

Supporting Information

Generalizable growth of 2D twisted transition metal dichalcogenide heterostructures under non-equilibrium thermodynamic conditions

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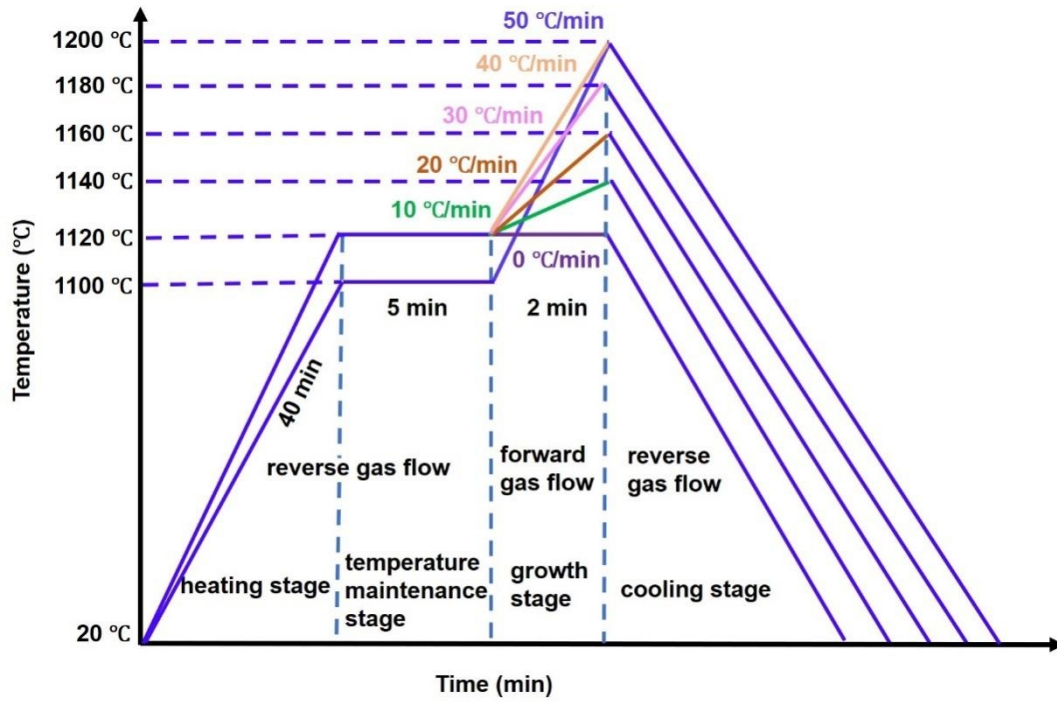
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Supplementary Note 1: Yield and density calculation

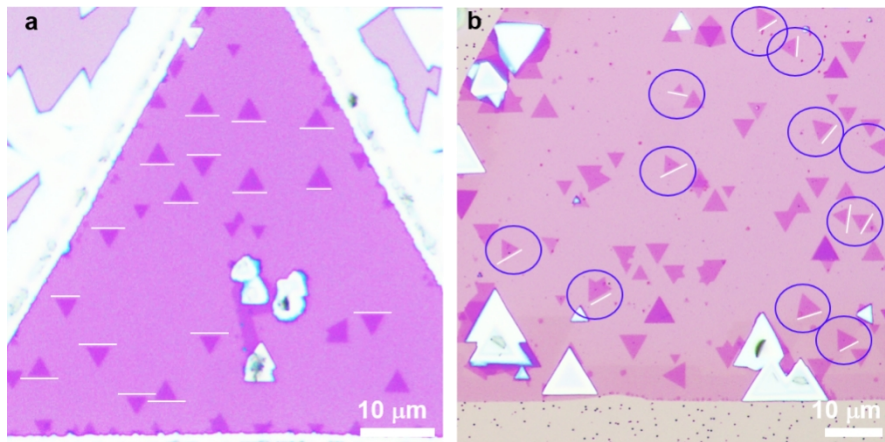
All yield and density data were obtained from at least three independent and repeated

experiments. For each experiment, no fewer than five representative fields of view were randomly selected on the substrate for statistical analysis. The OM under the 50X objective was selected due to the twisted bilayers heterostructures can be easily identified under the 50X objective. The number of twisted bilayers heterostructures and vertical heterostructures was calculated based on OM images under a 50X objective lens. The yield (Y) and density (D) can be determined using the total number of twisted samples (N_{twisted}), the total number of vertical heterostructure samples ($N_{\text{untwisted}}$), and the total substrate area (S).

$$Y = (N_{\text{twisted}} / (N_{\text{twisted}} + N_{\text{untwisted}})) \times 100\% \quad D = N / S.$$

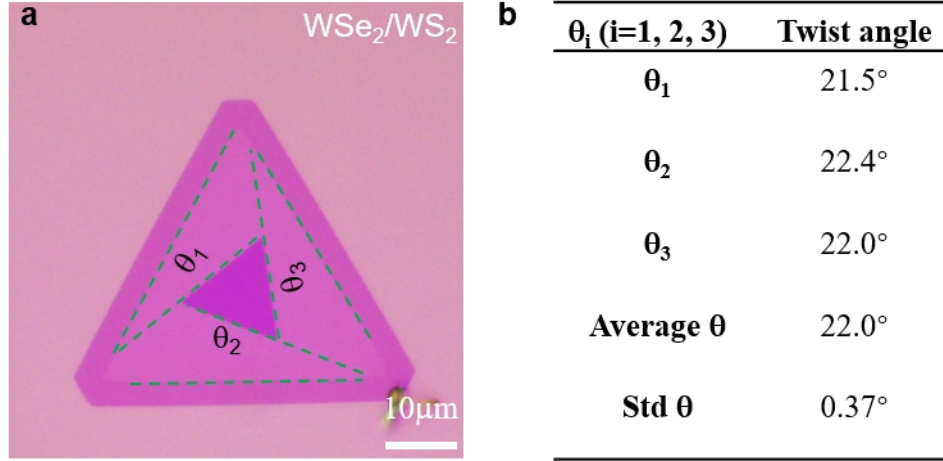


Supplementary Figure 1. The temperature rising process in our CVD synthesis strategy.

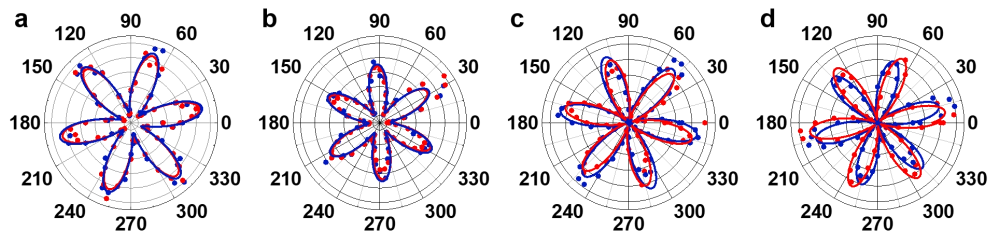


Supplementary Figure 2. The growth of WSe₂/WS₂ heterostructure. a, Optical microscope

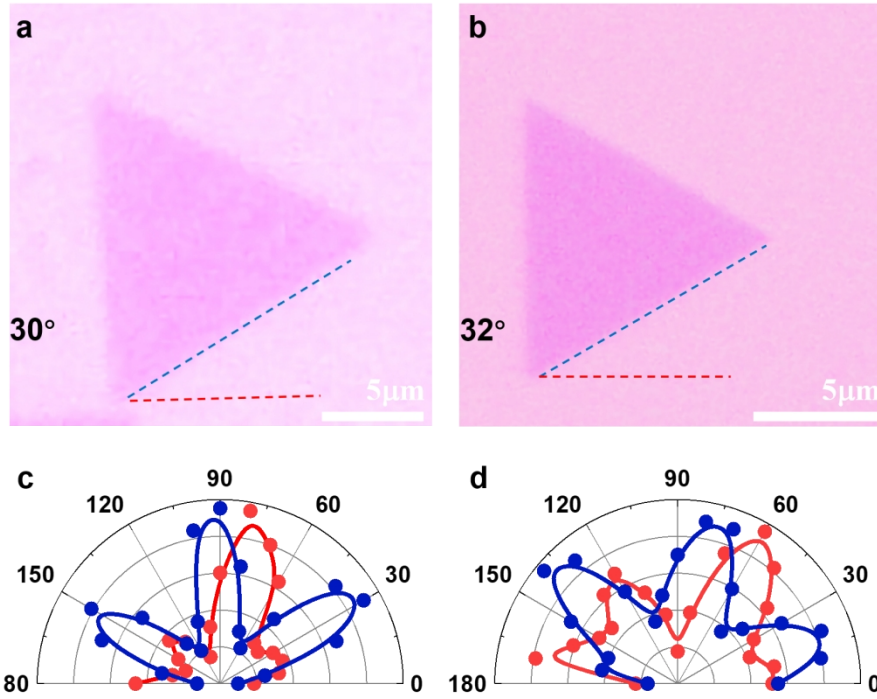
(OM) images of WSe₂/WS₂ heterostructures synthesized by conventional isothermal CVD process. **b**, OM images of WSe₂/WS₂ heterostructures synthesized by temperature continuously rising CVD process.



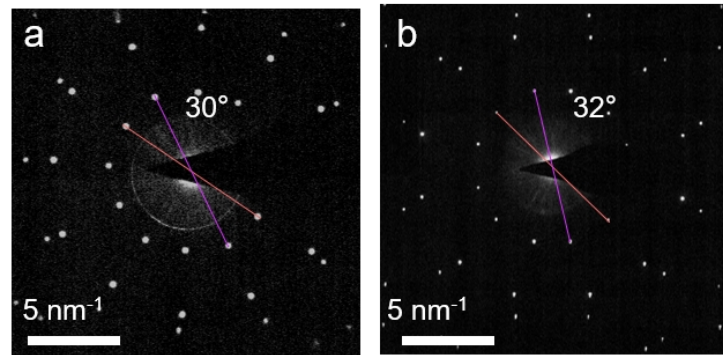
Supplementary Figure 3. Determination of the twist angle in heterostructures. **a**, The typical OM image of TB-WSe₂/WS₂. **b**, The measured twist angles based on the OM from the TB-WSe₂/WS₂ in (a).



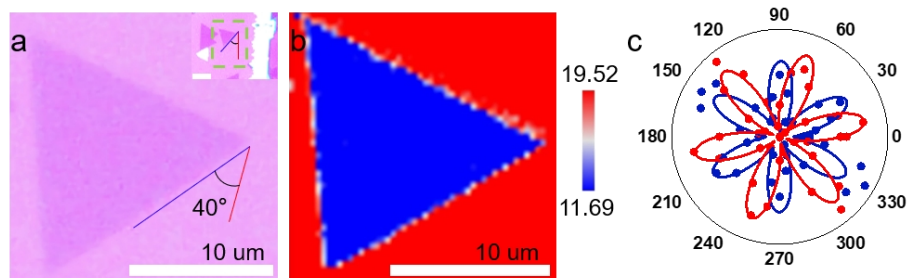
Supplementary Figure 4. Polar plots of normalized SHG intensities for TB-WSe₂/WS₂ heterostructures. **a-d**, Polar plots of normalized SHG intensities for 0°, 3.0°, 8.0°, and 15.0°-TB-WSe₂/WS₂ heterostructures, respectively.



Supplementary Figure 5. SHG characterization of twisted WSe₂/WS₂ heterostructure. a, OM images of 30° and 32°-TB-WSe₂/WS₂ heterostructures. c, d, Polar plots of normalized SHG intensities for 30° and 32°-TB-WSe₂/WS₂, respectively.

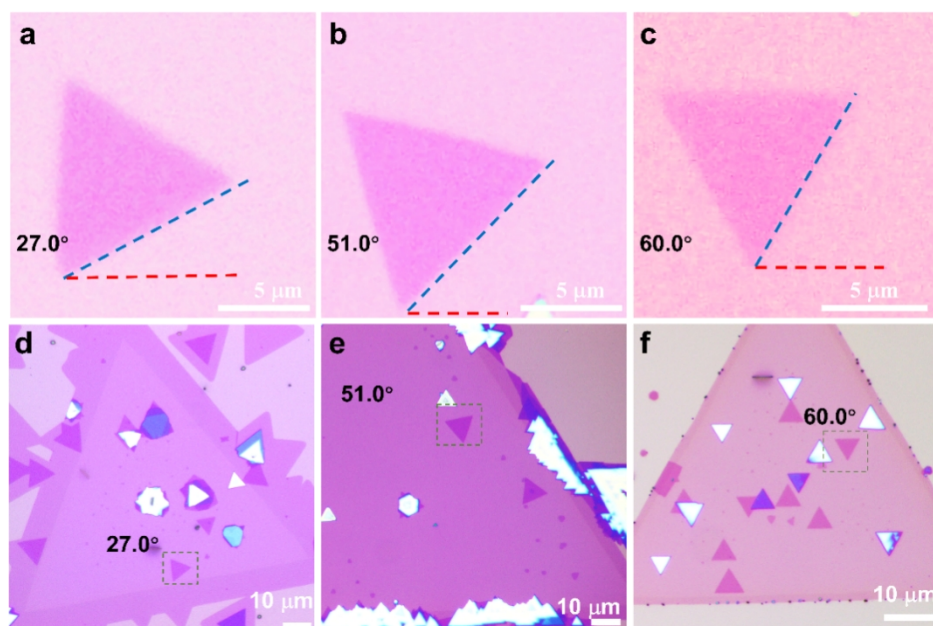


Supplementary Figure 6. SAED characteristic of twisted WSe₂/WS₂ heterostructures. a-b, The SAED patterns of 30.0°, and 32.0°-TB-WSe₂/WS₂. The pink and purple hexagons mark the diffraction patterns from WSe₂ and WS₂ lattices, respectively.

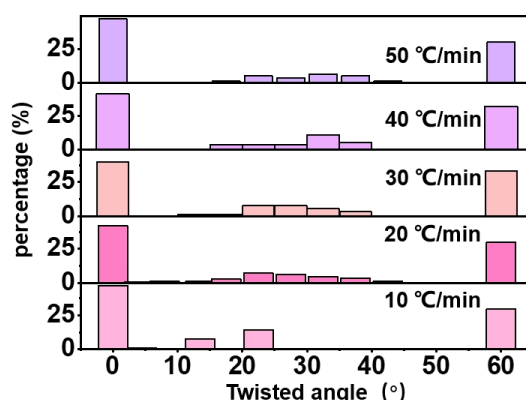


Supplementary Figure 7. The SHG measurements in twisted WSe₂/WS₂ Heterostructure.

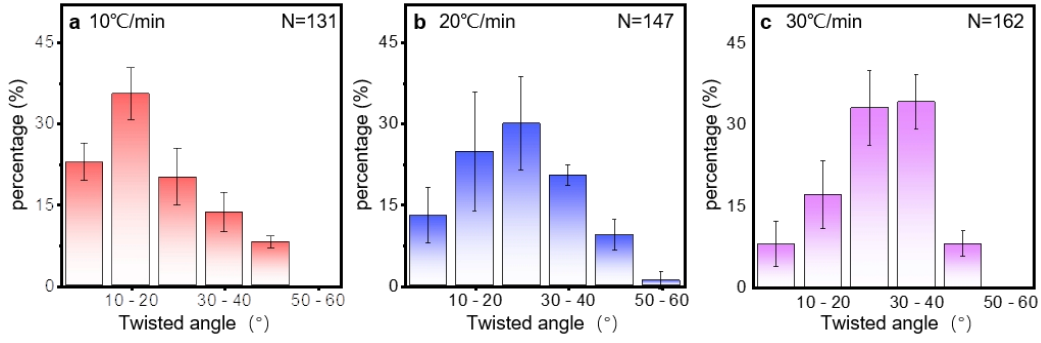
a, The OM image of the 40°-TB-WSe₂/WS₂ heterostructure, the WS₂ and WSe₂ regions with red and blue dashed outlines, respectively. The inset presents an overview of the 40° -twisted-WSe₂/WS₂ heterostructure. **b**, The SHG mapping of the area in (a), with the color bar unit in CCD cts. **c**, Polar plots of normalized SHG intensities for 40°-TB-WSe₂/WS₂.



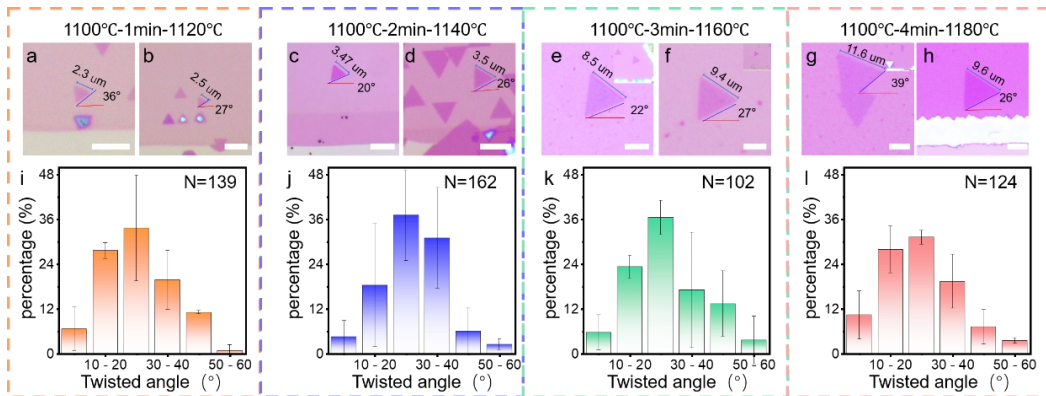
Supplementary Figure 8. Optical image of twisted WSe₂/WS₂ heterostructures. a-c, OM images of TB-WSe₂/WS₂ heterostructures with 27°, 51°, 60° twist angles. d-f, The original OM images corresponding to the (a-c).



Supplementary Figure 9. Angle distribution statistics of TB-WSe₂/WS₂ heterostructures at heating rate of 10 °C/min, 20 °C/min, 30 °C/min, 40 °C/min and 50 °C/min, including 0° and 60°.

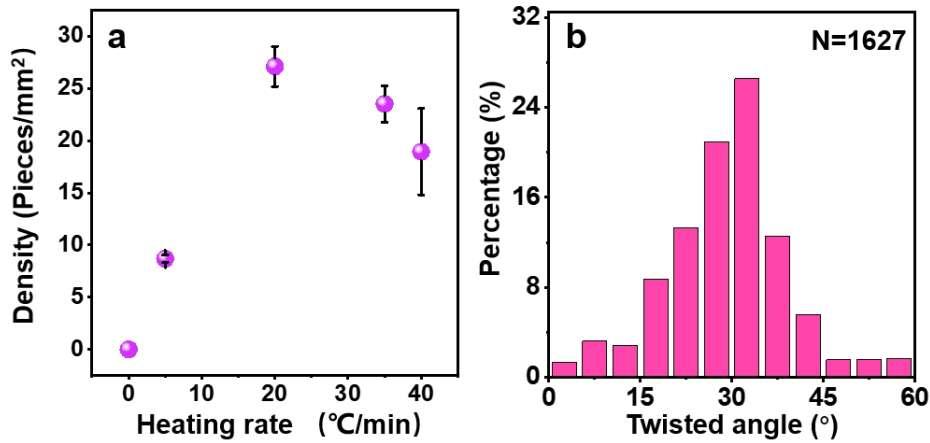


Supplementary Figure 10. Statistical charts of twist angles in WSe_2/WS_2 heterostructures under different heating rates: a, under a heating rate of $10^\circ\text{C}/\text{min}$. b, under a heating rate of $20^\circ\text{C}/\text{min}$. c, under a heating rate of $30^\circ\text{C}/\text{min}$. All data plots are derived from at least three independent and repeatable experiments. For each experiment, no fewer than five representative fields of view were randomly selected for statistical analysis. The error bars (representing the standard deviation (s.d.)) are derived from three independent experiments at the corresponding heating rates. The error bars for the five sets in (a) are $22.84\% \pm 3.46\%$, $35.48\% \pm 4.79\%$, $20.06\% \pm 5.27\%$, $13.56\% \pm 3.63\%$, and $8.06\% \pm 1.09\%$, respectively. The error bars for the six sets in (b) are $12.93\% \pm 5.10\%$, $24.77\% \pm 10.96\%$, $29.96\% \pm 8.63\%$, $20.37\% \pm 1.97\%$, $9.35\% \pm 2.84\%$, and $0.95\% \pm 1.65\%$, respectively. The error bars for the five sets in (c) are $7.85\% \pm 4.17\%$, $17.04\% \pm 6.22\%$, $33.01\% \pm 6.85\%$, $34.16\% \pm 5.07\%$, and $7.93\% \pm 2.33\%$, respectively.



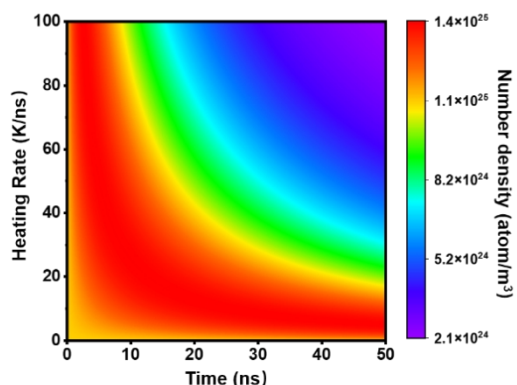
Supplementary Figure 11. Statistical charts of twist angles in WSe_2/WS_2 heterostructures across different growth temperature ranges. a-h, Heating rate: $20^\circ\text{C}/\text{min}$, with temperature ranges: $1100 \rightarrow 1120^\circ\text{C}$ (1min) (a, b), $1100 \rightarrow 1140^\circ\text{C}$ (2min) (c, d),

1100→1160 °C (3min) (e, f), 1100→1180 °C (4min) (g, h). The scale bar is 5 μm . i-l, i, for 1 min starting from 1100°C with a final temperature of 1120°C. j, for 2 min starting from 1100°C with a final temperature of 1140°C. k, for 3 min starting from 1100°C with a final temperature of 1160°C. l, for 4 min starting from 1100°C with a final temperature of 1180°C. All data plots are derived from at least three independent and repeatable experiments. For each experiment, no fewer than five representative fields of view were randomly selected for statistical analysis. The error bars (representing the standard deviation (s.d.)) are derived from three independent experiments at the corresponding heating rates. The error bars for the six sets in (i) are 6.70% $\pm$ 5.85%, 27.70% $\pm$ 2.08%, 33.69% $\pm$ 14.18%, 19.83% $\pm$ 7.97%, 11.18% $\pm$ 0.41% and 0.90% $\pm$ 1.56%, respectively. The error bars for the six sets in (j) are 4.54% $\pm$ 4.38%, 18.44% $\pm$ 16.48%, 37.19% $\pm$ 12.18%, 31.08% $\pm$ 13.57%, 6.15% $\pm$ 6.10%, and 2.59% $\pm$ 1.42%, respectively. The error bars for the six sets in (k) are 5.76% $\pm$ 4.70%, 23.36% $\pm$ 3.15%, 36.60% $\pm$ 4.60%, 17.13% $\pm$ 15.51%, 13.44% $\pm$ 8.79% and 3.70% $\pm$ 6.42%, respectively. The error bars for the six sets in (l) are 10.44% $\pm$ 6.42%, 28.01% $\pm$ 6.27%, 31.28% $\pm$ 1.85%, 19.45% $\pm$ 7.23%, 7.25% $\pm$ 4.61% and 3.57% $\pm$ 0.77%, respectively.

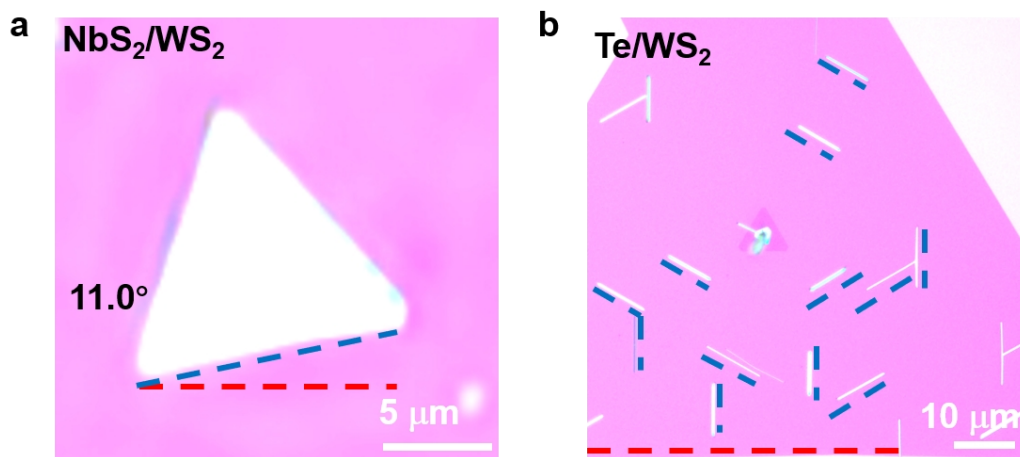


Supplementary Figure 12. Statistical analysis of density and twist angle distribution in WSe₂/WS₂ heterostructures under different heating rates. a, Statistical chart of the density of TB-WSe₂/WS₂ heterostructures under different heating rates. The density is defined as the number of TB-WSe₂/WS₂ heterostructures per unit area. The error bars (representing the standard deviation (s.d.)) are derived from four independent experiments at the corresponding

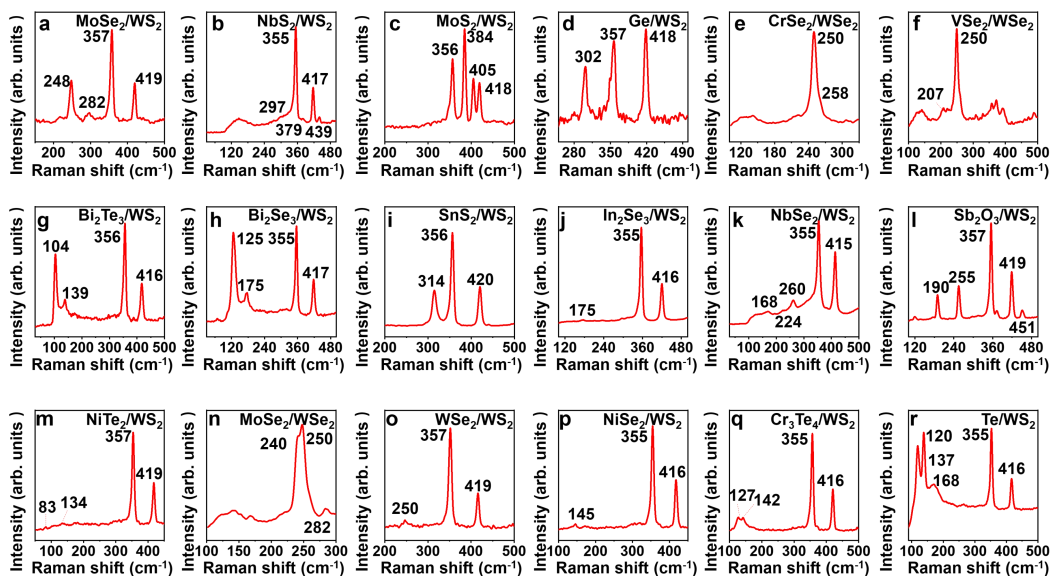
heating rates. The error bars for the five sets in (a) are 0.00 ± 0.00 pieces/mm², 8.67 ± 0.37 pieces/mm², 27.11 ± 1.92 pieces/mm², 23.5 ± 1.73 pieces/mm², and 18.94 ± 4.15 pieces/mm², respectively. **b**, Statistical chart of the angular distribution of TB-WSe₂/WS₂ heterostructures. The statistics were derived from all TB-WSe₂/WS₂ heterostructures prepared under multiple heating rates by the temperature-gradient-controlled CVD method.



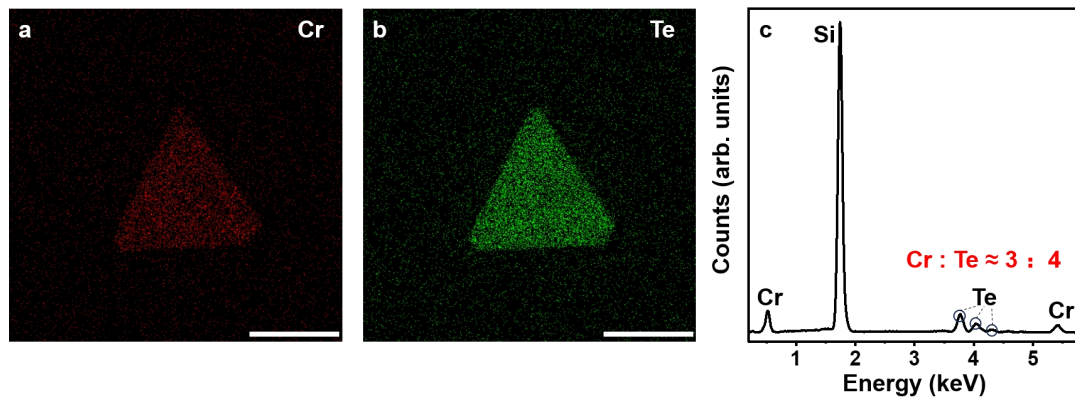
Supplementary Figure 13. 2D pseudocolor mapping of gas molecular density variation under different heating rates within the same time interval.



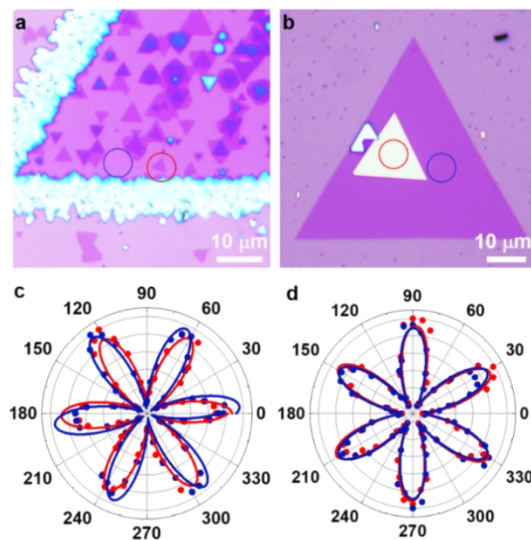
Supplementary Figure 14. OM images of twisted TMD vdW heterostructures. a, OM images of twisted NbS₂/WS₂ heterostructure. **b**, OM images of twisted Te/WS₂ heterostructures.



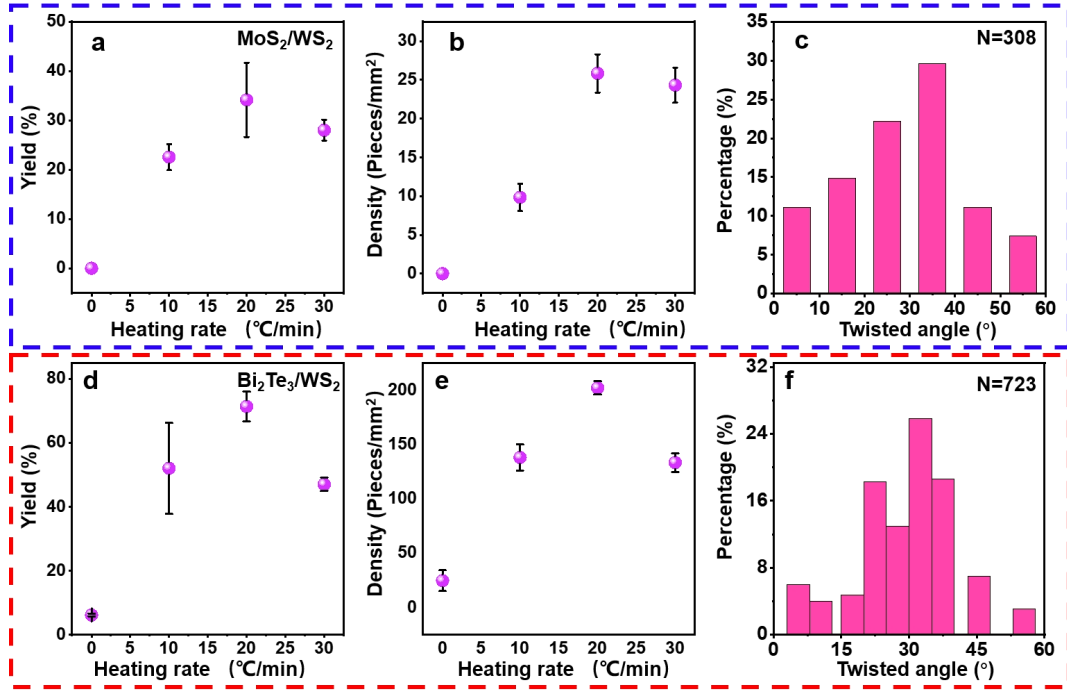
Supplementary Figure 15. General synthesis of diverse twisted TMD vdW heterostructures. a-r, The Raman spectra of MoSe₂/WS₂, NbS₂/WS₂, MoS₂/WS₂, Ge/WS₂, CrSe₂/WSe₂, VSe₂/WSe₂, Bi₂Te₃/WS₂, Bi₂Se₃/WS₂, SnS₂/WS₂, In₂Se₃/WS₂, NbSe₂/WS₂, Sb₂O₃/WS₂, NiTe₂/WS₂, MoSe₂/WSe₂, WSe₂/WS₂, NiSe₂/WS₂, Cr₃Te₄/WS₂, Te/WS₂. The Raman spectrum of WS₂ is approximately at 355 cm⁻¹ and 416 cm⁻¹. The Raman spectrum of WSe₂ is approximately at 250 cm⁻¹. The Raman spectrum of MoS₂ is approximately at 384 cm⁻¹ and 405 cm⁻¹. The Raman spectrum of MoSe₂ is approximately at 248 cm⁻¹ and 282 cm⁻¹. The Raman spectrum of SnS₂ is approximately at 314 cm⁻¹. The Raman spectrum of Bi₂Te₃ is approximately at 104 cm⁻¹ and 139 cm⁻¹. The Raman spectrum of Bi₂Se₃ is approximately at 125 cm⁻¹ and 175 cm⁻¹. The Raman spectrum of NbSe₂ is approximately at 168 cm⁻¹ and 224 cm⁻¹, and 260 cm⁻¹. The Raman spectrum of NbS₂ is approximately at 297 cm⁻¹ and 379 cm⁻¹, and 439 cm⁻¹. The Raman spectrum of Ge is approximately at 302 cm⁻¹. The Raman spectrum of CrSe₂ is approximately at 221 cm⁻¹. Raman spectrum of VSe₂ is approximately at 207 cm⁻¹. Raman spectrum of In₂Se₃ is approximately at 175 cm⁻¹. Raman spectrum of Sb₂O₃ is approximately at 190 cm⁻¹, 255 cm⁻¹ and 451 cm⁻¹. Raman spectrum of NiTe₂ is approximately at 83 cm⁻¹ and 134 cm⁻¹. Raman spectrum of NiSe₂ is approximately at 145 cm⁻¹. Raman spectrum of Cr₃Te₄ is approximately at 127 cm⁻¹ and 143 cm⁻¹.



Supplementary Figure 16. EDS characterization of Cr_3Te_4 in the $\text{Cr}_3\text{Te}_4/\text{WS}_2$ twisted heterostructure. a-b, The EDS spectrum of Cr_3Te_4 nanosheet. The scale bars are 1 μm . **c**, Spectrum from EDS analysis.



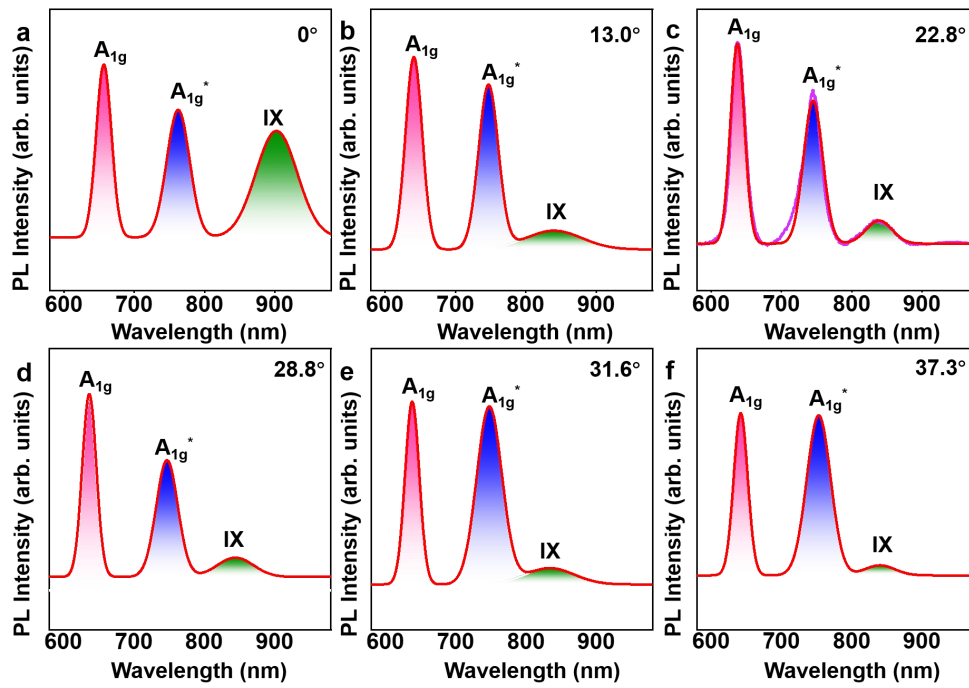
Supplementary Figure 17. SHG characterization of twisted MoS_2/WS_2 and $\text{Bi}_2\text{Te}_3/\text{WS}_2$ heterostructures. a, OM images of 12° - MoS_2/WS_2 heterostructure. **b**, OM images of 4° - $\text{Bi}_2\text{Te}_3/\text{WS}_2$ heterostructure. **c-d**, Polar plots of normalized SHG intensities for (a) and (b), respectively. The red and blue petals correspond to the position of the red circle and blue circle in (a) and (b), respectively.



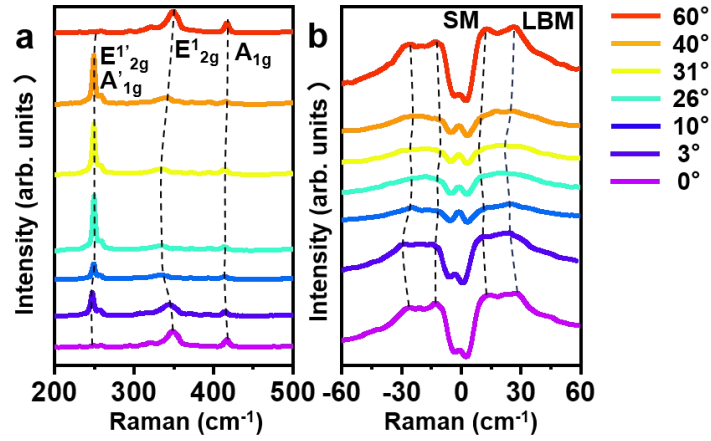
Supplementary Figure 18. Statistical analysis of yield, density and twist angle distribution in MoS₂/WS₂ and Bi₂Te₃/WS₂ heterostructures under different heating rates.

a, Statistical chart of the yield of TB-MoS₂/WS₂ heterostructures corresponding to different heating rates. The yield is dedicated to the proportion of TB-MoS₂/WS₂ heterostructures in MoS₂/WS₂ heterostructures. The error bars (representing the standard deviation (s.d.)) are derived from three independent experiments at the corresponding heating rates. The error bars for the four sets in (a) are 0.00% ± 0.00%, 22.54% ± 2.65%, 34.16% ± 7.52%, and 28.01% ± 2.13%, respectively. **b**, Statistical chart of the density of TB- MoS₂/WS₂ heterostructures under different heating rates. The density is defined as the number of TB-MoS₂/WS₂ heterostructures per unit area. Error bars represent mean ± standard deviation. The error bars for the four sets in (a) are 0.00± 0.00 pieces/mm², 9.83± 1.76 pieces/mm², 25.83± 2.47 pieces/mm², and 24.31± 2.25 pieces/mm², respectively. **c**, Statistical chart of the angular distribution of TB-MoS₂/WS₂ heterostructures. The statistics were derived from all TB-MoS₂/WS₂ heterostructures prepared by the temperature-gradient-controlled CVD method. **d**, Statistical chart of the yield of twisted Bi₂Te₃/WS₂ heterostructures corresponding to different heating rates. The yield is dedicated to the proportion of twisted Bi₂Te₃/WS₂ heterostructures in Bi₂Te₃/WS₂ heterostructures. Error bars represent mean ± standard deviation. The error bars

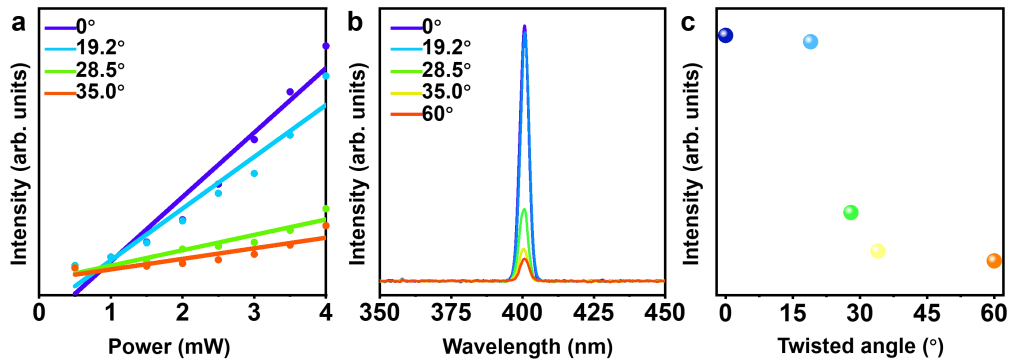
for the four sets in (a) are $6.16\% \pm 0.43\%$, $52.03\% \pm 14.22\%$, $71.37\% \pm 4.63\%$, and $47.01\% \pm 2.06\%$, respectively. **e**, Statistical chart of the density of twisted $\text{Bi}_2\text{Te}_3/\text{WS}_2$ heterostructures under different heating rates. The density is defined as the number of twisted $\text{Bi}_2\text{Te}_3/\text{WS}_2$ heterostructures per unit area. Error bars represent mean $\pm$ standard deviation. The error bars for the four sets in (a) are 24.67 ± 9.61 pieces/ mm^2 , $138.00 \pm 12.12\%$, $202.00\% \pm 6.08\%$, and $133.33\% \pm 8.62\%$, respectively. **f**, Statistical chart of the angular distribution of twisted $\text{Bi}_2\text{Te}_3/\text{WS}_2$ heterostructures. The statistics were derived from all twisted $\text{Bi}_2\text{Te}_3/\text{WS}_2$ heterostructures prepared by the temperature-gradient-controlled CVD method.



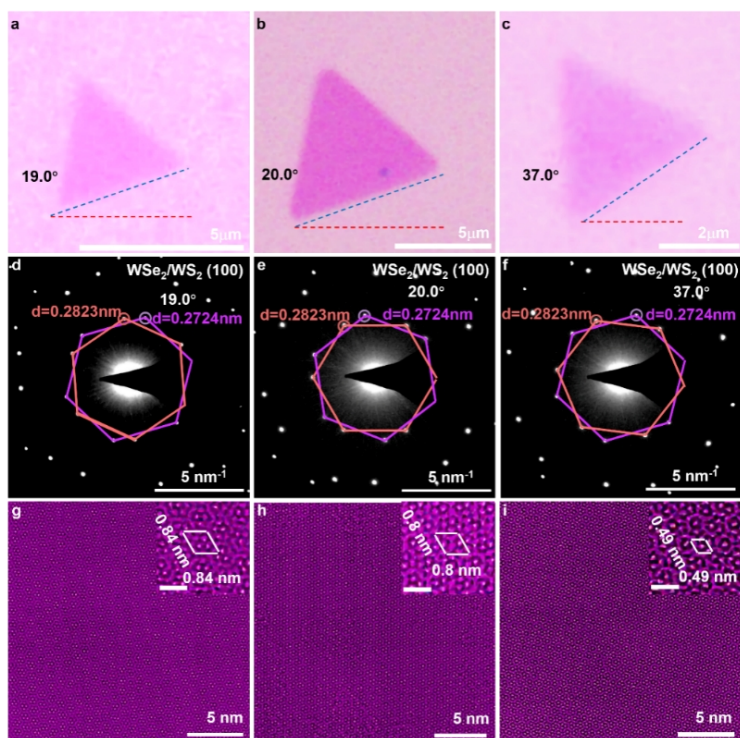
Supplementary Figure 19. PL spectra of TB-WSe₂/WS₂ heterostructures at 80 K. a, PL spectra of 0°-WSe₂/WS₂ heterostructure. **b**, PL spectra of 13.0°-WSe₂/WS₂ heterostructure. **c**, PL spectra of 22.8 ° -WSe₂/WS₂ heterostructure. **d**, PL spectra of 28.8 ° -WSe₂/WS₂ heterostructure. **e**, PL spectra of 31.6 ° -WSe₂/WS₂ heterostructure. **f**, PL spectra of 37.3 ° -WSe₂/WS₂ heterostructure. A_{1g} denotes the A exciton peak in WS₂, A_{1g}* denotes the A exciton peak in WSe₂, IX denotes the interlayer exciton peak of the WS₂/WSe₂ heterostructure respectively. PL is an abbreviation for photoluminescence spectra.



Supplementary Figure 20. The Raman spectra of WSe₂/WS₂ Heterostructures with different twist angles. **a, b** the low-frequency raman spectra and the high-frequency raman spectra of WSe₂/WS₂ Heterostructures with different twist angles. Peaks at 249 cm⁻¹, 257 cm⁻¹, 348 cm⁻¹ and 416 cm⁻¹ are consistent with the E'₁₂g, A'₁g mode of WSe₂, the E'₁₂g, A₁g mode of WS₂, respectively.

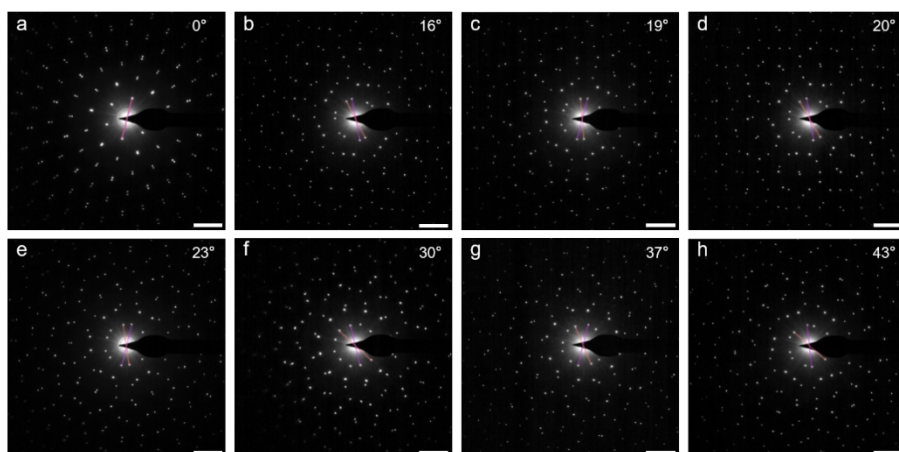


Supplementary Figure 21. SHG optical characterization of WSe₂/WS₂ heterostructures with different twist angles. **a**, Pump power dependence of SHG intensity in TB-WSe₂/WS₂ with the various twist angles. **b**, SHG spectrum of 0°, 19.2°, 28.5°, 35°, and 60°-TB-WSe₂/WS₂. **c**, The variation in SHG intensity for 0°, 19.2°, 28.5°, 35°, and 60°-TB-WSe₂/WS₂ is plotted as a function of the twist angle. Laser power is 1 mW.



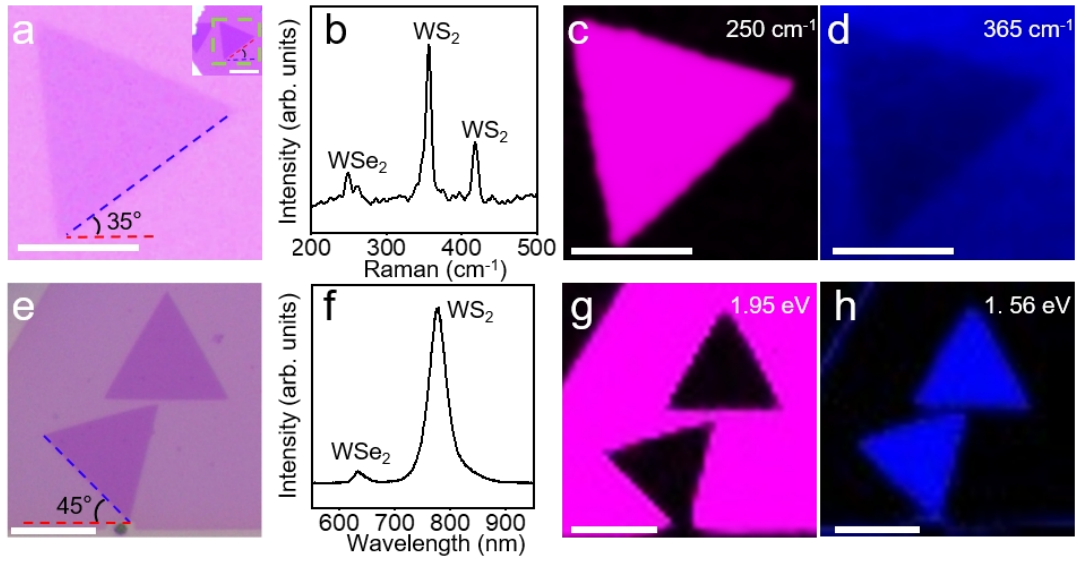
Supplementary Figure 22. STEM characterization of twisted WSe₂/WS₂ heterostructures.

a-c, OM images of 19°, 20°, and 37°-WSe₂/WS₂ heterostructures. **d-f**, The SAED patterns for structural characterization of 19°, 20°, and 37°-WSe₂/WS₂ heterostructures. The pink and purple hexagons mark the diffraction patterns from WSe₂ and WS₂ lattices, respectively. **g-i**, STEM images for structural characterization of 19°, 20°, and 37°-WSe₂/WS₂ heterostructures. Insets show magnified views of the moiré superlattices. The white dashed box indicates a Moiré period.

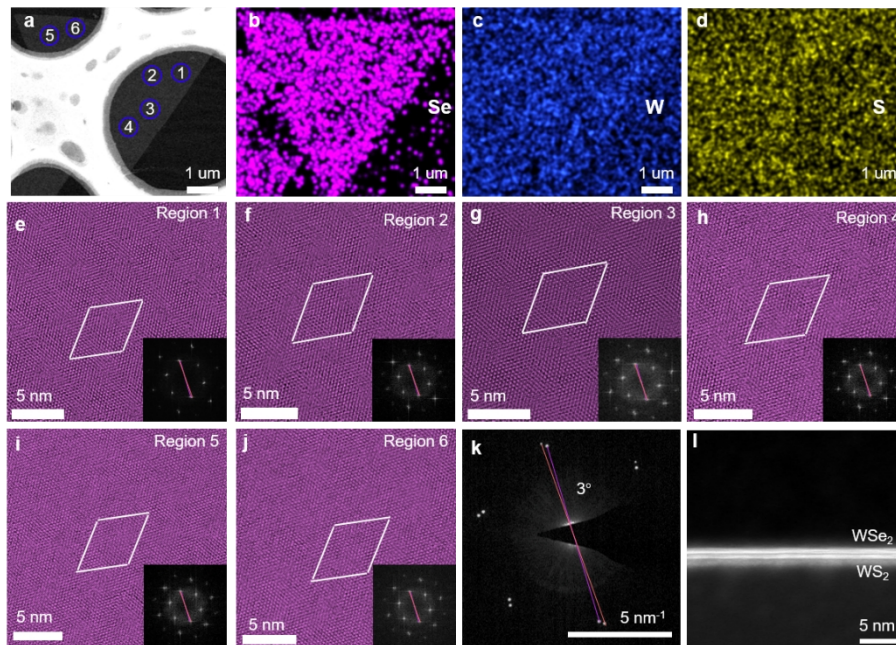


Supplementary Figure 23. Twist Angle Characterization of Twisted WSe₂/WS₂ Heterostructures. **a-h**, The SAED patterns for structural characterization of 0°, 16°, 19°, 20°, 23°, 30°, 37°, and 43°.

23°, 30°, 37° and 43.0°-TB-WSe₂/WS₂, respectively. The pink and purple hexagons mark the diffraction patterns from WSe₂ and WS₂ lattices, respectively. Scale bar: 5 nm⁻¹

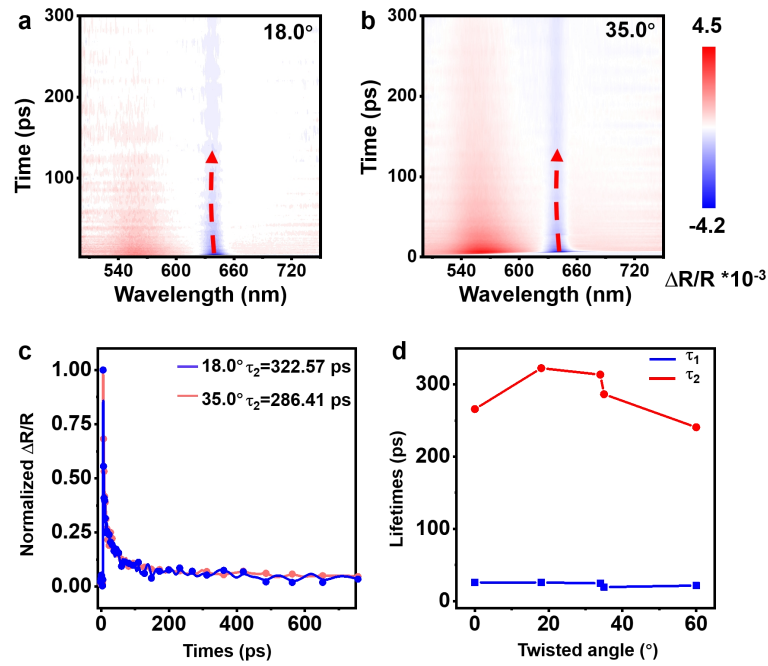


Supplementary Figure 24. The OM image, Raman, and PL measurements of twisted WSe₂/WS₂ Heterostructures. a, The OM image of 35°-twisted-WSe₂/WS₂ heterostructure. b, c) The raman mapping image of 35°-twisted-WSe₂/WS₂ heterostructure in (a) at the characteristic peak. d, The OM image of 45°-twisted-WSe₂/WS₂ heterostructure. e, f) The PL mapping image of 45°-twisted-WSe₂/WS₂ heterostructure in (d) at the characteristic peak. Scale bar: 10 μm.



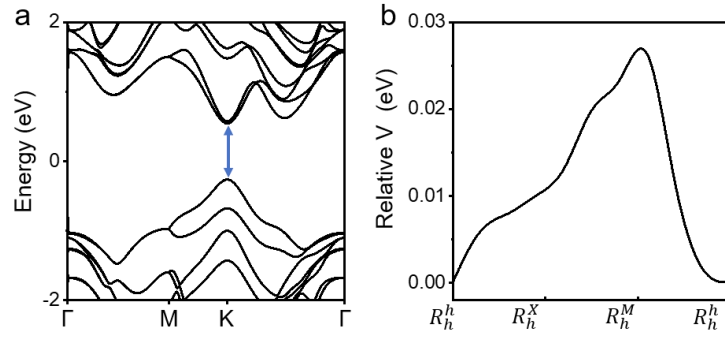
Supplementary Figure 25. The structural characterization of twisted WSe₂/WS₂

heterostructure. a, The TEM image of 3°-WSe₂/WS₂ heterostructure. **b-d**) The corresponding EDS mapping images of 3°-WSe₂/WS₂ heterostructure. **b**) Se, **c**) W, and **d**) S. **e-j**) The TEM images of 3°-WSe₂/WS₂ heterostructure. The white dashed rhombus exhibits the periodically repeating moiré cells of the 3°-WSe₂/WS₂ heterostructure. The moiré period in the figure is 5.16 nm ± 0.07 nm. **k**, The SAED image of 3°-WSe₂/WS₂ heterostructure. The pink and purple hexagons mark the diffraction patterns from WSe₂ and WS₂ lattices, respectively. **l**, The cross-sectional STEM image of twisted WSe₂/WS₂ heterostructure.



Supplementary Figure 26. TR characterization of twisted WSe₂/WS₂ heterostructures.

a-b, Spectro-temporal transient reflectance maps for 18° and 35°-TB-WSe₂/WS₂ heterostructures. **c**, Ground-state bleaching kinetics of 18° and 35°-TB-WSe₂/WS₂ heterostructures. The ΔR/R represents reflectance change. **d**, the statistical plot of τ₁ and τ₂ from the biexponential fitting of the ground-state bleaching curves for the 0°, 18°, 34°, 35° and 60°-TB-WSe₂/WS₂ heterostructures. The data points represent the lifetime values of τ₁ and τ₂ corresponding to 0°, 18°, 34°, 35° and 60°-TB-WSe₂/WS₂ heterostructures, respectively. The solid lines indicate the approximate trends of τ₁ and τ₂ as the twist angle increases from 0° to 60°-TB-WSe₂/WS₂ heterostructures. Here, τ₁ and τ₂ are time constants, with τ₁ representing the fast decay component and τ₂ the slow decay component.



Supplementary Figure 27. Moiré potential in WS₂/WSe₂ heterostructures obtained via theoretical calculation. **a**, Schematic band structure of a representative WSe₂/WS₂ heterostructure. **b**, Relative moiré exciton potential in different stacking configurations. Here the moiré exciton potential of the R_h^h configuration is set to zero for clarity.

Supplementary Table 1. The number of statistically twisted WSe₂/WS₂ samples, the total number of statistically vertical WSe₂/WS₂ heterostructures, the substrate area and the yield and density for each set of experiments. Note: Samples with angles of 0° and 60° are defined as non-twisted, while all angles other than 0° and 60° are considered twisted.

Heating rate (°C/min)	Proportion of twist angles (pieces)	Substrate area (mm ²)	Yield (%)	Density (pieces/ mm ²)
0	0/303	3	0.00, 0.00, 0.00	0.00, 0.00, 0.00
5	73/335	40	17.12, 28.13, 25.42, 20.74	9.50, 9.00, 7.50, 10.50
20	1341/3839	50	46.87, 28.85, 25.49, 22.9	29.04, 24.07, 25.88, 23.67
35	85/241	6	20.83, 21.74, 34.78, 32.76	24.00, 24.00, 25.00, 21.00
40	108/445	5.5	27.22, 27.40, 26.73, 16.25	15.00, 23.75, 21.00, 16.00

Supplementary Table 2. The number of statistically twisted MoS₂/WS₂ samples, the total number of statistically vertical MoS₂/WS₂ heterostructures, the substrate area and the yield and density for each set of experiments. Note: Samples with angles of 0° and 60° are defined as non-twisted, while all angles other than 0° and 60° are considered twisted.

Heating rate (°C/min)	Proportion of twist angles (pieces)	Substr ate area	Yield (%)	Density (pieces/ mm ²)
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(mm ²)				
0	0/303	3	0.00, 0.00, 0.00	0.00, 0.00, 0.00
10	132/385	5	25.29, 22.33, 20.00	11.50, 10.00, 8.00
20	61/270	3	42.67, 31.40, 28.40	27.00, 27.00, 23.00
30	65/233	3	30.14, 28.00, 25.88	26.50, 24.44, 22.00

Supplementary Table 3. The number of statistically twisted Bi₂Te₃/WS₂ samples, the total number of statistically vertical Bi₂Te₃/WS₂ heterostructures, the substrate area and the yield and density for each set of experiments. Note: Samples with angles of 0° and 60° are defined as non-twisted, while all angles other than 0° and 60° are considered twisted.

Heating rate (°C/min)	Proportion of twist angles (pieces)	Substrate area (mm ²)	Yield (%)	Density (pieces/mm ²)
0	42/705	1.8	5.75, 6.60, 6.12	26, 15, 23
10	372/719	2.6	36.70, 54.59, 64.79	122, 168, 144
20	513/711	1.8	67.04, 76.25, 70.28	200, 285, 206
30	115/245	0.9	47.41, 48.84, 44.78	128, 127, 150

Supplementary Table 4. Preparation methods for other twisted 2D structures.

Type	Materials	Preparation Method	Angle of rotation range	Reference
Homostructure	twisted bilayer graphene	Mechanical Stacking	1.05°, 1.16°	1
Homostructure	Twisted Bilayer MoS ₂	gas-flow perturbation CVD	8° to 56°	2
Heterostructure	Twisted WS ₂ /WSe ₂	Mechanical Stacking	0°, 30°, 60°	3
Homostructure	Twisted MoS ₂ bilayers	Mechanical Stacking	22° to 38°	4
Homostructure	twisted bilayer TaS ₂	Mechanical Stacking	5.1° to 27.8°	4
Heterostructure	Twisted MoSe ₂ /WSe ₂	Mechanical Stacking	0° to 60°	5
Heterostructure	Twisted MoSe ₂ /WSe ₂	Mechanical Stacking	1° to 44.3°	6
Homostructure	Twisted WSe ₂	CVD	15°	7
Homostructure	twisted bilayer MoS ₂	CVD	0°, 15°, 30°	8
Homostructure	twisted bilayer WS ₂	CVD	19°	8
Homostructure	twisted bilayer graphene	CVD	4.1°	9
Homostructure	Twisted graphene	Mechanical Stacking	4.8° to 30°	10
Homostructure	Twisted graphene	Mechanical Stacking	7.5° to 55.5°	11
Homostructure	twisted black phosphorus	Mechanical Stacking	0° to 90°	12
Heterostructure	Twisted WS ₂ /Sb ₂ S ₃	CVD	0° to 180°	13
Homostructure	twisted bilayer graphene	Gas-flow perturbation CVD	3° to 30°	14
Heterostructure	MoS ₂ -graphene	Mechanical Stacking	2° to 14°	15
Homostructure	supertwisted spirals WS ₂	CVD	0° to 15°	16
Homostructure	twisted WS ₂	Sn atom-assisted CVD	0° to 60°	17
Heterostructure	Twisted WS ₂ /WSe ₂	Mechanical Stacking	58°	18
Homostructure	twisted bilayer graphene	prerotated-assisted CVD	1° to 30°	19
Homostructure	twisted WS ₂	tilted substrate-assisted CVD	0° to 60°	20
Homostructure	twisted WSe ₂	Sn atom-assisted CVD	0° to 50°	21

Supplementary Table 5. Amplitude (A_i) and lifetime (τ_i) values obtained from biexponential fitting of ultrafast transient reflection (TR) curves for WSe₂/WS₂ heterostructures at different twist angles.

Twisted angles	A_1	τ_1 (ps)	A_2	τ_2 (ps)
0°	0.0062	25.842	0.00077	265.875
18°	0.000665	25.84	0.000103	322.57
34°	0.00709	24.54	0.00108	313.62
35°	0.00261	19.428	0.000348	286.408
60°	0.01073	21.56	0.00149	240.68

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