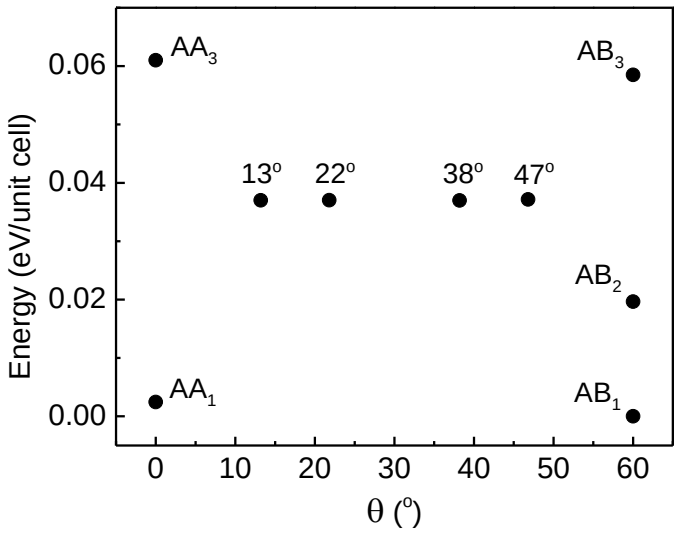
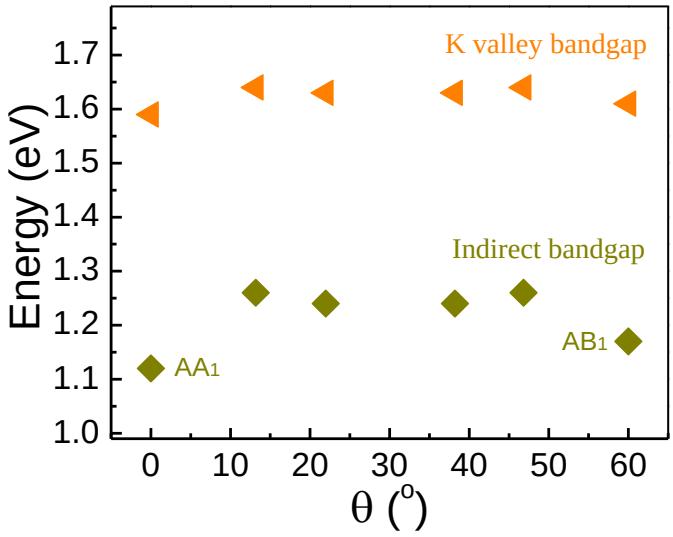
Supplementary Figures



Supplementary Figure 1. Calculated LDA relative energy of different configurations as calibrated to the lowest energy AB_1 configuration.



Supplementary Figure 2. Calculated values for the Kohn-Sham PBE-D direct bandgap at the K valley (orange) and indirect bandgap (dark yellow) for the energetically favorable structures at each twist angle.

Supplementary Table 1. Calculated interlayer distance in twisted graphene bilayers

Configuration	LDA interlayer distance (nm)	Van der Waals (PBE-D) interlayer distance (nm)
0° (higher energy)	0.35	0.34
0° (Bernal stacking)	0.33	0.32
13.2°	0.34	0.33
21.8°	0.34	0.33

Supplementary Note 1. Determination of stacking order for $\theta = 0°, 60°$ bilayers by SHG

The bilayers with twist angle of $\theta = 0°, 60°$ can have AA or AB stacking. In the AA stacking case, the in-plane Mo-S bond direction is the same for the two layers, and in the AB-stacking case the in-plane Mo-S bond direction is opposite for the two layers. In order to distinguish these two stacking orders, we employ nonlinear optical second-harmonic generation (SHG), which is very sensitive to asymmetry of the surface layers[1]. Monolayer MoS_2 belongs to the noncentrosymmetric D_{3h}^1 group and therefore has a strong SHG signal (black curve in Fig. 1j). In contrast, AB-stacked bilayer MoS_2 belongs to the centrosymmetric D_{3d}^1 group, and therefore, has no appreciable SHG signal (pink curve in Fig. 1j, $\theta = 60°$). Owing to the absence of an inversion center, the SHG signal is very strong for AA-stacked bilayer MoS_2 (dark orange curve in Fig. 1j, $\theta = 0°$), roughly two times stronger than for the monolayer. The fact that the $\theta = 0°(\theta = 60°)$ bilayer is AA (AB) stacking confirms that the two layers have the same edge types[2-4].

Supplementary References:

1 Li, Y. *et al.* Probing symmetry properties of few-layer MoS2 and h-BN by optical second-harmonic generation. *Nano Lett* **13**, 3329 (2013).

2 Lauritsen, J. V. *et al.* Size-dependent structure of MoS2 nanocrystals. *Nature Nanotech* **2**, 53-58 (2007).

3 van der Zande, A. M. *et al.* Grains and grain boundaries in highly crystalline monolayer molybdenum disulphide. *Nature Materials* **12**, 554-561 (2013).

4 Bollinger, M. V. *et al.* One-dimensional metallic edge states in MoS2. *Phys Rev Lett* **87**, 196803 (2001).