparticle bandgap. We excited the samples with a 100 fs pump pulse at 2.45 eV (505 nm), above the ionization continuum onset of the A-exciton transition, and monitored the photoinduced absorption change (ΔA) using a broadband white-light probe from 520 to 800 nm. Figure 3b presents the TA spectra of the 0° -twisted bilayer WS₂, recorded at a delay time of 0.35 ps after excitation, for pump fluences ranging from $160 \mu\text{J}\cdot\text{cm}^{-2}$ to $1930 \mu\text{J}\cdot\text{cm}^{-2}$.

At relatively low fluences, the band-edge transitions exhibit bleaching of excitonic resonances accompanied by broadening of the exciton peaks. This Coulombic effect becomes increasingly pronounced with rising fluence[48]. As the conduction and valence bands are populated by high-density electrons and holes, the renormalized bandgap shifts below the original exciton energy. In the spectral domain, this process appears as a low-energy negative differential absorption feature, originating from population inversion near the band edge. The onset of this negative signal, highlighted in the pseudocolor map in Figure 3c, is used to determine the renormalized bandgap edge. The detailed procedure for extracting the BGR magnitude from the TA spectra is described in Supporting Information Note 2. The negative differential absorption feature observed below the original exciton energy (highlighted by the box) corresponds to the onset of the renormalized bandgap edge. For the 0° bilayer near the Mott density, the equilibrium quasiparticle bandgap is determined from the A-exciton energy and exciton binding energy ($E_g^0 = E_A + E_b$). The photoexcited quasiparticle bandgap shifts to 1.57 eV, resulting in a BGR energy of 645 meV (see Supporting Information Note 2 for details).

To determine the critical carrier density for the Mott phase transition, we systematically measured the transient bleaching signal amplitude at the A-exciton resonance energy as a function of pump fluence (Figure 3d). As shown in Figure 3e, the signal initially increases linearly with injected carrier density due to phase-space filling and then gradually saturates, indicating that

modulation of the probe pulse by excitonic states approaches its limit. The power density at signal amplitude saturation is $1930 \mu\text{J}\cdot\text{cm}^{-2}$ (corresponding to a carrier density of $2.45 \times 10^{14} \text{ cm}^{-2}$), which is on the same order of magnitude as the estimated n_{Mott} . Near this critical point, the rapid rise in free carrier density induces strong Coulomb screening, sharply reducing exciton binding energy until the excitonic states fully dissociate. Beyond this threshold, the exciton population ceases to grow significantly, because excess photogenerated carriers exist predominantly as unbound electron-hole pairs. The saturation of the signal amplitude thus serves as an experimental signature of the transition from an exciton gas to an electron-hole plasma. Moreover, the absorption features at the band-edge exciton resonance peaks were significantly suppressed (Figure S5), and this experimental result further confirmed the occurrence of the Mott transition.

Figure 3f shows the pseudocolor TA map of the 28° -twisted sample under excitation close to the Mott density, with other twist angles shown in Figure S6. The evolution of the A exciton energy with increasing excitation power is governed by the competition between two distinct many-body mechanisms. First, screening effects reduce the exchange-correlation energy, driving bandgap renormalization characterized by a downward shift of the conduction band minimum and an upward shift of the valence band maximum[49]. This bandgap contraction promotes a spectral redshift. Second, the enhanced dielectric screening weakens the electron-hole Coulomb coupling, reducing exciton binding energy. This binding energy decrease counteracts the bandgap narrowing, favoring a blueshift of the exciton peak. The net redshift observed in Figure 3g indicates that bandgap narrowing dominates over the reduction in exciton binding energy. To quantitatively evaluate the evolution of the A-exciton energy as a function of photoexcitation density, fluence-dependent TA spectra were collected, and the A-exciton resonance positions were extracted by fitting the spectral features around the exciton peak (Fig. S7). Notably, the magnitude of the A-exciton redshift is substantially larger in samples near 0° than in those near 30° . It should be noted that this fluence-dependent exciton redshift represents the net optical transition shift and should not be directly compared with the BGR extracted at the Mott transition density. At high carrier densities, the BGR is governed by the many-body quasiparticle renormalization, where the weaker background screening near 30° produces a larger renormalization effect. In the 0° samples, stronger interlayer interactions likely make the bandgap more sensitive to carrier density, leading to a more pronounced net redshift under low excitation conditions.

Figure 3h illustrates the twist-angle dependence of the BGR magnitude in bilayer WS_2 . At pump fluences corresponding to the Mott transition density, the BGR exhibits opposite trends for the two representative samples: the sample near 30° displays a more pronounced BGR, whereas the sample near 0° shows a smaller BGR. This angle-dependent BGR response reflects how the initial dielectric environment constrains many-body interactions in 2D semiconductors. Weaker dielectric screening amplifies many-body effects such as exchange and correlation[50]. Consequently, under the same photogenerated carrier density, the 28° sample, with its weaker initial background screening, exhibits stronger Coulomb enhancement. As the system approaches the Mott transition, photogenerated

carriers induce a dramatic change in screening strength, resulting in a much larger bandgap renormalization. In contrast, the stronger initial interlayer coupling in the 0° sample partially counteracts the contribution from photogenerated carriers, suppressing the net renormalization at the Mott transition. TA spectra for other twist angles are provided in Figure S8. Overall, twist-angle tuning enables controllable BGR from 645 to 751 meV, demonstrating that interlayer-coupling engineering provides an effective route to manipulate the electronic band structure of TMDs under extreme nonequilibrium conditions.

Subsequently, we investigated the twist-angle dependence of the A-exciton lifetime and mobility in bilayer WS_2 (Figure 4). As established above, interlayer coupling in bilayer WS_2 is strongly modulated by the twist angle. Figure 4a schematically illustrates that the interlayer coupling in this van der Waals homobilayer is highly anisotropic and sensitive to stacking configuration. At 0° , favorable orbital alignment and energy degeneracy produce strong interlayer coupling, promoting electron-wavefunction delocalization and reconstruction of excitonic Rydberg states. In contrast, when the twist angle approaches 30° , momentum-space misalignment sharply reduces orbital overlap, suppressing interlayer interactions and driving the excitonic properties toward the monolayer-like regime[14,51]. From the TA dynamics, we observe a significant angular dependence of the exciton lifetime (Figure 4b), fitted with a double-exponential function. The fast lifetime component (τ_1) shows little variation with twist angle, as it is associated with ultrafast relaxation or nonradiative processes that are less affected by interlayer coupling. The slow component (τ_2), which reflects exciton diffusion and recombination, is highly sensitive to the degree of exciton wavefunction delocalization.

As the twist angle increases from 0° to 28° , τ_1 decreases from 3 to 1.7 ps, whereas τ_2 drops markedly from 103 to 29 ps (Figures 4c,d). This behavior arises from twist-angle-dependent interlayer coupling. In the 0° sample, interlayer charge redistribution reduces electron-hole wavefunction overlap, suppressing recombination and extending the exciton lifetime. In the 28° sample, weakened interlayer coupling enhances the intralayer exciton character and increases electron-hole overlap, leading to faster recombination. Samples measured at low power also exhibit a strong twist angle dependence (Figure S9), with generally longer lifetimes, confirming that many-body interactions (such as Auger recombination and exciton-exciton annihilation) dominate charge carrier dynamics under extreme non-equilibrium conditions. The stronger lifetime reduction in the 0° sample at high fluence is attributed to its indirect-bandgap character induced by interlayer coupling, where intense Coulomb screening activates previously suppressed nonradiative channels[52]. In contrast, the 28° -twisted sample exhibits a recombination rate closer to that of a direct transition, with a lower initial lifetime. Under weak coupling, restricted exciton diffusion causes the many-body annihilation effect to saturate rapidly, making the lifetime less sensitive to further power increases. These observations clearly demonstrate the ultrafast exciton dynamics and significant many-body effects in non-equilibrium twisted bilayer WS_2 .

To further validate the influence of interlayer coupling on exciton spatial dynamics, we compared exciton migration in bilayer WS_2 with twist angles of 0° and 28° . Figure 4e presents transient absorption microscopy (TAM) images of the

ground- state band region for the 0°-twisted sample, acquired over time delays from 1 to 21 ps. The corresponding images for the 28° sample are shown in Figure S10. The experimental data were fitted with a two- dimensional Gaussian function, and the variance (σ^2) of the exciton distribution evolves linearly with time t , consistent with the diffusion equation:

$$\sigma^2 = \sigma_0^2 + 2Dt$$

Here, D represents the diffusion coefficient, which is extracted from the temporal evolution of the spatial variance of the exciton distribution. To quantitatively compare the exciton diffusion behavior, the spatial TA intensity profiles at selected delay times were extracted and plotted in Figures 4f and 4g for the 0° and 28° samples, respectively. The experimental profiles were fitted with Gaussian functions, from which the variance σ^2 at each delay time was obtained. The temporal broadening of the contours visually illustrates exciton diffusion. Using the Einstein relation, an effective exciton transport mobility (μ_{eff}) was estimated from the diffusion coefficient:

$$\mu_{\text{eff}} = \frac{eD}{k_B T}$$

In this formula, e represents the elementary charge, k_B denotes the Boltzmann constant, and T denotes the temperature. Figure 4h shows the linear fits of σ^2 versus time for bilayer WS₂ with twist angles of 0° and 28°. The 0°-twisted sample, with the strongest interlayer coupling, exhibits delocalized exciton wavefunctions and enhanced interlayer transport, resulting in a high mobility of 134 cm²/(V·s). In contrast, due to weak interlayer coupling, the 28°-twisted sample exhibits significantly suppressed long-range exciton diffusion, thereby compromising efficient carrier transport and reducing the mobility to 40 cm²/(V·s). This angle-dependent mobility evolution provides crucial physical insights and design guidelines for developing optoelectronic devices based on exciton transport.

3 Conclusions

In summary, twist-angle engineering provides a structural route to regulate many-body Coulomb interactions in bilayer WS₂. By tuning interlayer coupling, the exciton binding energy varies from 265 to 336 meV, while the bandgap renormalization changes by more than 100 meV near the Mott transition. The same coupling also governs exciton dynamics: the strongly coupled 0° bilayer suppresses recombination, extends the slow lifetime to 103 ps, and enhances exciton mobility to 134 cm²/(V·s), nearly three times that of the 28° sample. These results link twist-controlled screening, quasiparticle renormalization, and exciton transport, offering design principles for ultrafast excitonic and optoelectronic devices.

4 Methods

4.1 Material Preparation

The WS₂ flakes used in this study were synthesized by chemical vapor deposition (CVD) on sapphire substrates. A precursor mixture consisting of 2.3 mg of tungsten trioxide powder (99.9%, Alfa Aesar), 1 g of sulfur powder (99.5%, Sigma-Aldrich), and 1 mg of NaCl (99.9%, Alfa Aesar; added as a growth promoter) was thoroughly ground. The mixture was then loaded into a ceramic boat placed at the center of the furnace. To relieve substrate-induced strain, the as-grown flakes were fabricated into bilayer WS₂ using a polystyrene (4 wt%) wet

transfer method. The flakes were detached from the sapphire substrate by etching with a 30% potassium hydroxide solution, followed by immersion in deionized water to remove residual KOH. The sample was subsequently transferred onto a monolayer WS₂ grown on a sapphire substrate and heated on a hot plate at 100°C for approximately 10 minutes. The heterostructure was then delaminated from the sapphire substrate and transferred onto a SiO₂/Si substrate following the same procedure described above. The polystyrene layer was removed by immersing the sample in acetone. Finally, to eliminate polymer residues, the transferred bilayer WS₂ samples were annealed at 150°C for 3 hours under an argon flow (500 sccm).

4.2 Measurements and Characterization

The twist angle θ of each bilayer WS₂ sample was determined by exciting isolated monolayer regions with a linearly polarized laser and rotating the polarization plane. The thickness of the samples was characterized using a Bruker Icon AFM instrument operating in tapping mode. PL spectra were acquired using a continuous-wave laser emitting at 532 nm. Raman spectra were obtained using a confocal micro-Raman spectrometer (HR Evolution, Horiba). TA measurements were performed using a 1030 nm femtosecond laser with a pulse duration of 100 fs as the fundamental driving source. A portion of this beam was focused into a YAG (yttrium aluminum garnet) crystal to generate a white-light continuum. This beam was then frequency-doubled using a beta-barium borate (BBO) crystal to produce the pump pulse. The temporal delay relative to the probe was controlled using a high-precision motorized translation stage. The remaining fundamental light was focused into a separate nonlinear crystal to generate an ultrabroadband white light probe (450nm-800 nm). Both pulses were focused onto the sample using a microscope equipped with a 50× transmission objective to ensure spatial overlap at the focal plane. The transmitted probe signal was collected and analyzed using a linear array detector (TA-100, Time-Tech Spectra, LLC). For the TAM test, the pump light wavelength was set to 505 nm and the detection light wavelength was set to 630 nm.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. Z. L.: Conceptualization, data curation, formal analysis, funding acquisition, investigation, methodology, writing original draft, reviewing and editing manuscript. Y. S. G. and J. L. C.: Data curation, investigation, methodology, writing original draft, reviewing and editing manuscript. C. X. Y.: Investigation. Q. B. W.: Investigation. W. X.: Methodology. W. Z. L.: Conceptualization. H. Y. X.: Conceptualization, funding acquisition, project administration, resources, software, supervision, reviewing and editing manuscript. Y. C. L.: Conceptualization. All the authors have approved the final manuscript.

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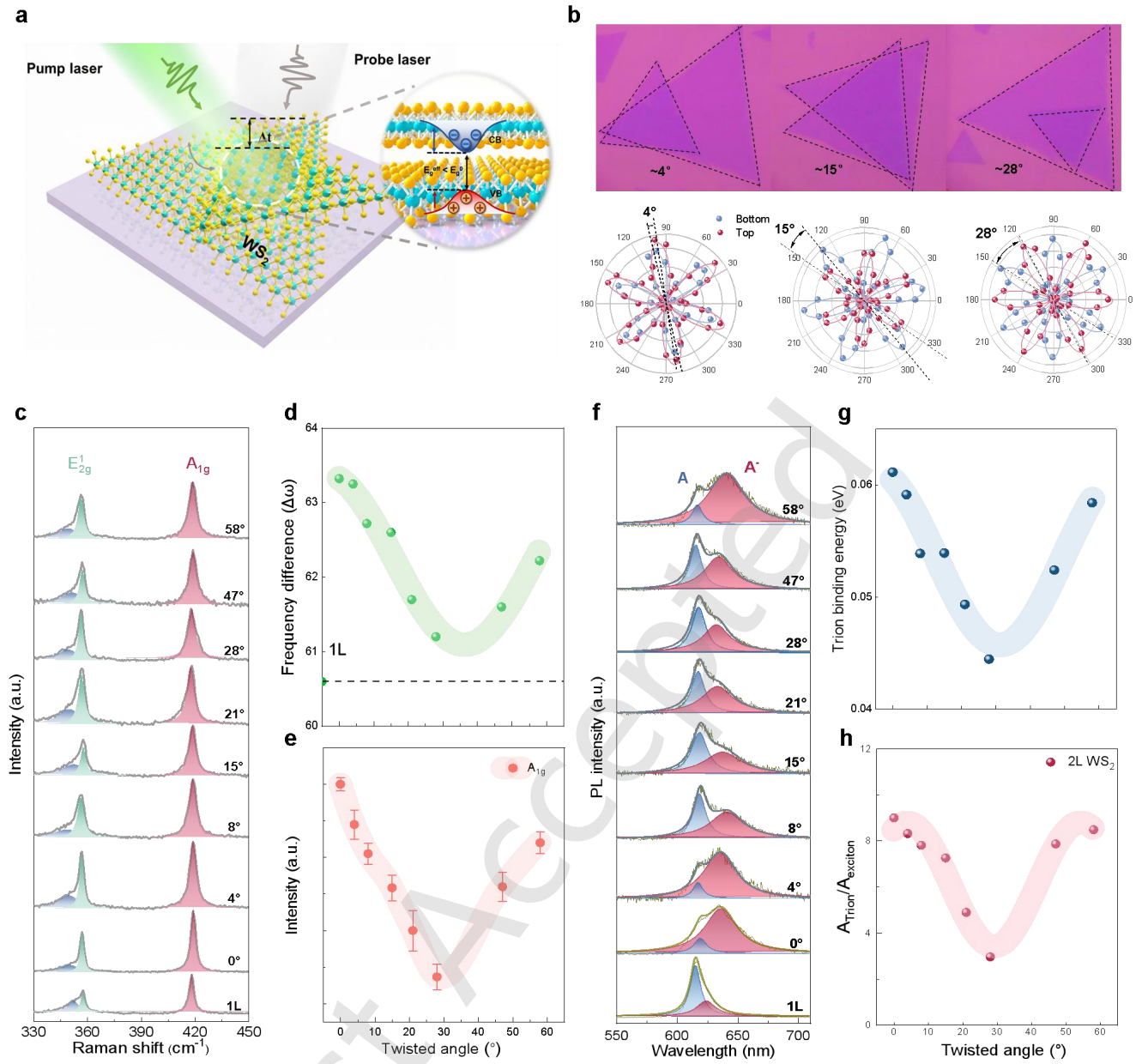


Figure 1. Structural and excitonic optical properties of twisted bilayer WS₂. (a) Schematic illustration of many-body interactions in bilayer WS₂. (b) Optical images and SHG polar plots of representative bilayers with twist angles of 4°, 15°, and 28°. (c) Raman spectra at different twist angles. (d,e) Twist-angle dependence of the Raman frequency difference and A_{1g} intensity. (f) Fitted PL spectra of twisted bilayer WS₂. (g,h) Twist-angle-dependent trion binding energy and A^-/A intensity ratio.

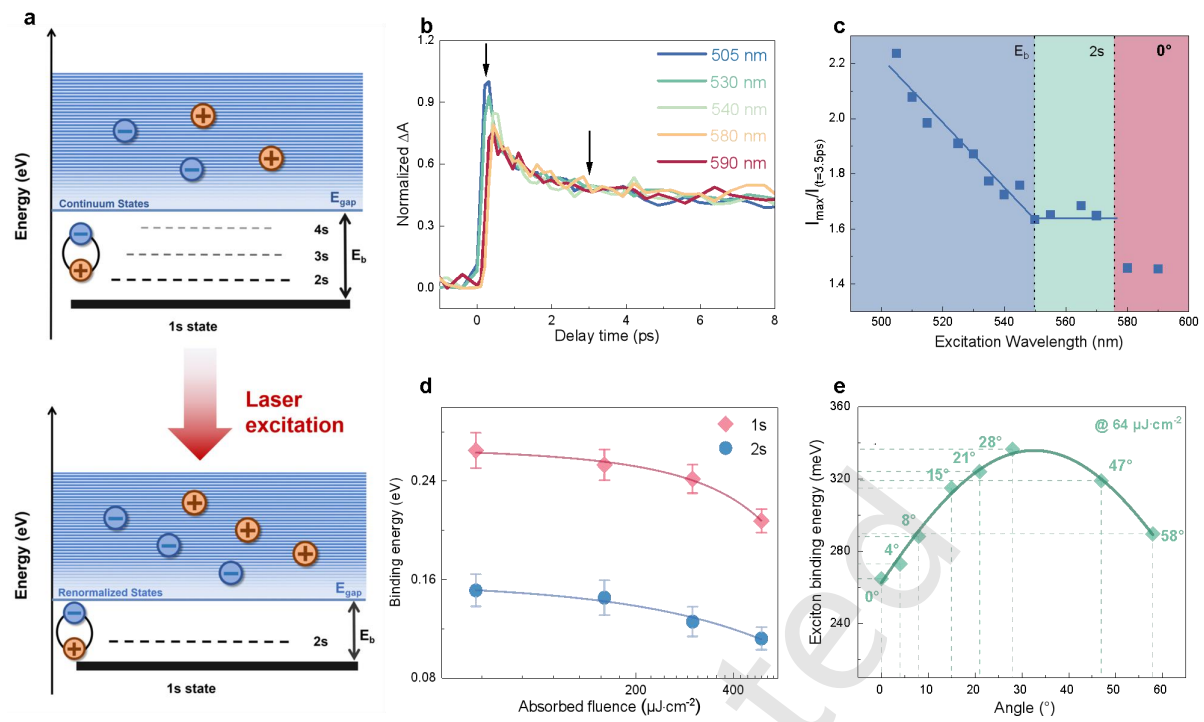


Figure 2. Photoinduced Coulomb screening and exciton binding energy in twisted bilayer WS₂. (a) Schematic evolution of Rydberg states and bandgap under low- and high-density excitation. (b) A-exciton bleach dynamics at 630 nm under different excitation wavelengths at 64 $\mu\text{J}\cdot\text{cm}^{-2}$. (c) Excitation-wavelength-dependent amplitude ratio $I_{\text{max}}/I_{t=3.5 \text{ ps}}$ for the 0° sample. (d) Fluence dependence of the 1s and 2s A-exciton states in the 0° sample. (e) Twist-angle dependence of the A-exciton binding energy at 64 $\mu\text{J}\cdot\text{cm}^{-2}$.

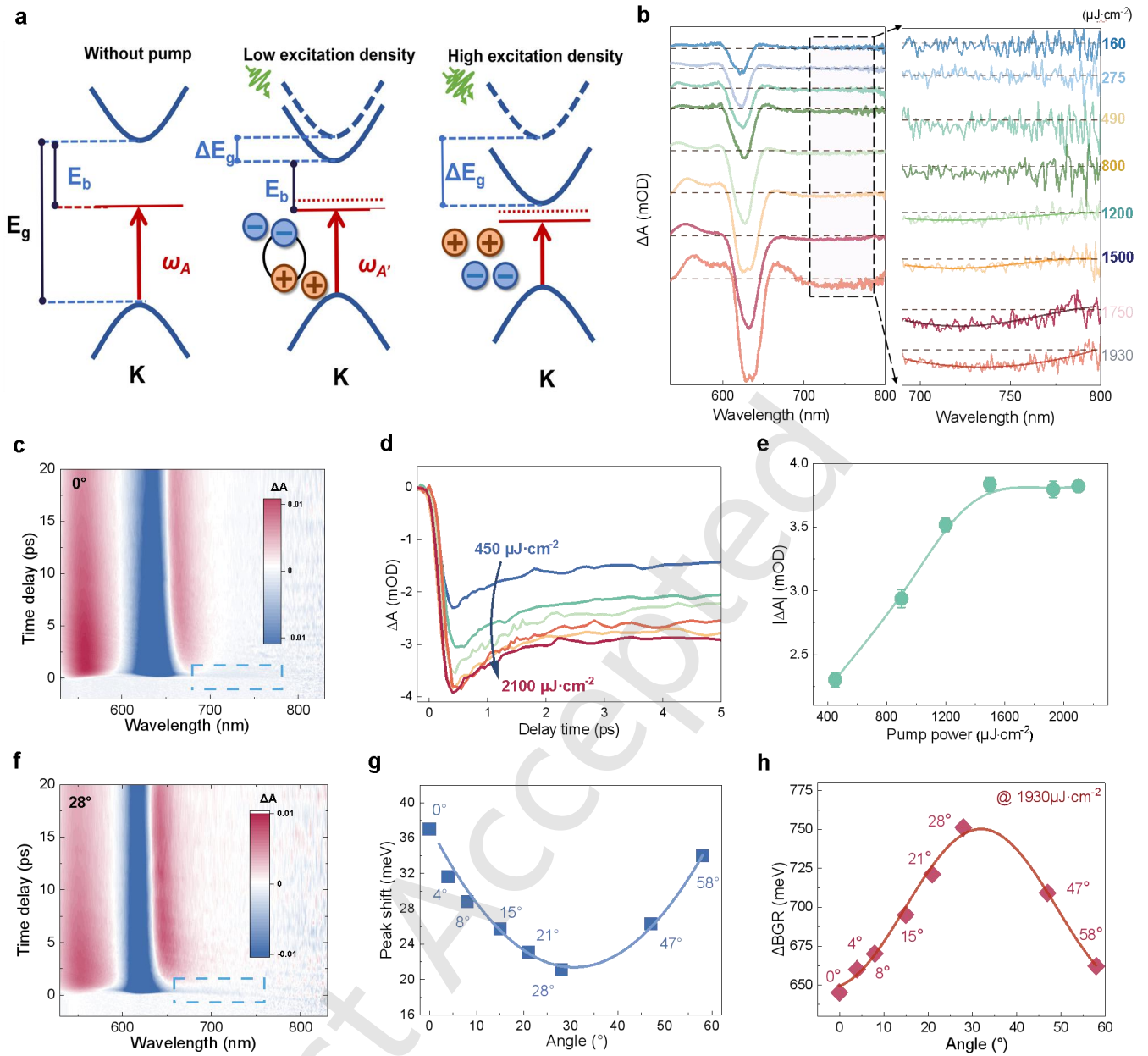


Figure 3. Photoinduced bandgap renormalization in twisted bilayer WS₂. (a) Schematic of bandgap renormalization under increasing excitation density. (b) TA spectra of the 0° bilayer at different pump fluences. (c) Pseudocolor TA map of the 0° sample at 1930 $\mu\text{J}\cdot\text{cm}^{-2}$. (d,e) Fluence-dependent A-exciton bleaching dynamics and saturation behavior. (f) Pseudocolor TA map of the 28° sample. (g,h) Twist-angle-dependent A-exciton redshift and BGR magnitude.

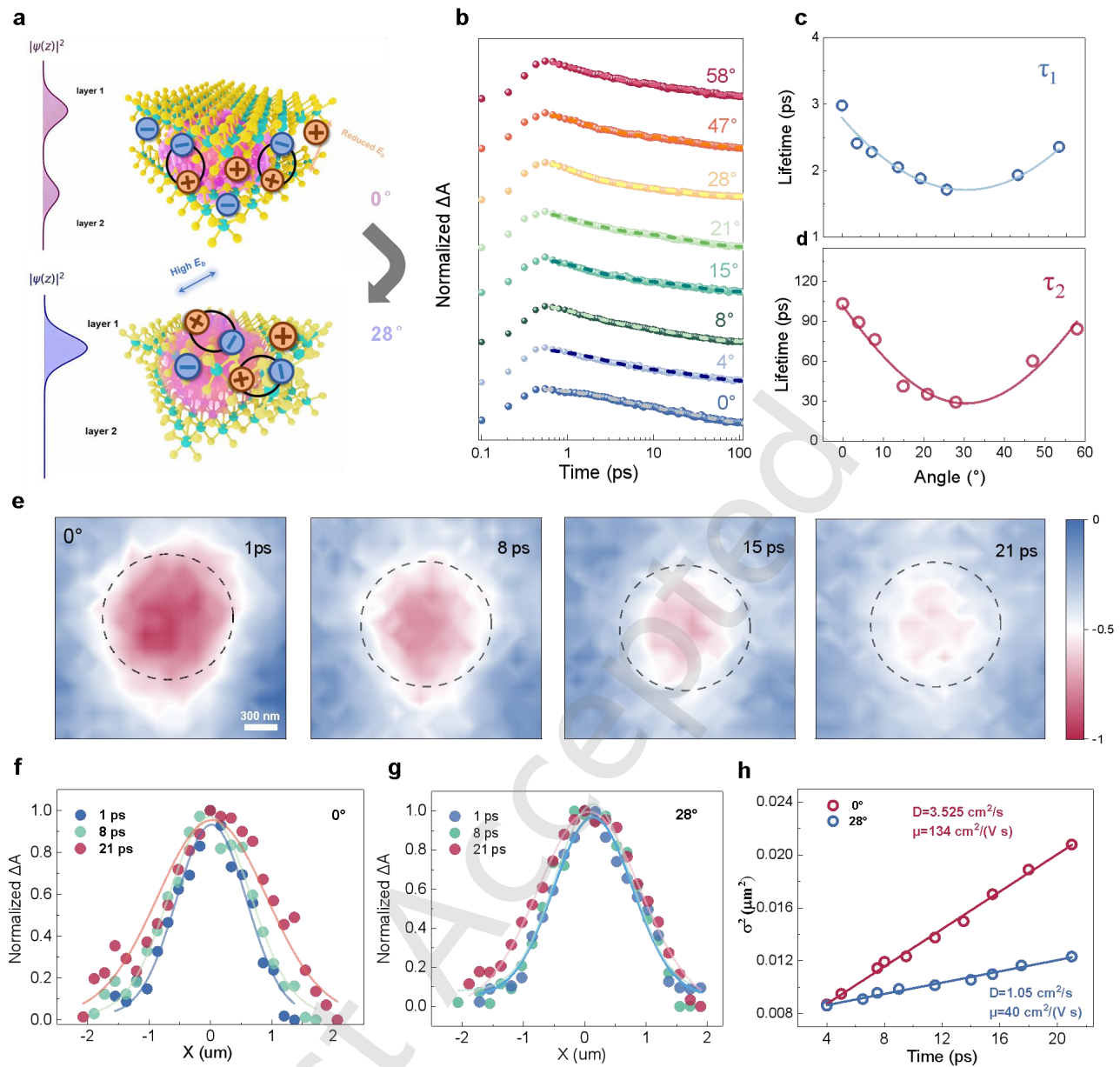


Figure 4. Twist-angle-dependent exciton dynamics and transport in bilayer WS₂. (a) Schematic illustration of exciton delocalization at different twist angles. (b) Twist-angle-dependent A-exciton bleach decay curves under 800 $\mu\text{J}\cdot\text{cm}^{-2}$ excitation. (c,d) Fast and slow lifetime components extracted from double-exponential fitting. (e) Temporal evolution of the TAM images. (f-g) Normalized spatial TA profiles at different delay times for the 0° and 28° samples, respectively, together with Gaussian fits. The pump power density is 160 $\mu\text{J}\cdot\text{cm}^{-2}$. (h) Time-dependent variance extracted from Gaussian fitting, yielding exciton diffusion coefficients and mobilities.

Electronic Supplementary Material

Twist-angle tunable bandgap renormalization and exciton binding energy in WS₂ bilayers

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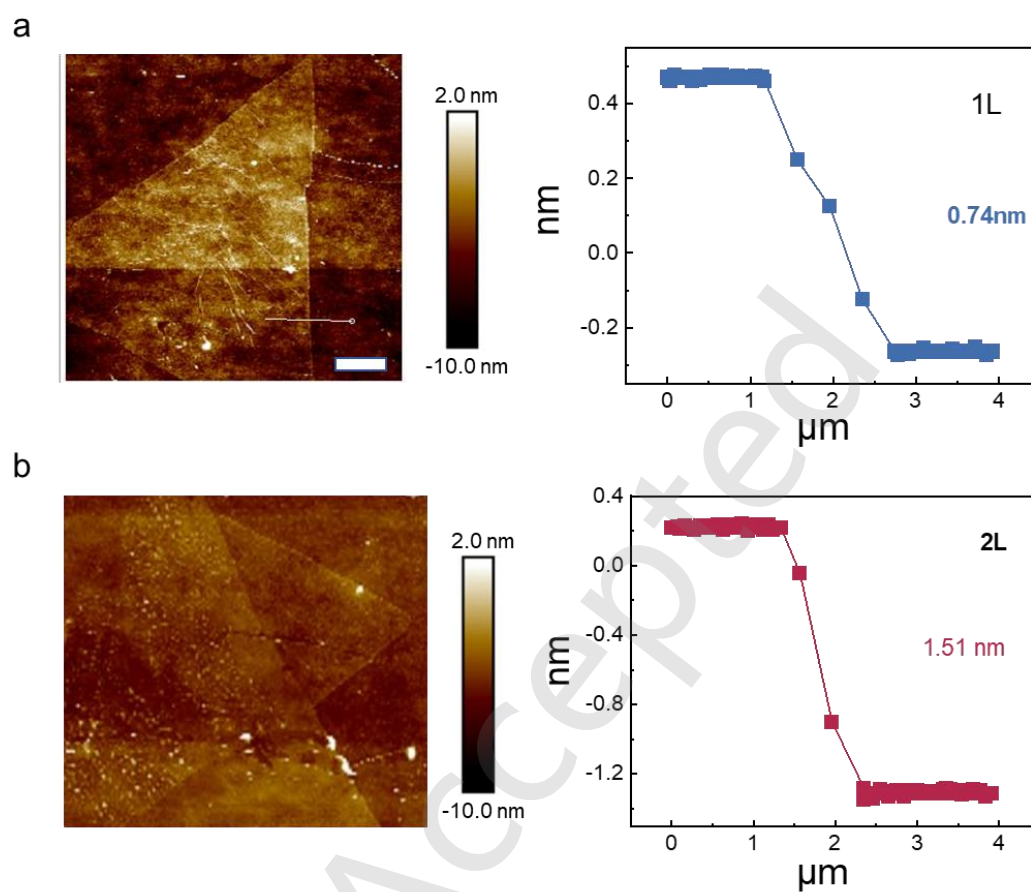


Figure S1. AFM images of (a) 1L and (b) 2L WS₂ nanosheets, together with height profiles taken along the white scan lines. Scale bar: 2 μm.

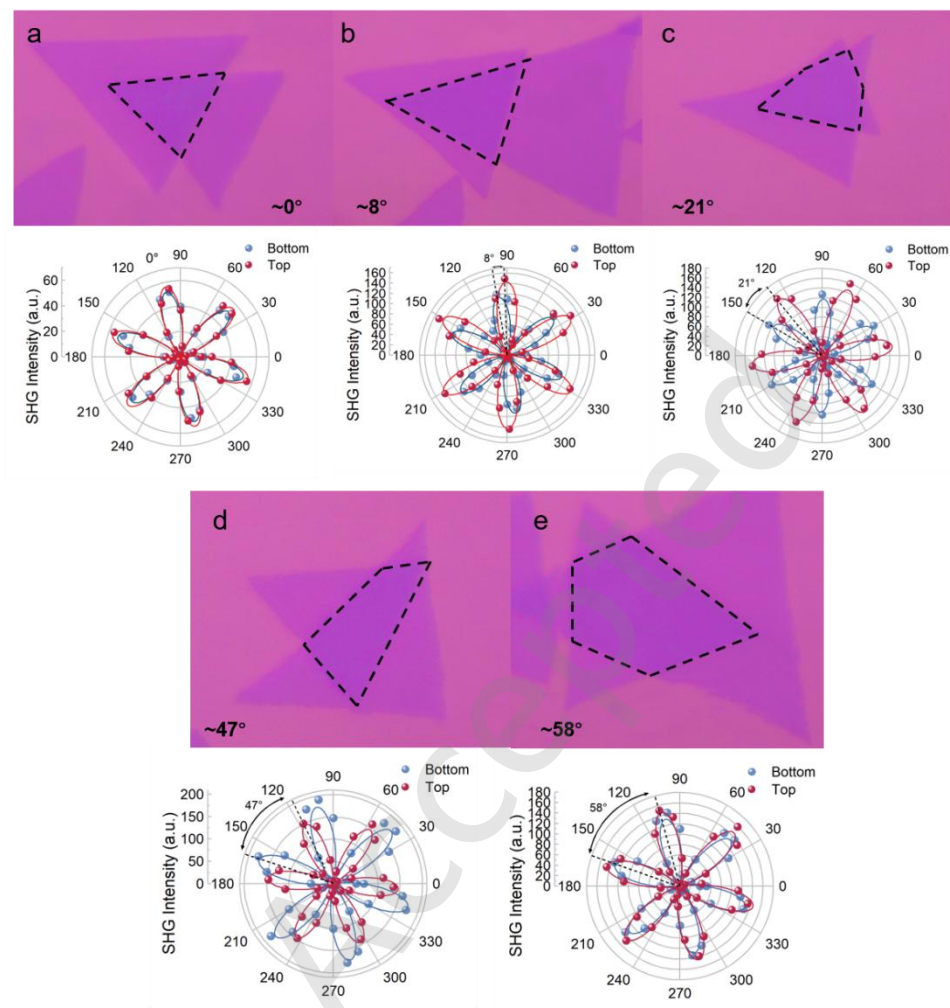


Figure S2. Optical microscopy images and polarization-resolved SHG characterizations of additional twisted bilayer WS2 samples.

Optical microscopy images and corresponding polarization-resolved SHG intensity mappings of bilayer WS2 samples with twist angles of (a) 0° , (b) 8° , (c) 21° , (d) 47° , and (e) 58° . The crystallographic orientations of the individual WS2 layers were determined from the polarization-dependent SHG patterns, and the relative orientations between the two layers were used to verify the corresponding twist angles.

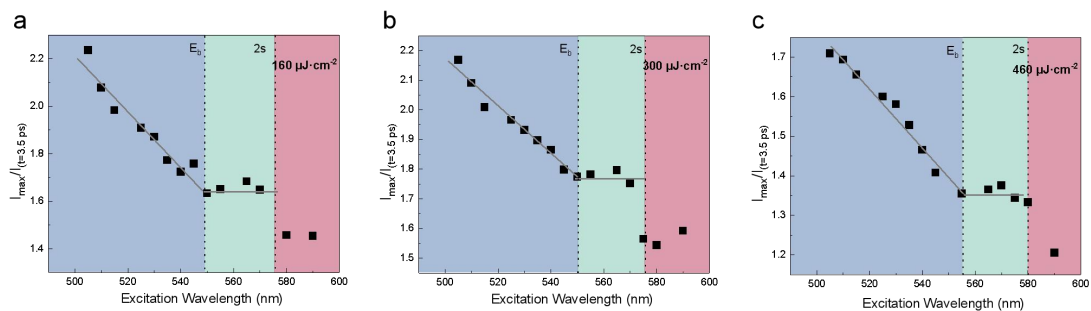


Figure S3. Excitation-wavelength dependence of the transient absorption (TA) signal amplitude ratio, $I_{\max}/I_{(+3.5 \text{ ps})}$, for the 0° twisted sample at pump fluences of (a) $160 \mu\text{J}\cdot\text{cm}^{-2}$, (b) $300 \mu\text{J}\cdot\text{cm}^{-2}$, and (c) $460 \mu\text{J}\cdot\text{cm}^{-2}$.

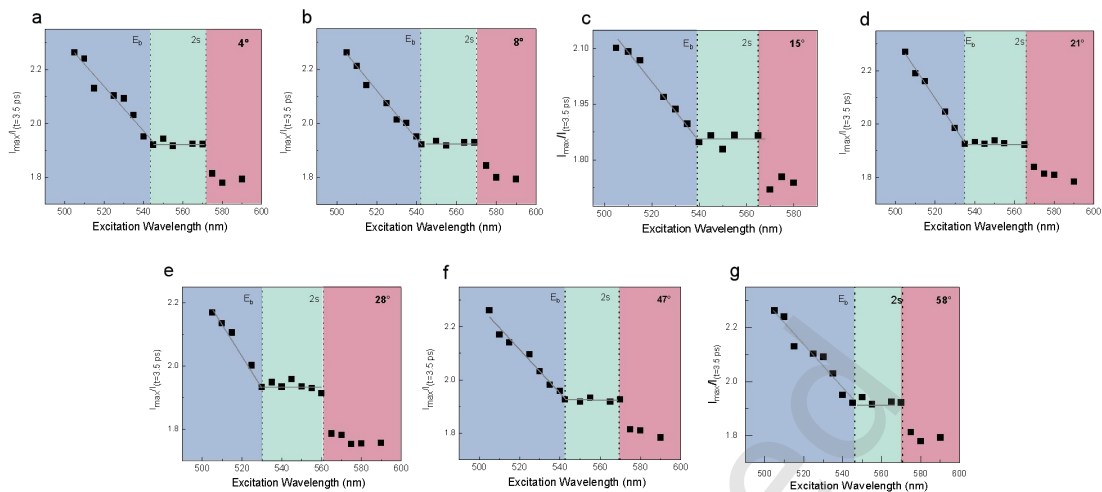


Figure S4. Excitation-wavelength dependence of the amplitude ratio ($I_{\max} / I_{(+3.5 \text{ ps})}$) of the TA signal for bilayer WS₂ samples at twist angles of 4°, 8°, 15°, 21°, 28°, 47°, and 58°.

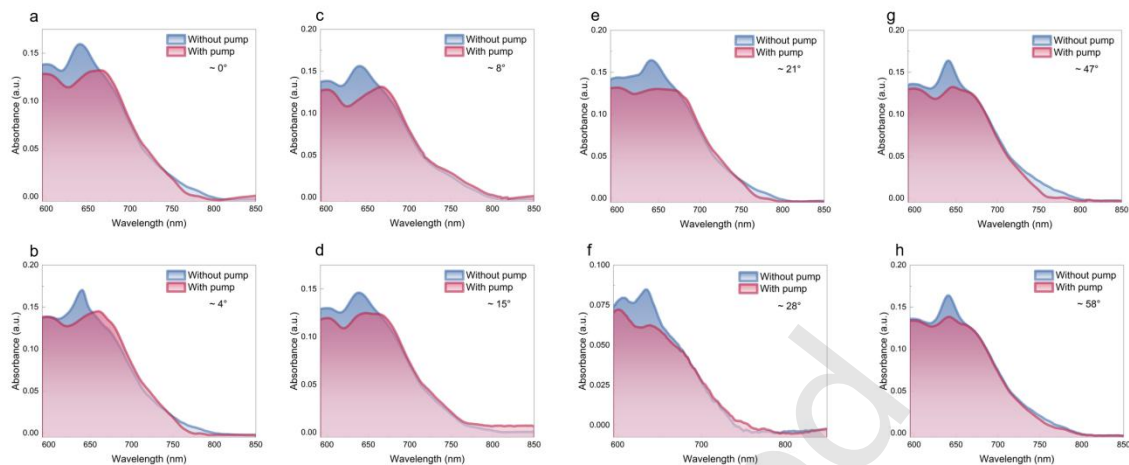


Figure S5. Absorbance spectra without (blue) and with (red) pump excitation for bilayer WS₂ with various twist angles. The A-exciton resonance is strongly suppressed at a pump fluence of 1930 $\mu\text{J}\cdot\text{cm}^{-2}$, revealing pronounced photoinduced exciton screening across different twist angles.

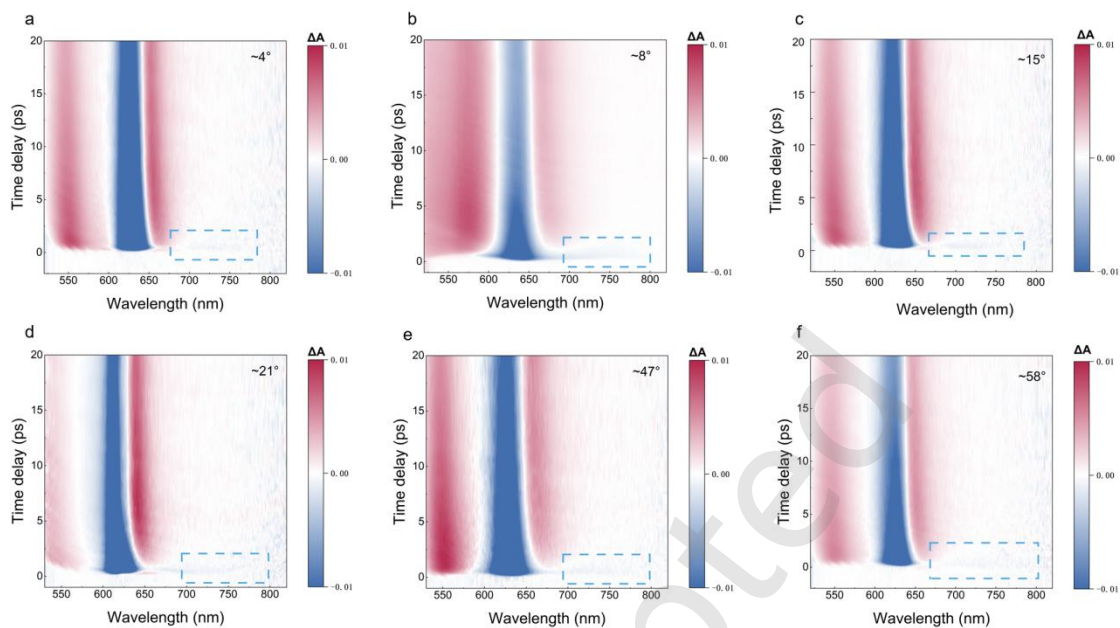


Figure S6. Pseudocolor maps of the TA signal as a function of time delay and wavelength for samples with twist angles of (a) 4° , (b) 8° , (c) 15° , (d) 21° , (e) 47° , and (f) 58° .

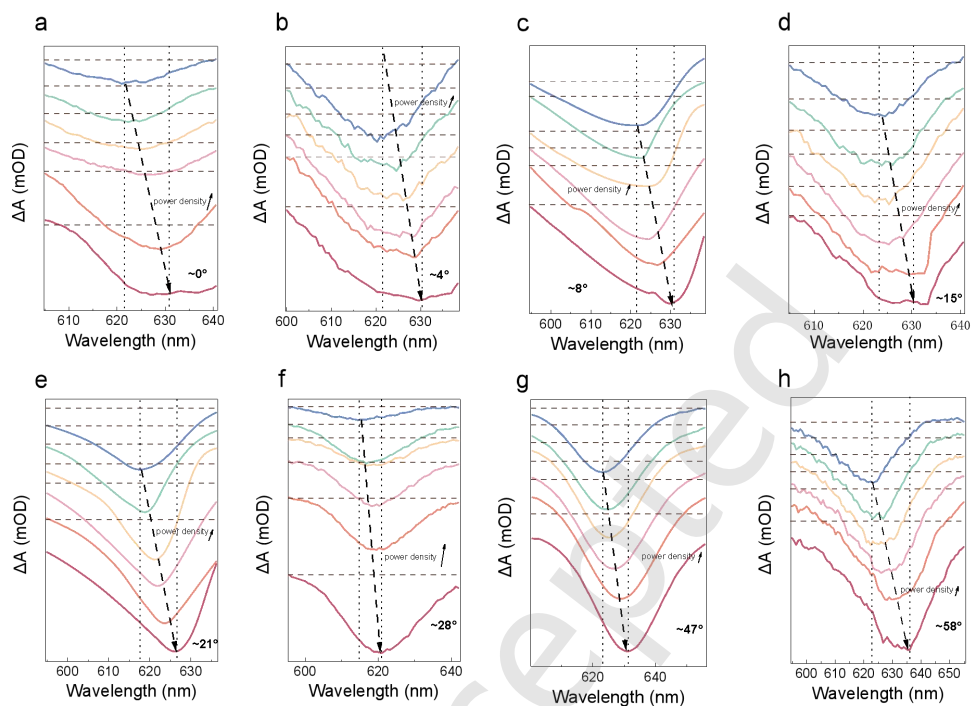


Figure S7. (a–h) Representative fluence-dependent TA spectra and fitted A-exciton peak positions for bilayer WS₂ samples with twist angles of 0°, 4°, 8°, 15°, 21°, 28°, 47°, and 58°, respectively. The A-exciton resonance energies were extracted by fitting the exciton bleaching peaks at different pump fluences.

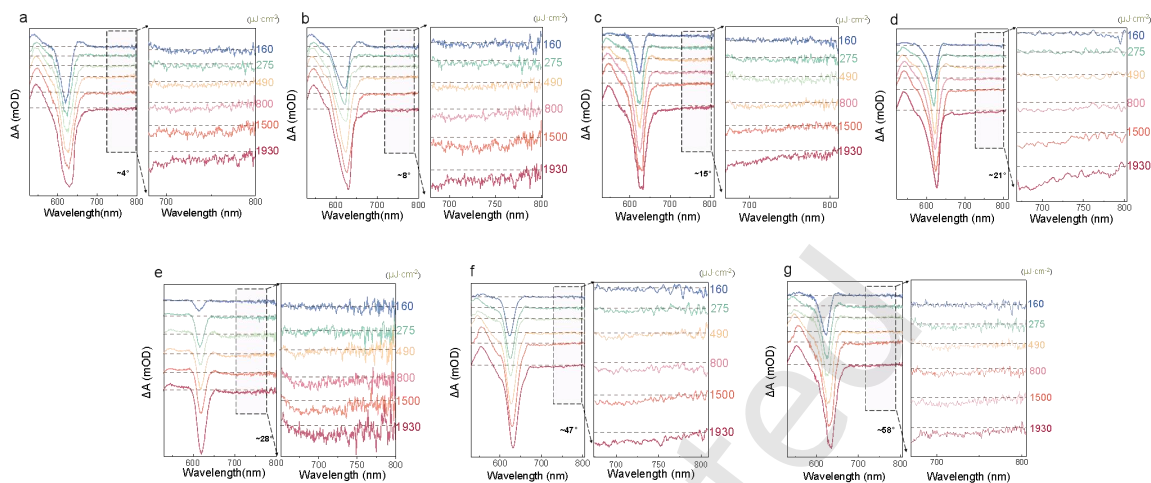


Figure S8. TA spectra of bilayer WS₂ under pump fluences of 160–1930 $\mu\text{J}\cdot\text{cm}^{-2}$ at a time delay of 0.35 ps. The low-energy population inversion region is highlighted by a rectangle.

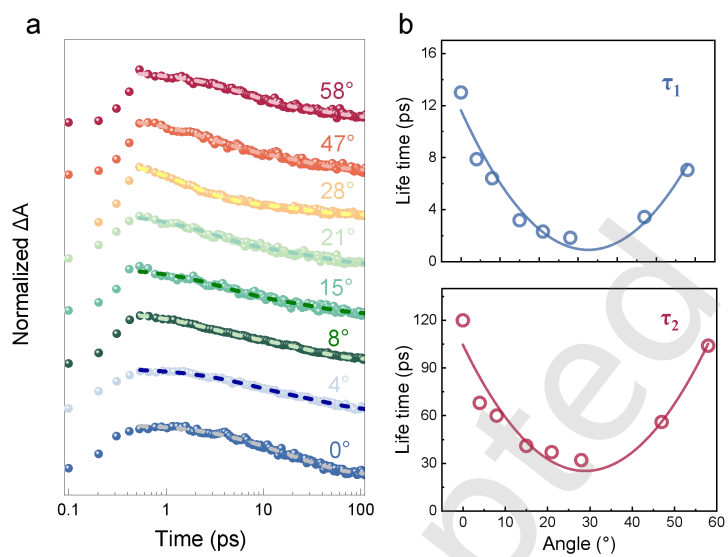


Figure S9. (a) Bleaching signal attenuation as a function of twist angle at a pump fluence of $160 \mu\text{J}\cdot\text{cm}^{-2}$. (b) Lifetime components τ_1 and τ_2 extracted from double-exponential fitting, corresponding to fast and slow exciton recombination, respectively.

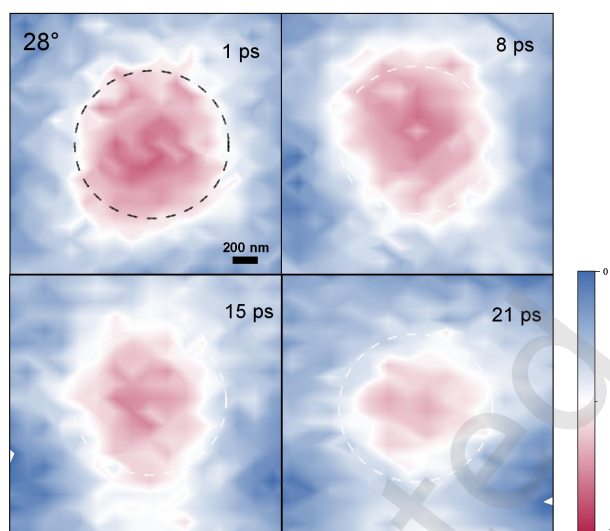


Figure S10. Temporal evolution of transient absorption microscopy (TAM) images for the 28° twisted sample.

Note 1: Details for exciton binding energy measurements

The exciton binding energy, E_b , is defined as the energy required to dissociate a bound electron-hole pair into free carriers. In optical measurements, E_b can be extracted by identifying the onset of continuum states relative to the excitonic resonance[1]. Specifically, the excess photon energy is defined as follows:

$$\Delta E = h\nu - E_A$$

where $h\nu$ is the excitation energy and E_A is the 1s exciton resonance. Experimentally, this threshold can be extracted by fitting a spectroscopic observable (e.g., TA amplitude ratio) with a piecewise function:

$$S(h\nu) = \begin{cases} a, & h\nu < E_b \\ a + b(h\nu - E_b), & h\nu \geq E_b \end{cases}$$

where a and b are fitting parameters, and $h\nu$ is the photon energy. The breakpoint of the fit yields E_b . This approach is consistent with the physical picture in which excitonic states lie below the continuum edge, with the binding energy corresponding to their separation[2]. The threshold method therefore provides a direct, experimentally accessible route and is particularly well suited for low-dimensional systems exhibiting strong Coulomb interactions.

Note 2: Determination of bandgap renormalization

The bandgap renormalization (BGR) magnitude was experimentally extracted from the transient absorption spectra. Unlike phenomenological models based on carrier-density-dependent fitting, the present approach directly determines the photoinduced quasiparticle bandgap shift from the spectral evolution.

The equilibrium quasiparticle bandgap E_g^0 is related to the optical exciton transition energy E_A and exciton binding energy E_b :

$$E_g^0 = E_A + E_b$$

where E_A is obtained from the A-exciton resonance position and E_b is extracted using the excitation-wavelength-dependent TA threshold method described in Note 1. Under high-density photoexcitation, many-body Coulomb screening induces bandgap renormalization. The transient quasiparticle bandgap E_g^* is determined from the onset of the low-energy population inversion feature in the TA spectra (Figure 3c and Figure S6).

Therefore, the BGR magnitude is calculated as:

$$\Delta E_{BGR} = E_g^0 - E_g^*$$

For example, for the 0° twisted bilayer WS₂ near the Mott transition density, $E_A = 1.95$ eV and $E_b = 265$ meV, giving $E_g^0 = 2.215$ eV. The renormalized bandgap extracted from the TA spectrum is $E_g^* = 1.57$ eV, resulting in:

$$\Delta E_{BGR} = 2.215 - 1.57 = 645 \text{ meV}$$

This experimental extraction method is consistent with previous studies of photoinduced BGR in atomically thin TMDs, where transient optical spectroscopy was used to directly monitor the quasiparticle bandgap renormalization[3-5].

Note 3: Conversion from pump fluence to photoexcited carrier density

The photoexcited carrier density was estimated from the incident pump fluence according to:

$$n = \frac{F(1-R)A}{h\nu}$$

where n is the generated photoexcited carrier density, F is the incident pump fluence, R is the reflection coefficient of bilayer WS₂, A is the optical absorption of bilayer WS₂ at the pump wavelength, and $h\nu$ is the photon energy. For the excitation condition used in this work, the pump wavelength was 505 nm, corresponding to a photon energy of:

$$h\nu = \frac{hc}{\lambda} = 2.45 \text{ eV}$$

and the maximum pump fluence applied near the Mott transition was:

$$F = 1930 \mu\text{J cm}^{-2} = 1.93 \times 10^{-3} \text{ J cm}^{-2}$$

The corresponding photoexcited carrier density is calculated to be:

$$n \approx 2.45 \times 10^{14} \text{ cm}^{-2}$$

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