Supplementary Information to

"Twist Angle-Dependent Interlayer Exciton in MoS_2 Bilayers Revealed by Room-Temperature Reflectance"

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Contents

SI-1 Details on the sample preparation

Firstly, we put a Polydimethylsiloxane (PDMS) strip on a glass slide, then the MoS_2 flakes were exfoliated onto the surface of the PDMS and examined by optical microscopy to find a sizeable smooth monolayer MoS_2. We repeated the above steps to find another monolayer MoS_2 of similar size. Next, we cleaned a quartz/sapphire substrate, and the side of the glass slide with PDMS was parallel to the quartz/sapphire substrate. The relative position of the monolayer on PDMS and the quartz/sapphire substrate was observed through the microscope. Then, the PDMS was slowly moved close to the quartz/sapphire substrate and finally making the monolayer MoS_2 closely fit the quartz/sapphire substrate. The stage temperature was set at 80 °C for 5 min to release the monolayer. The process was then repeated to transfer another monolayer MoS_2 to the monolayer MoS_2 on a quartz/sapphire substrate with specific twist angles.

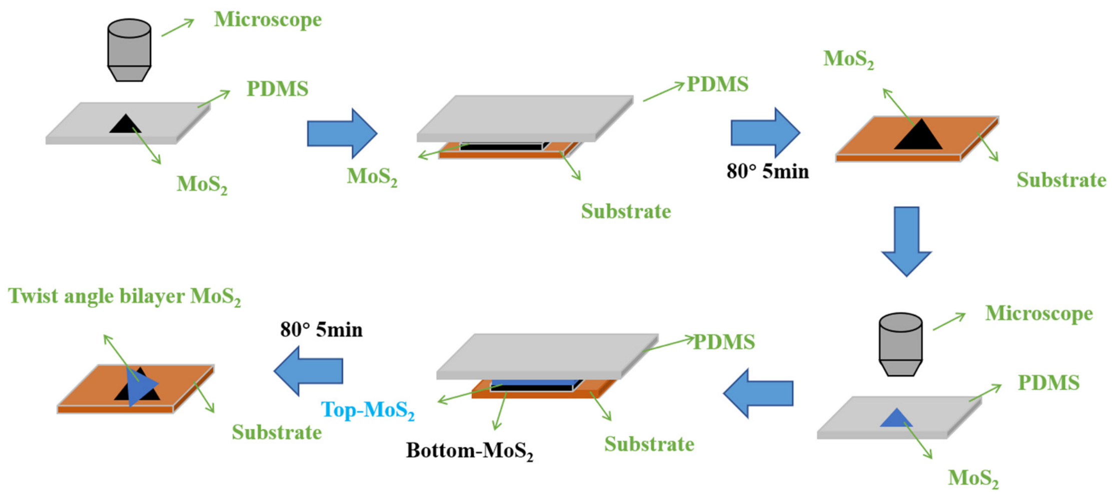


Figure S1-1 Schematics of the transfer process.

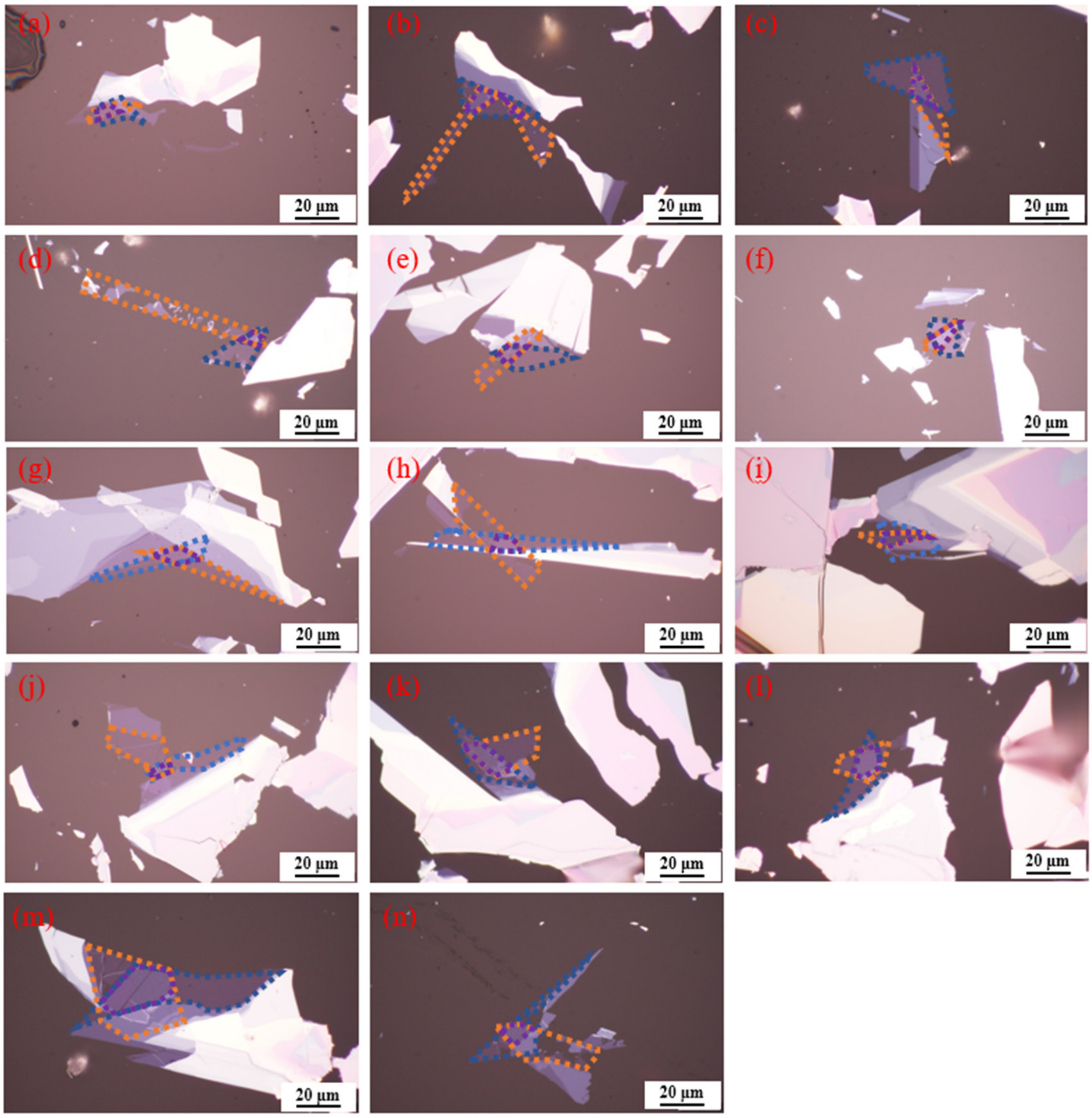
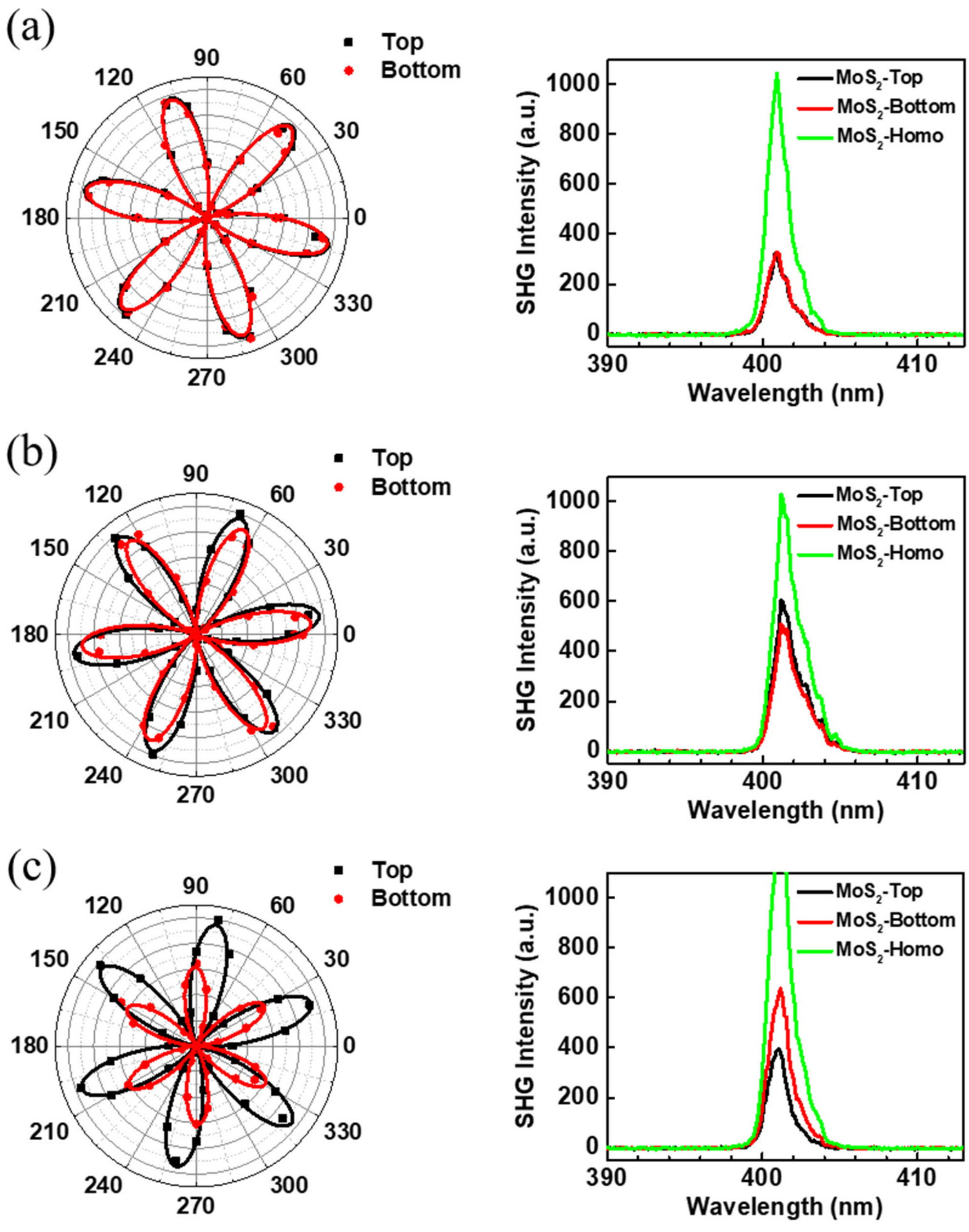


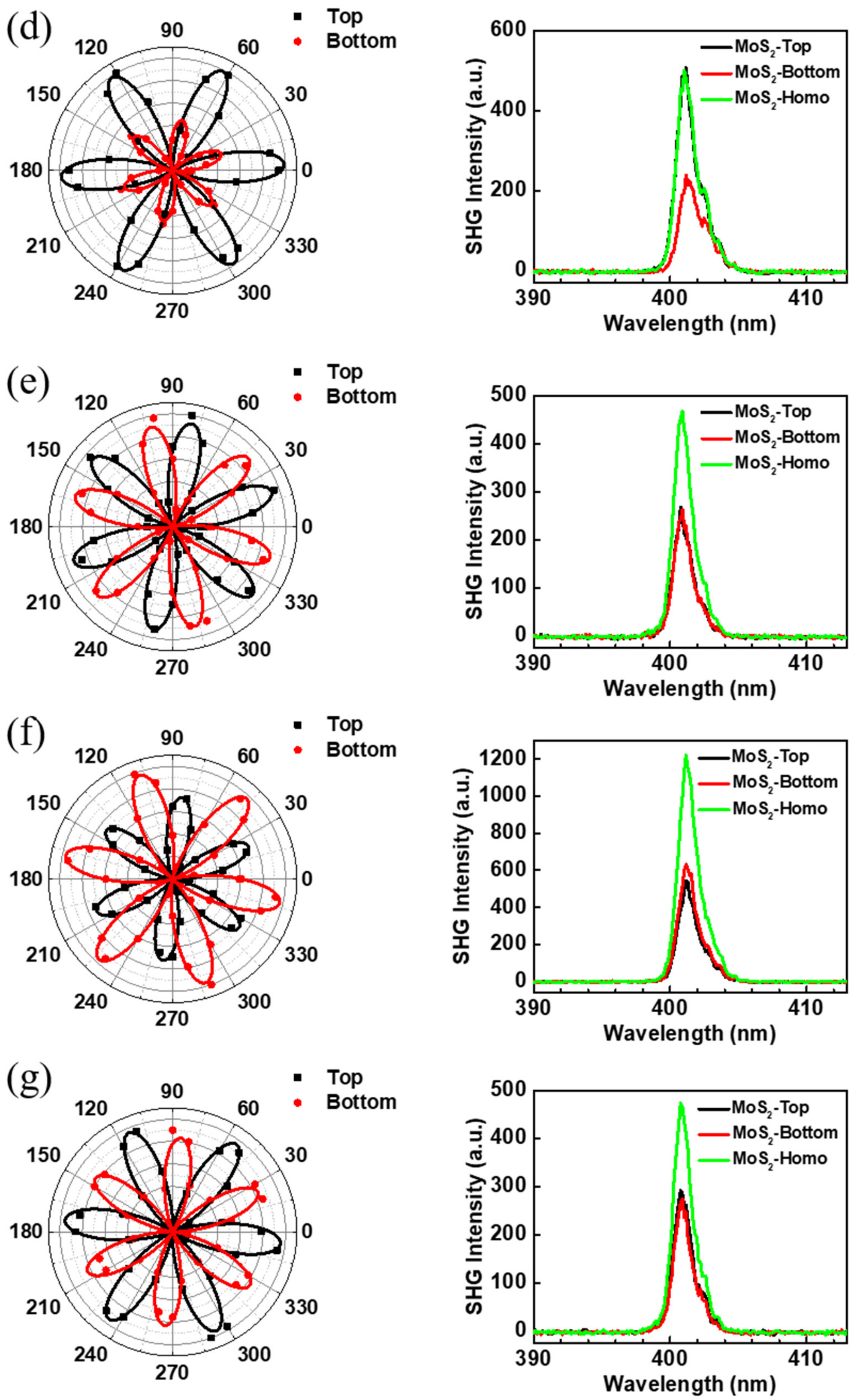
Figure S1-2. **(a-n)** Optical microscopy images of twisted bilayer MoS_2 with the twist angle of 0°, 3°, 10°, 16°, 23°, 24°, 31°, 41°, 42°, 50°, 55°, 56°, 58° and 60°, respectively. The yellow dotted line represents the upper monolayer MoS_2, the blue dotted line represents the bottom monolayer MoS_2, and the purple dotted line represents the twist angle bilayer region.

SI-2 SHG measurements of monolayers and twisted bilayers

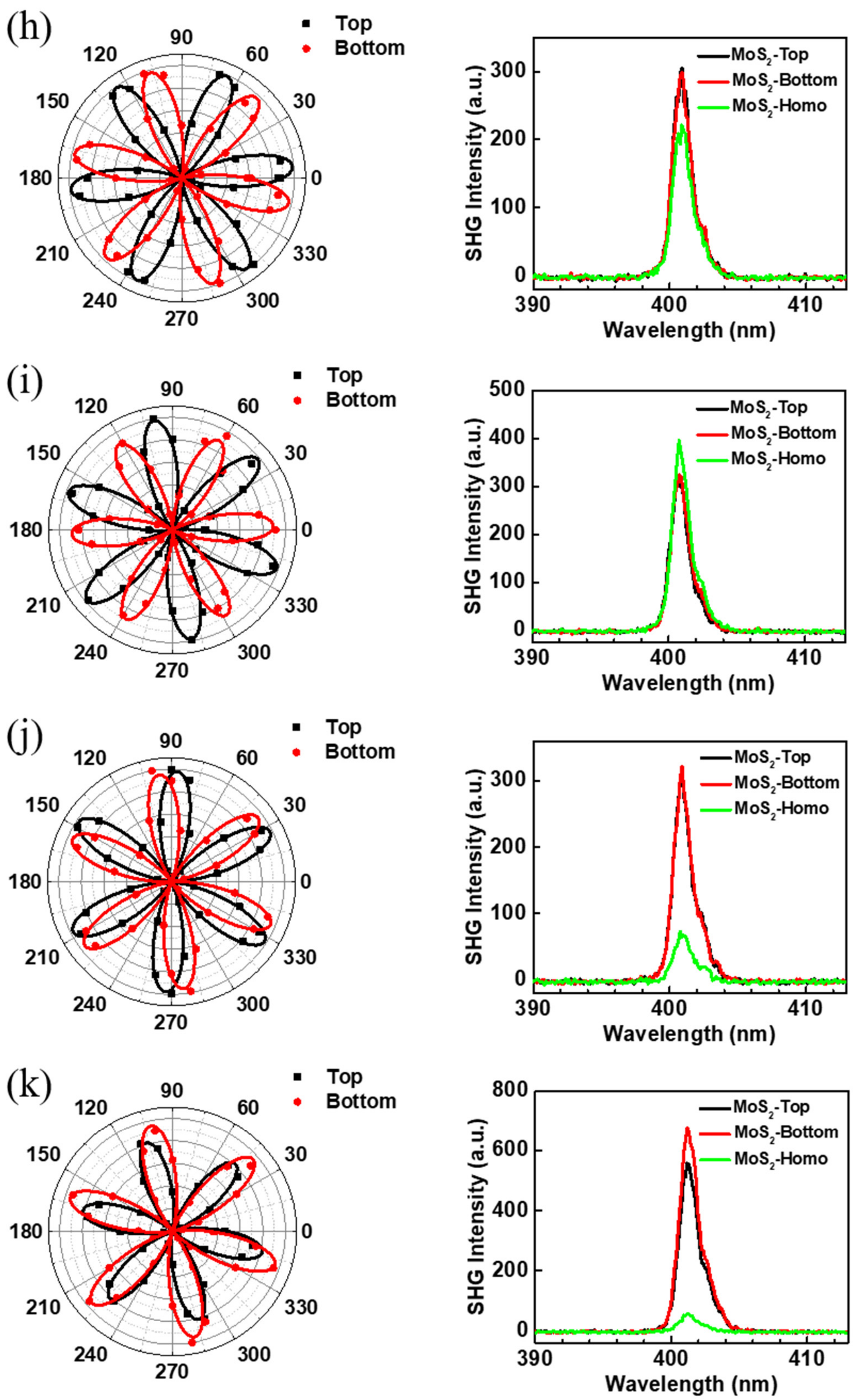
The crystal orientation of each layer in MoS_2 bilayer samples can be obtained by polarization-resolved second harmonic generation (PSHG). However, the six-fold SHG pattern cannot distinguished the R stacking (~0° twist angle) and H (~60° twist angle) stacking directly since the three-fold rotation symmetry nature of TMDCs monolayer. By comparing the SHG intensity of the bilayer and monolayer region directly, the two cases can be easily separated [S1].

The PSHG measurements were performed by a 150 fs Ti-sapphire oscillator (Coherent Chameleon II) or a 6 ps Supercontinuum laser. The repetition rate was both at 80 MHz and centered at a wavelength of 800 nm. The excitation laser was focused by a 50X objective (Nikon MUE31500) on the sample, and the SHG signal was collected by the same objective and detected by a spectrometer (Princeton Instruments HRS-500).





(d)
(e)
(f)
(g)
Top
Bottom
90
60
30
0
330
300
270
240
210
180
150
120
SHG Intensity (a.u.)
Wavelength (nm)
MoS2-Top
MoS2-Bottom
MoS2-Homo
390
400
410



(h)
Top
Bottom
90
60
30
0
330
300
270
240
210
180
150
120
SHG Intensity (a.u.)
MoS2-Top
MoS2-Bottom
MoS2-Homo
0
100
200
300
390
400
410
Wavelength (nm)
(i)
Top
Bottom
SHG Intensity (a.u.)
MoS2-Top
MoS2-Bottom
MoS2-Homo
0
100
200
300
400
500
390
400
410
Wavelength (nm)
(j)
Top
Bottom
SHG Intensity (a.u.)
MoS2-Top
MoS2-Bottom
MoS2-Homo
0
100
200
300
390
400
410
Wavelength (nm)
(k)
Top
Bottom
SHG Intensity (a.u.)
MoS2-Top
MoS2-Bottom
MoS2-Homo
0
200
400
600
800
390
400
410
Wavelength (nm)

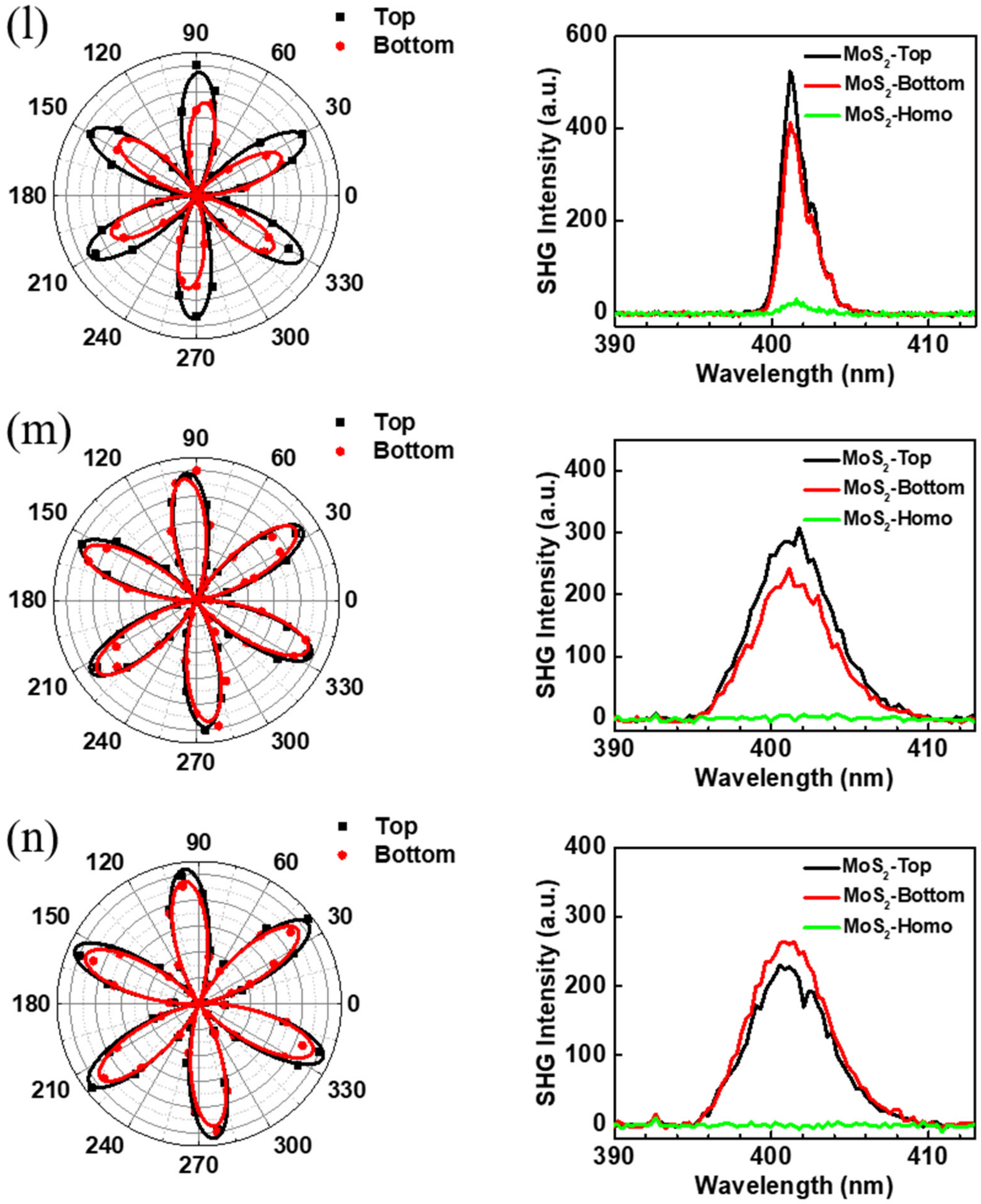


Figure S2. Left. Polar plot for PSHG from the top (black square) and bottom (red circle) layers of each sample, the solid line (black for top layers and red for bottom) is the corresponding fitting curve. **Right.** The SHG intensity for monolayer regions and homojunction regions. (a)-(n) represents the samples at a twist angle of 0°, 3°, 10°, 16°, 23°, 24°, 31°, 41°, 42°, 50°, 55°, 56°, 58° and 60°, respectively.

SI-3 Twist-angle dependence of the indirect emissions

To investigate the interlayer coupling in the twist angle-dependent bilayer, we measured the PL spectra of each sample in the range of 0° to 60°. The indirect exciton energy as a function of twist angle is shown in Figure S3. For the indirect exciton emission (peak I) of bilayer MoS_2, the emission energies decrease at twists of 0° or 60°, while the other energies are relatively high and unchanged with the twist angle. Liu et al. showed that the higher the indirect bandgap energy, the weaker the interlayer coupling strength [S2]. Here, our experimental results show that the interlayer coupling of the bilayers in this work demonstrates similar behavior.

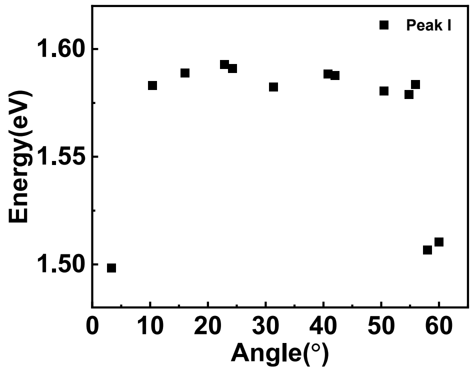


Figure S3. Twist angle-dependent energy positions of indirect excitons obtained by Gaussian fitting of PL spectra.

SI-4 Homogeneity of bilayers characterized by PL over the samples

In our work, the samples were stacked directly in an atmospheric environment. This type of sample is suitable for microscopic optical measurements, as the required uniform scale could be similar to or larger than the excitation/detection size. We annealed samples under an argon atmosphere of 200°C for 2 hours. The annealing will improve the interlayer coupling. We present the PL spectra of several points of representative twist angles that show the homogeneity of our samples is at a reasonable level.

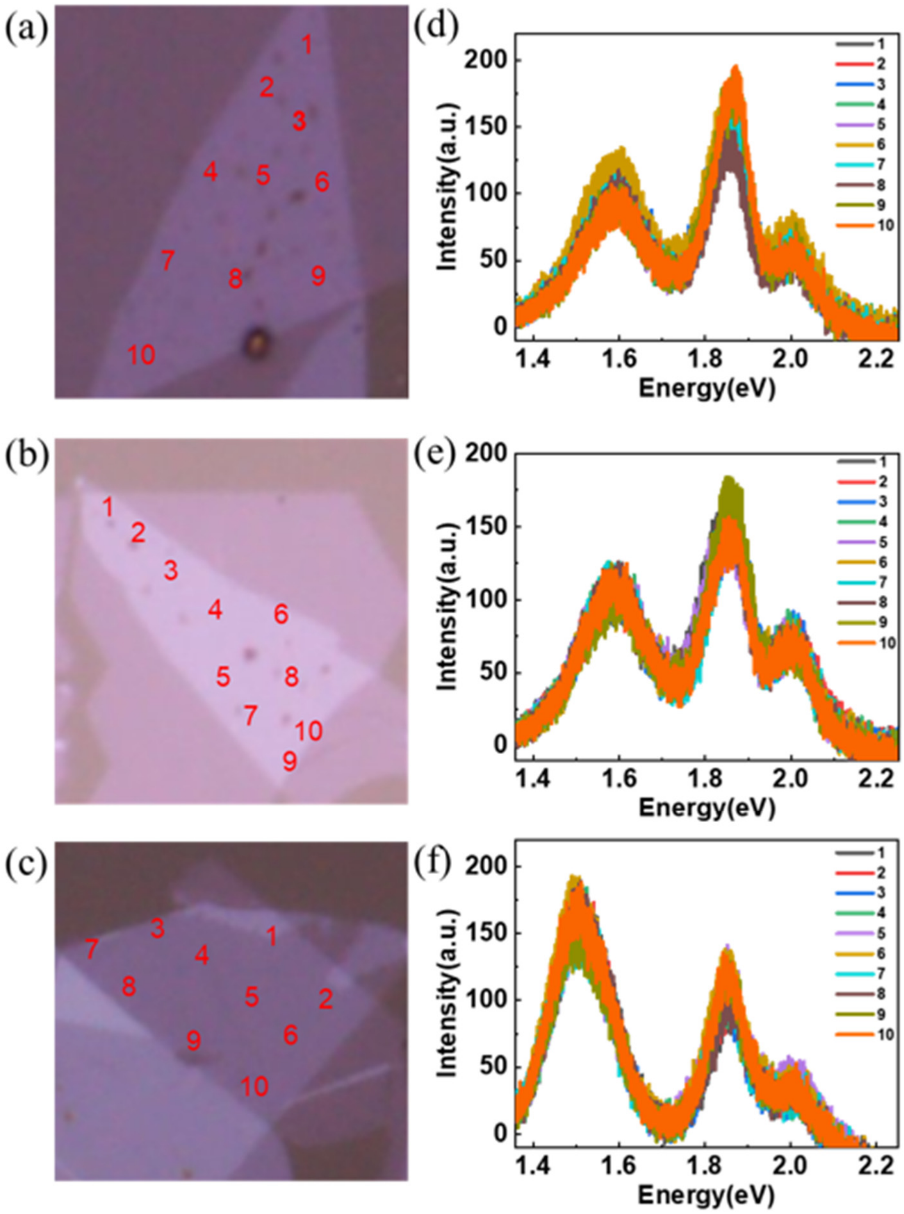


Figure S4. (a–c) Optical microscopy image of MoS_2 bilayer with the twist angles of 10°, 23° and 60°, respectively. The numbers represent the different positions where the PL are detected. (d–f) PL spectra of different positions correspond to the positions in (a-c), respectively.

Supplementary references:

[S1] Hsu, W.T.; Zhao, Z.A.; Li, L.J.; Chen, C.H.; Chiu, M.H.; Chang, P.S.; Chou, Y.C.; Chang, W. H. Second harmonic generation from artificially stacked transition metal dichalcogenide twisted bilayers. ACS nano, 2014, 8, 2951-2958.

[S2] Choi, J.; Florian, M.; Steinhoff, A.; Erben, D.; Tran, K.; Kim, D. S.; Sun, L.; Quan, J.; Claassen, R.; Majumder, S.; Hollingsworth, J. A.; Taniguchi, T.; Watanabe, K.; Ueno, K.; Singh, A.; Moody, G.; Jahnke, F.; Li, X. Twist angle-dependent interlayer exciton lifetimes in van der Waals heterostructures. Phys. Rev. Lett. 2021, 126, 047401.