Electronic Supplementary Material (ESI) for CrystEngComm.

Formation Mechanism and Twist-angle Dependent Optical Properties of Bilayer MoS_2 Grown by Chemical Vapor Deposition

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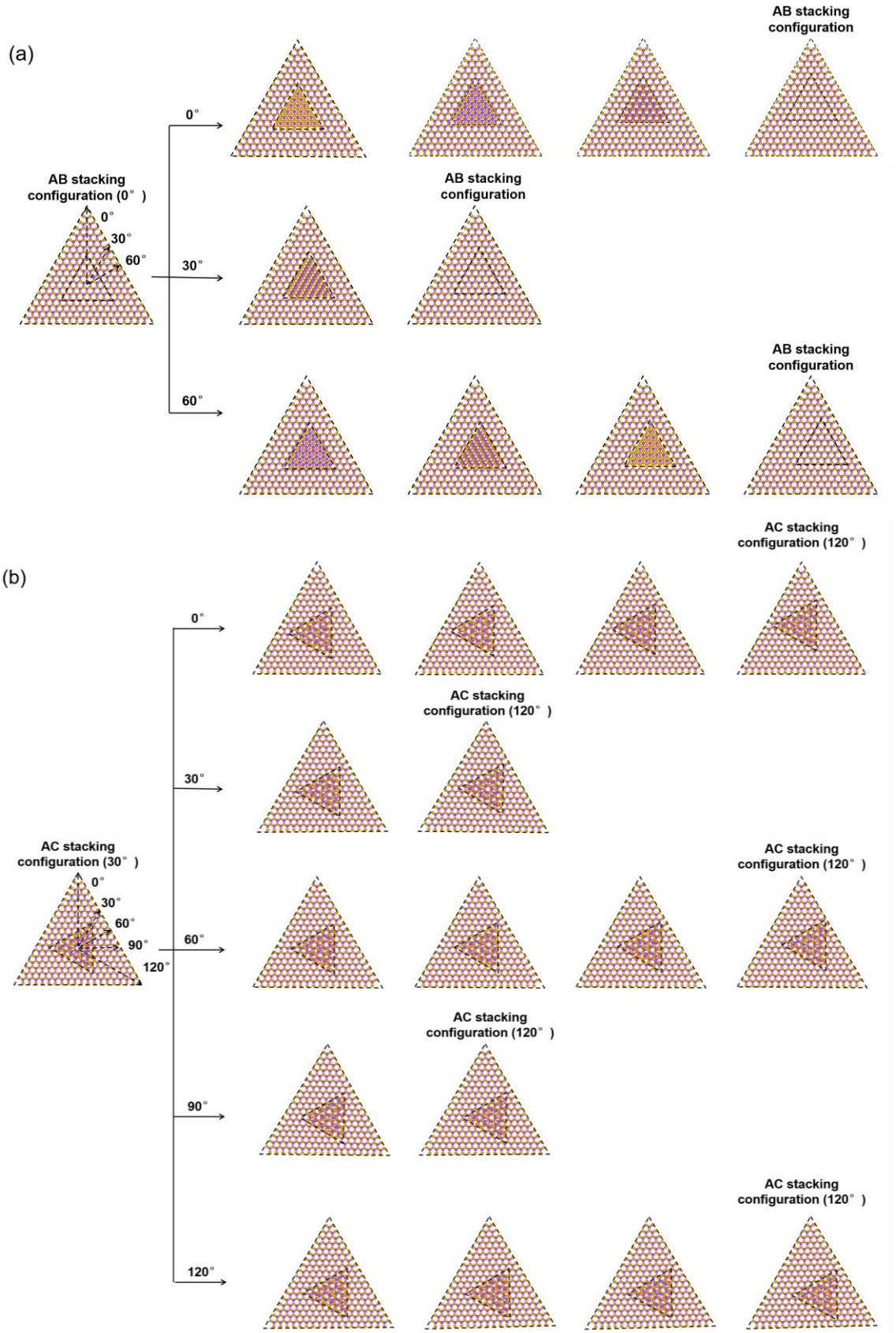

(a)
AB stacking configuration
0°
AB stacking configuration (0°)
0°
30°
60°
30°
AB stacking configuration
AB stacking configuration
60°
(b)
AC stacking configuration (120°)
0°
AC stacking configuration (120°)
30°
AC stacking configuration (30°)
0°
30°
60°
90°
120°
60°
AC stacking configuration (120°)
AC stacking configuration (120°)
90°
AC stacking configuration (120°)
120°

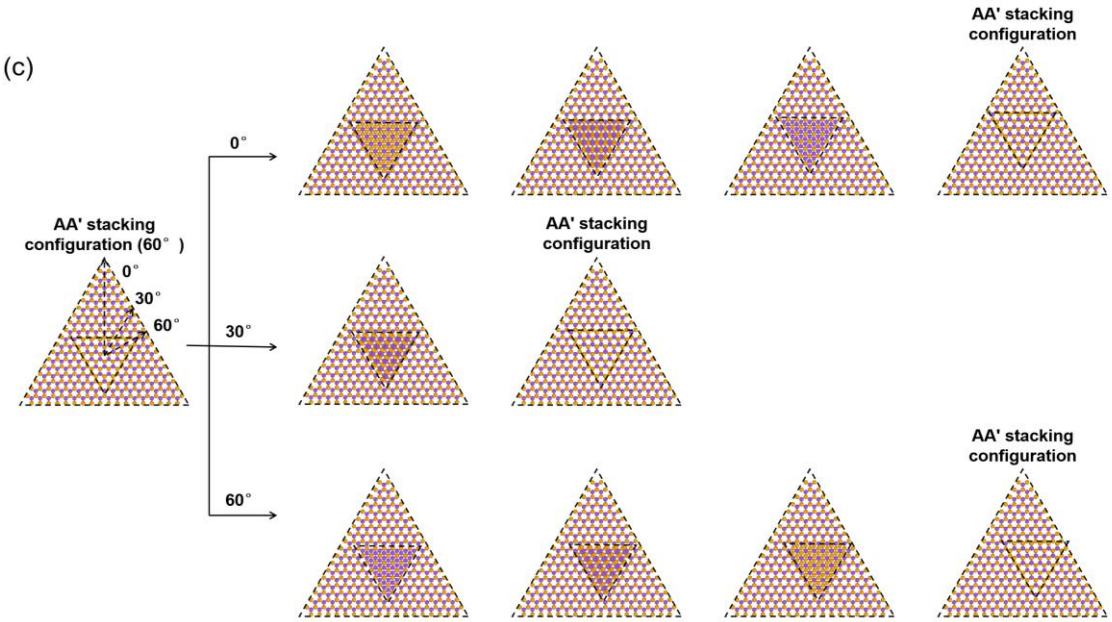
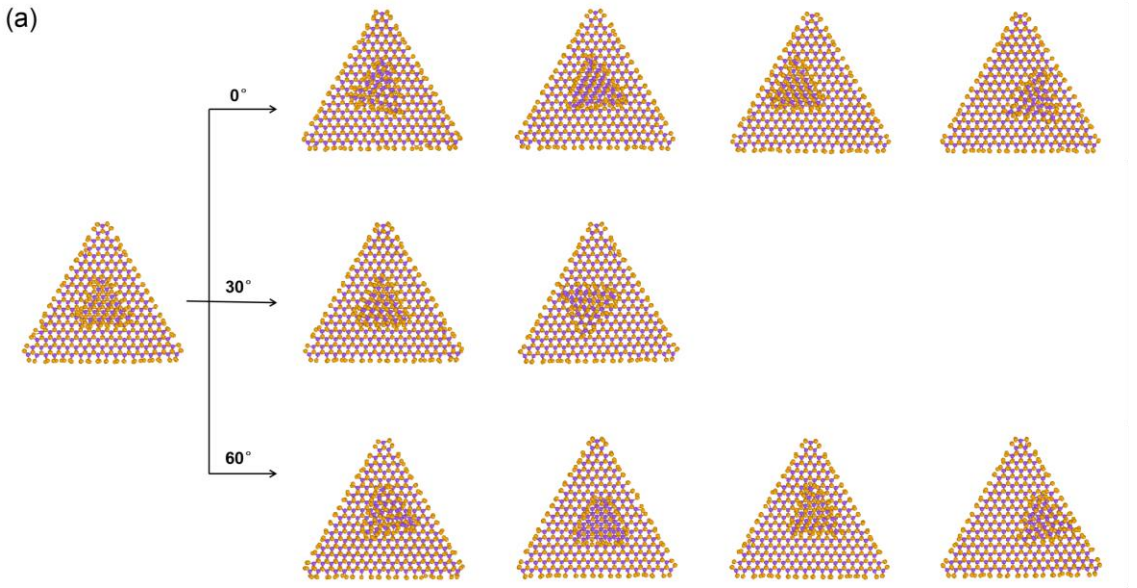
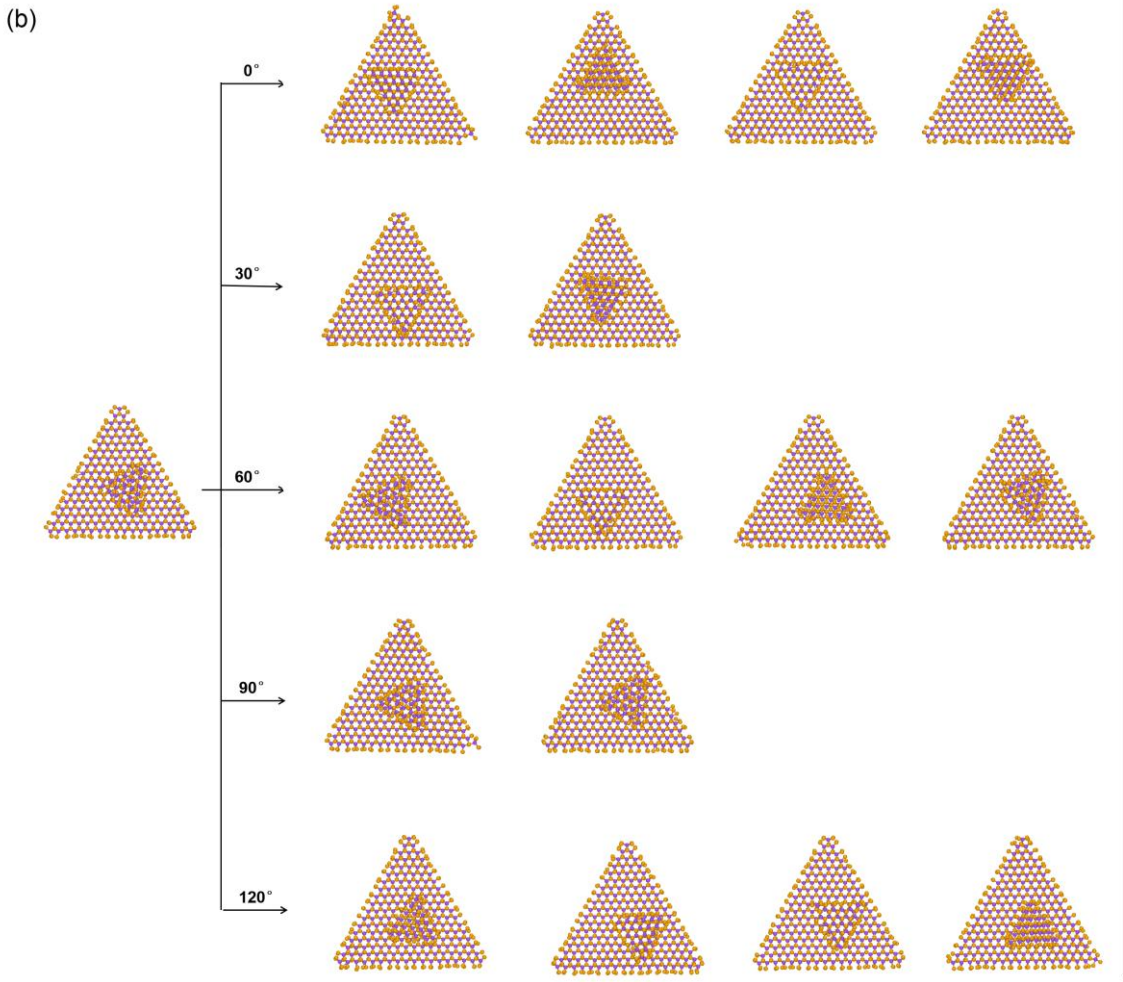


Fig. S1. The stacking configurations with twist angles of (a)0° (with the nucleation sites of top layers moving along 0°, 30° and 60° direction relative to that of the bottom layers), (b)30° (with the nucleation sites of top layers moving along 0°, 30°, 60°, 90° and 120° direction relative to that of the bottom layers) and (c)60° (along 0°, 30° and 60° direction relative to that of the bottom layers) simulated in our study, regardless of the temperature.


(a)
0°
30°
60°


(b)
0°
30°
60°
90°
120°

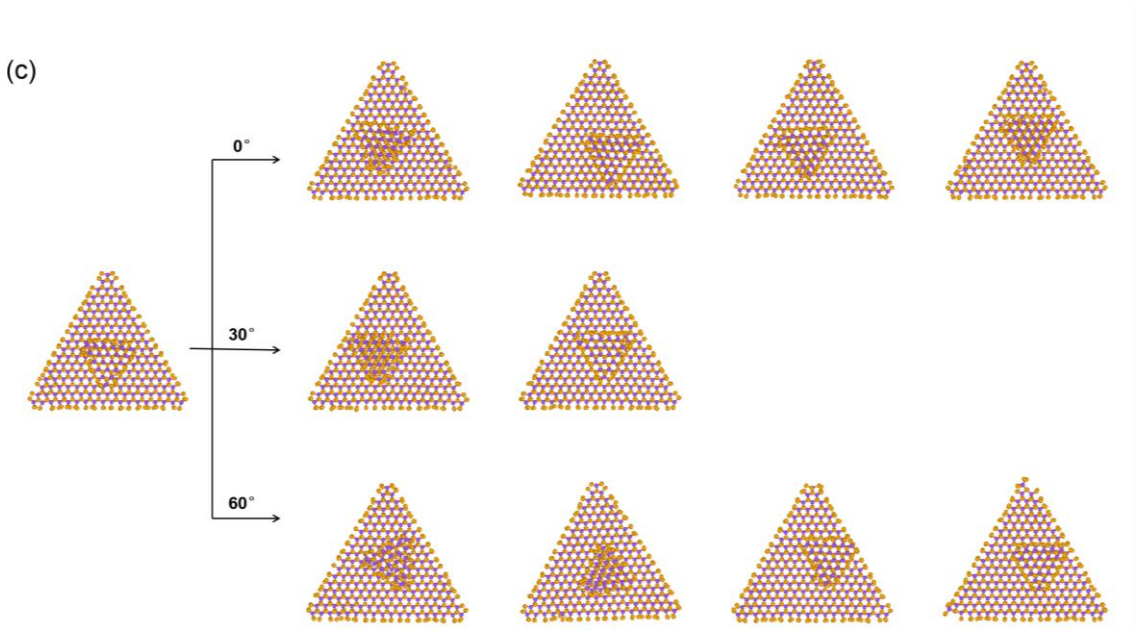
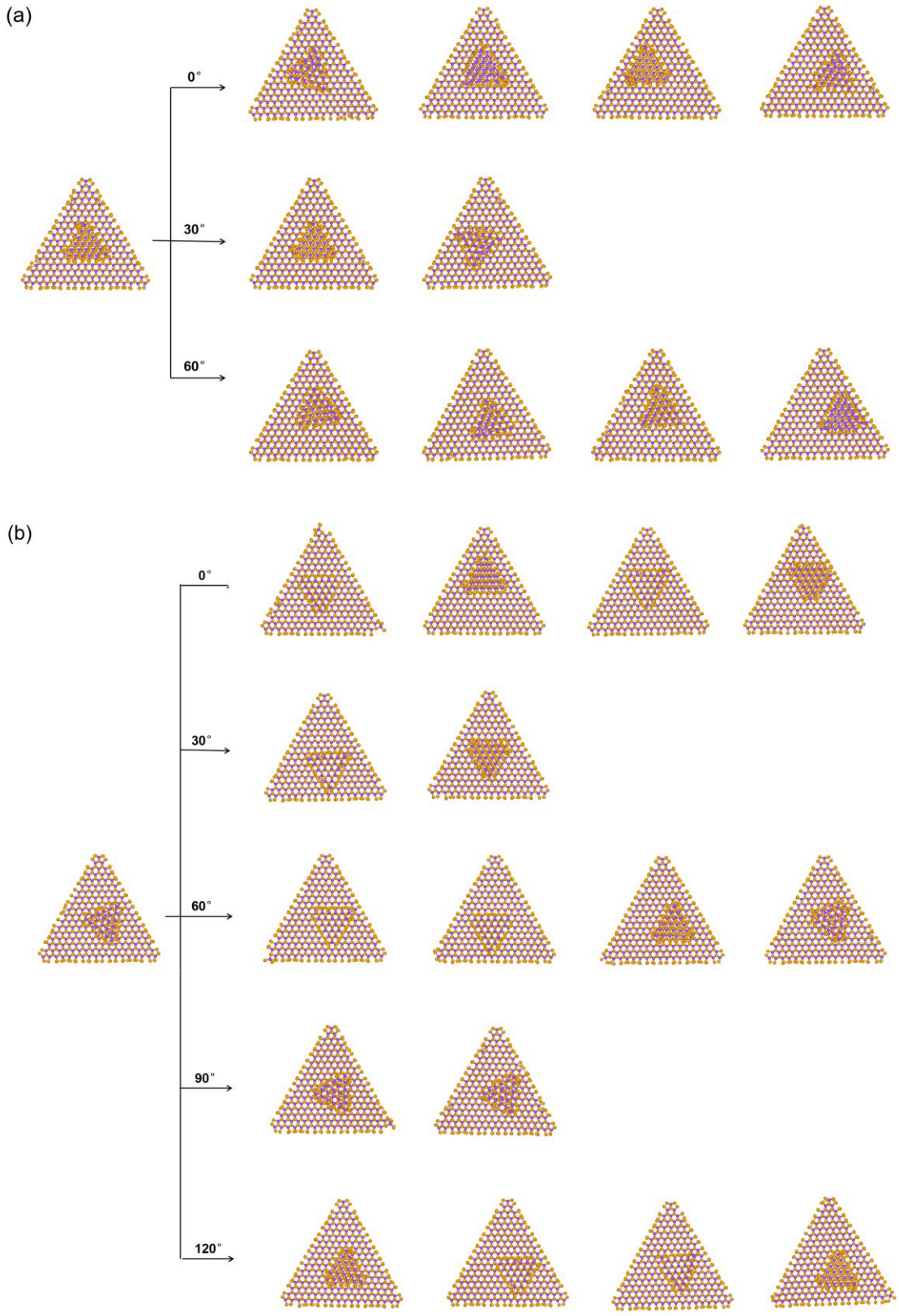


Fig. S2. The corresponding stacking configurations with twist angles of (a) 0°, (b) 30° and (c) 60° after fully relaxed at 300 K.


(a)
0°
30°
60°
(b)
0°
30°
60°
90°
120°

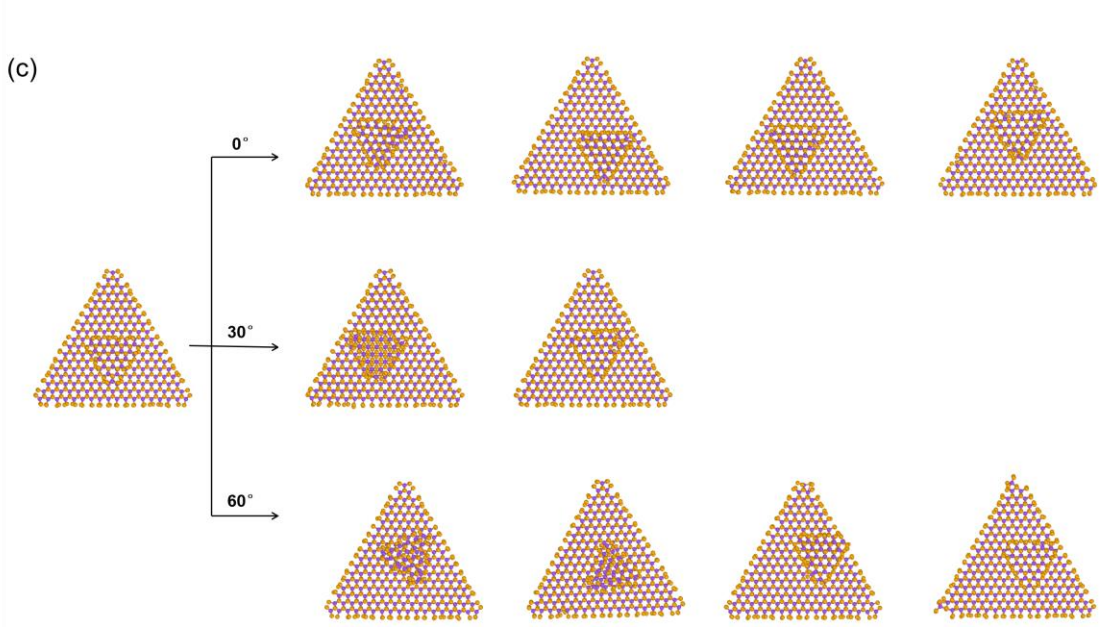


Fig. S3. The corresponding stacking configurations with twist angles of (a) 0°, (b) 30° and (c) 60° after fully relaxed at 0.3 K.

Table S1 0° stacking configurations simulated in our study at minimized state, 300 K and 0.3 K.

0° stacking configurations	Minimized state	300 K	0.3 K
Structure 1 (along 0° direction)	AB stacking	AB’	AB’
Structure 2 (along 0° direction)	AB stacking	twisted	twisted
Structure 3 (along 0° direction)	AB stacking	AB’’	AB’’
Structure 4 (along 0° direction)	AB stacking	AB’	AB’
Structure 5 (along 0° direction)	AB stacking	twisted	twisted
Structure 2 (along 30° direction)	AB stacking	AB’	AB’
Structure 3 (along 30° direction)	AB stacking	twisted	twisted
Structure 2 (along 60° direction)	AB stacking	twisted	twisted
Structure 3 (along 60° direction)	AB stacking	AB’’	AB’’
Structure 4 (along 60° direction)	AB stacking	AB’	AB’
Structure 5 (along 60° direction)	AB stacking	AB’’	AB’’

Table S2 30° stacking configurations simulated in our study at minimized state, 300 K and 0.3 K.

30° stacking configurations	Minimized state	300 K	0.3 K
Structure 1 (along 0° direction)	AC stacking	twisted	twisted
Structure 2 (along 0° direction)	AC stacking	AA’	AA’
Structure 3 (along 0° direction)	AC stacking	AB’	AB’
Structure 4 (along 0° direction)	AC stacking	AA’	AA’
Structure 5 (along 0° direction)	AC stacking	AA’’	AA’’
Structure 2 (along 30° direction)	AC stacking	AA’	AA’
Structure 3 (along 30° direction)	AC stacking	AA’’	AA’’
Structure 2 (along 60° direction)	AC stacking	AA’	AA’
Structure 3 (along 60° direction)	AC stacking	AA’	AA’
Structure 4 (along 60° direction)	AC stacking	AB’	AB’
Structure 5 (along 60° direction)	AC stacking	twisted	twisted
Structure 2 (along 90° direction)	AC stacking	twisted	twisted
Structure 3 (along 90° direction)	AC stacking	twisted	twisted
Structure 2 (along 120° direction)	AC stacking	AB’	AB’
Structure 3 (along 120° direction)	AC stacking	AA’	AA’
Structure 4 (along 120° direction)	AC stacking	AA’	AA’
Structure 5 (along 120° direction)	AC stacking	AB’	AB’

Table S3 60° stacking configurations simulated in our study at minimized state, 300 K and 0.3 K.

60° stacking configurations	Minimized state	300 K	0.3 K
Structure 1 (along 0° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 2 (along 0° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 3 (along 0° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 4 (along 0° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 5 (along 0° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 2 (along 30° direction)	AA’ stacking	AA’’ stacking	AA’’ stacking
Structure 3 (along 30° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 2 (along 60° direction)	AA’ stacking	twisted	twisted
Structure 3 (along 60° direction)	AA’ stacking	twisted	twisted
Structure 4 (along 60° direction)	AA’ stacking	AA’ stacking	AA’ stacking
Structure 5 (along 60° direction)	AA’ stacking	AA’ stacking	AA’ stacking