Electronic Supplementary Material (ESI) for Nanoscale.

The favorite of large misorientationanglegrain boundaries in graphene

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Supplementary Information

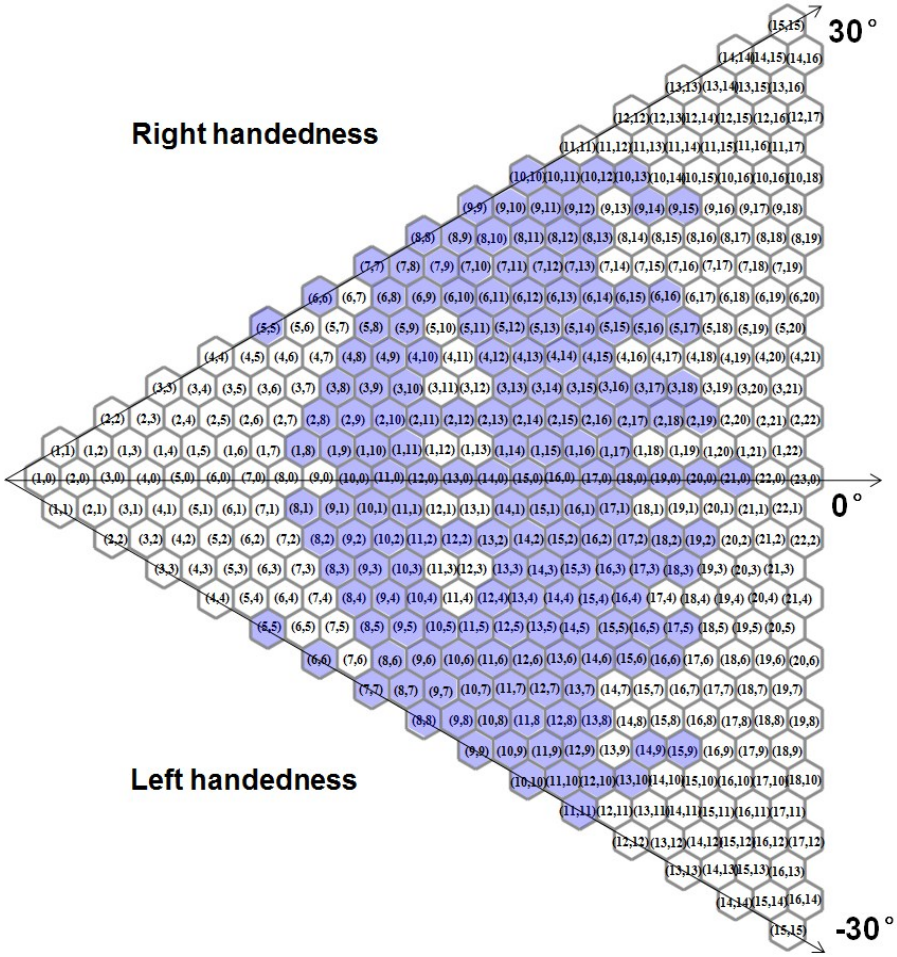

Fig. S1 The map of used carbon nanotube (*n*, *m*) used for constructing the studied GB structures.

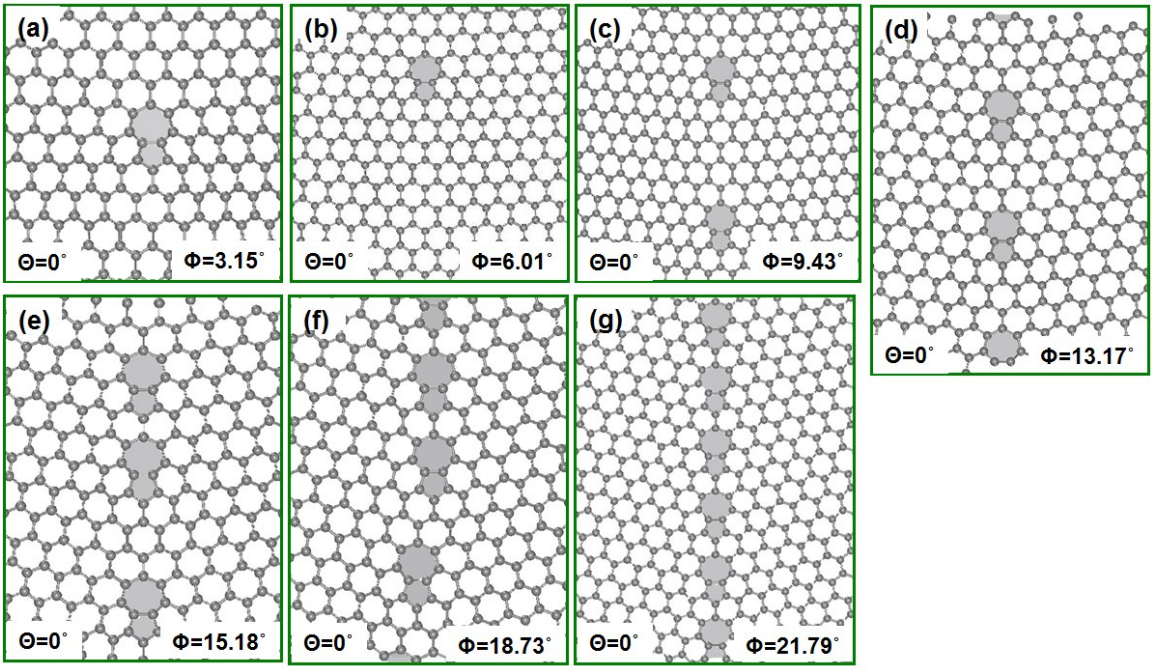

Fig.S2 (a-g) GB-Sstructures at misorientation angles of 3.15°, 6.01°, 9.43°, 13.17°, 15.18°,18.73° and 21.79°.

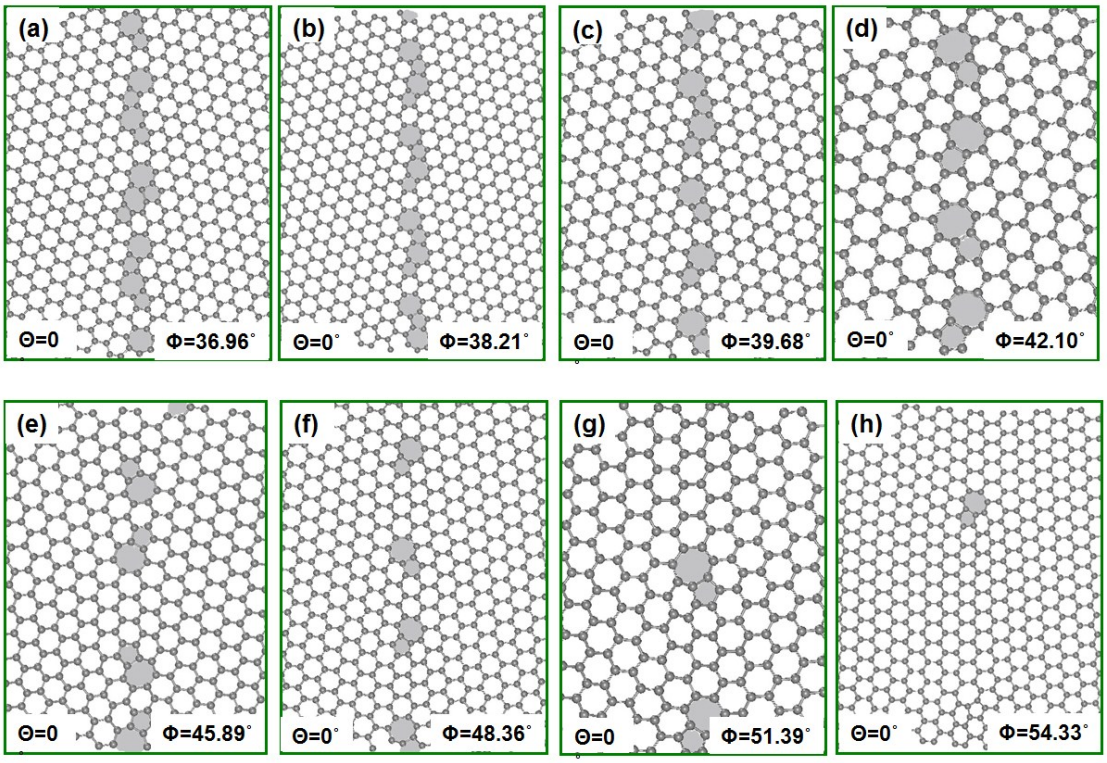

Fig.S3 (a-h) GB-S structures at misorientation angles of 36.96°, 38.21°, 39.68°, 42.10°, 45.89°, 48.36°, 51.39° and 54.33°.

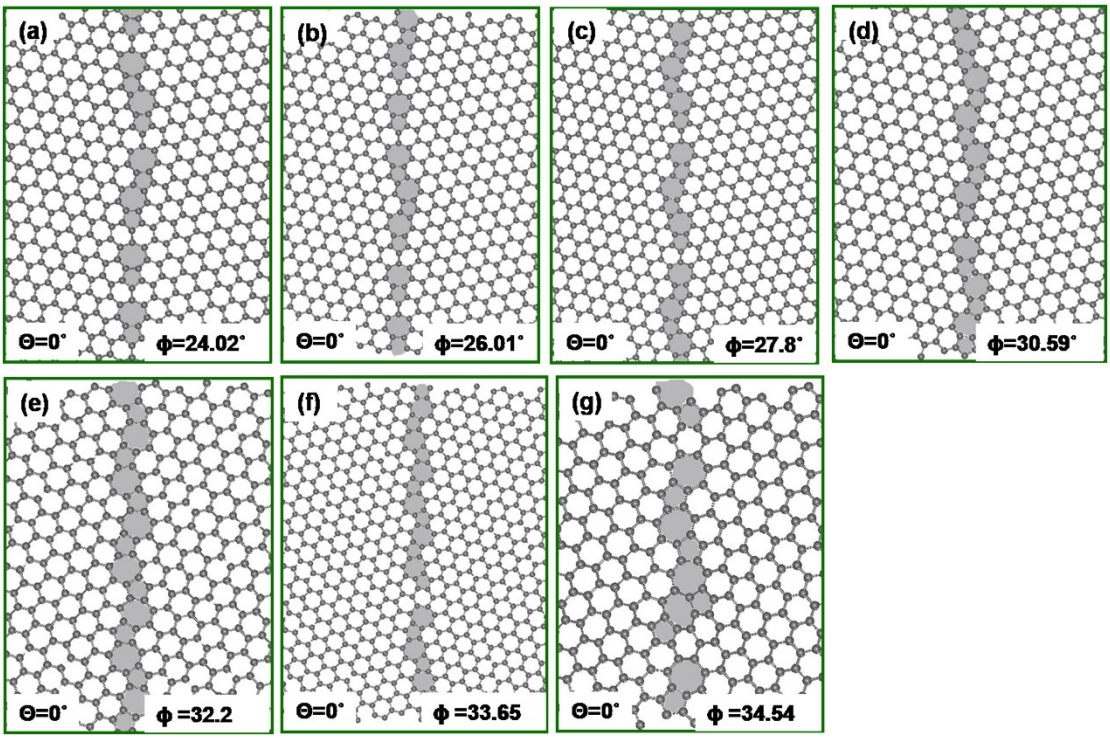

Fig.S4 (a-g) GB-S structures at misorientation angles of 24.02°, 27.80°, 30.59°, 32.20°, 33.65°and 34.54°.

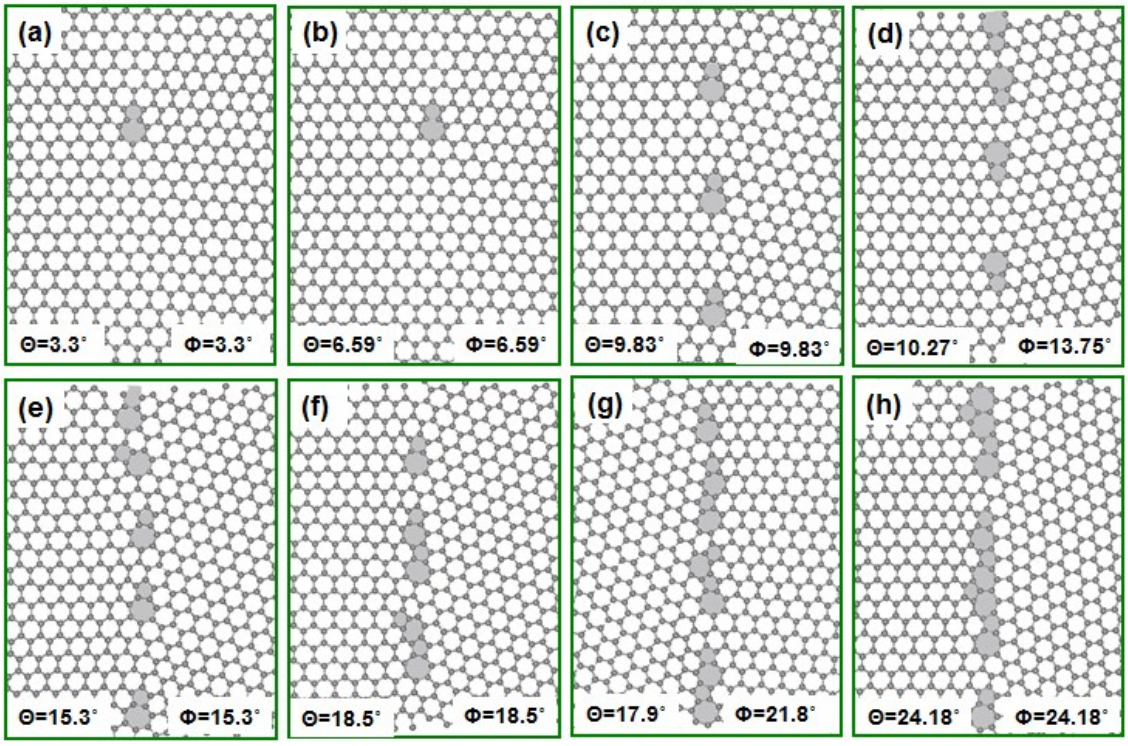

Fig.S5(a-g) GB-LR structures at misorientation angles of 3.3°, 5.63°, 9.83°, 13.75°, 15.3°, 18.5°, 21.8° and 24.18°.

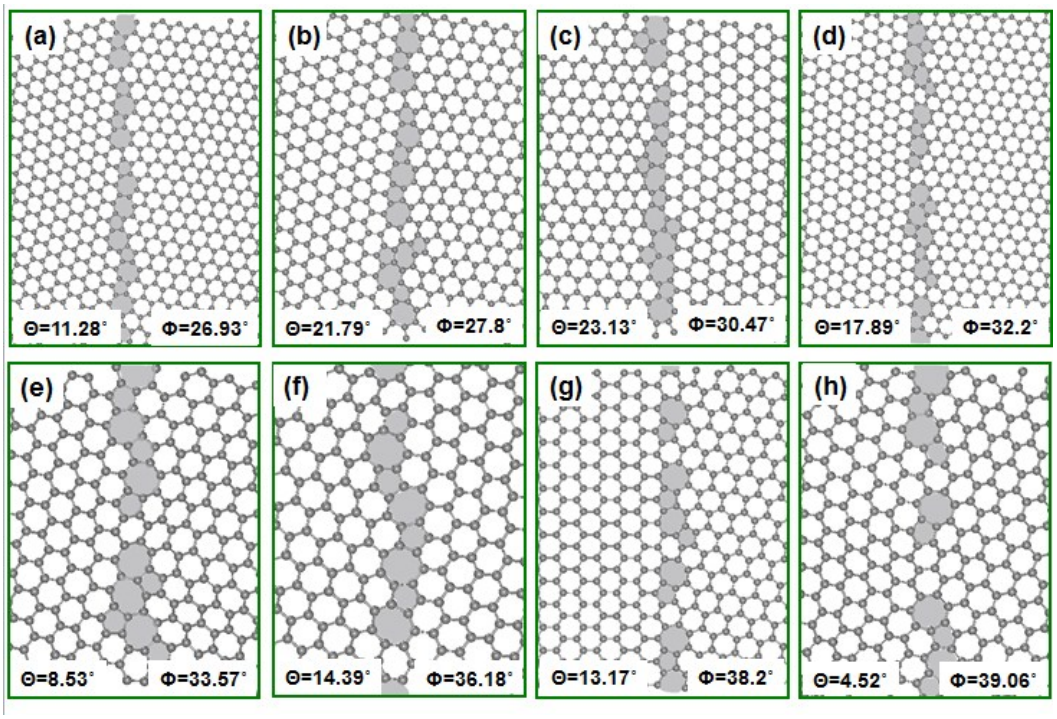

Fig.S6 (a-g) GB-LR structures at misorientation angles of 26.93°, 27.8°, 30.47°, 32.2°, 33.57°, 36.18°, 38.2° and 39.06°.

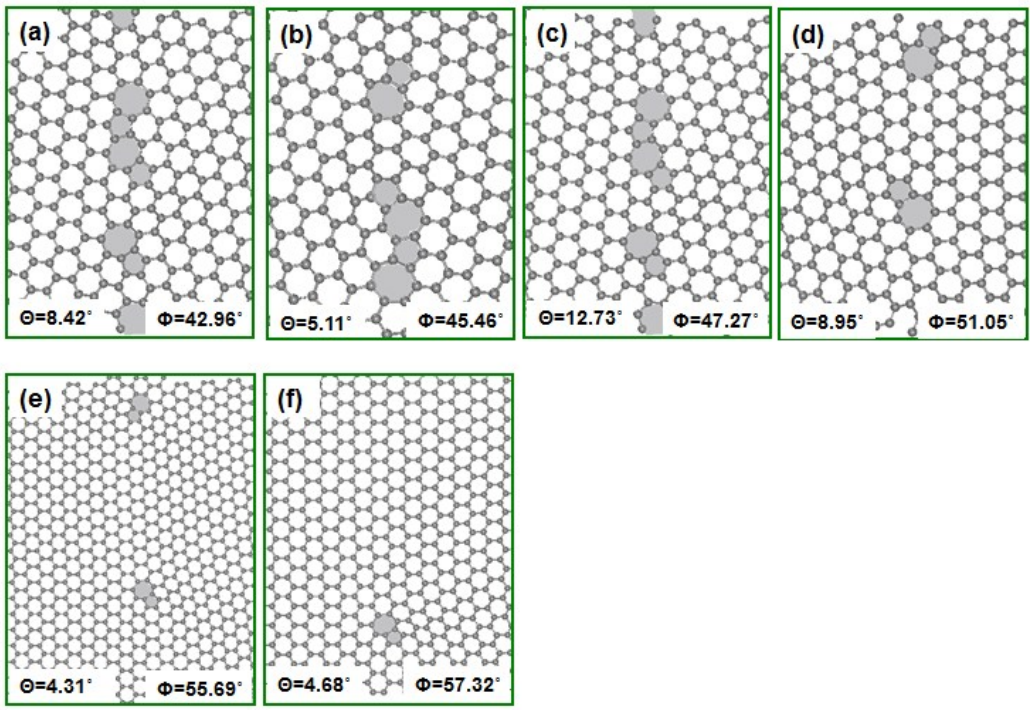

Fig.S7 (a-g) GB-LR structures at misorientation angles of 42.96°, 45.46°, 47.27°, 51.05°, 55.69° and 57.32°.

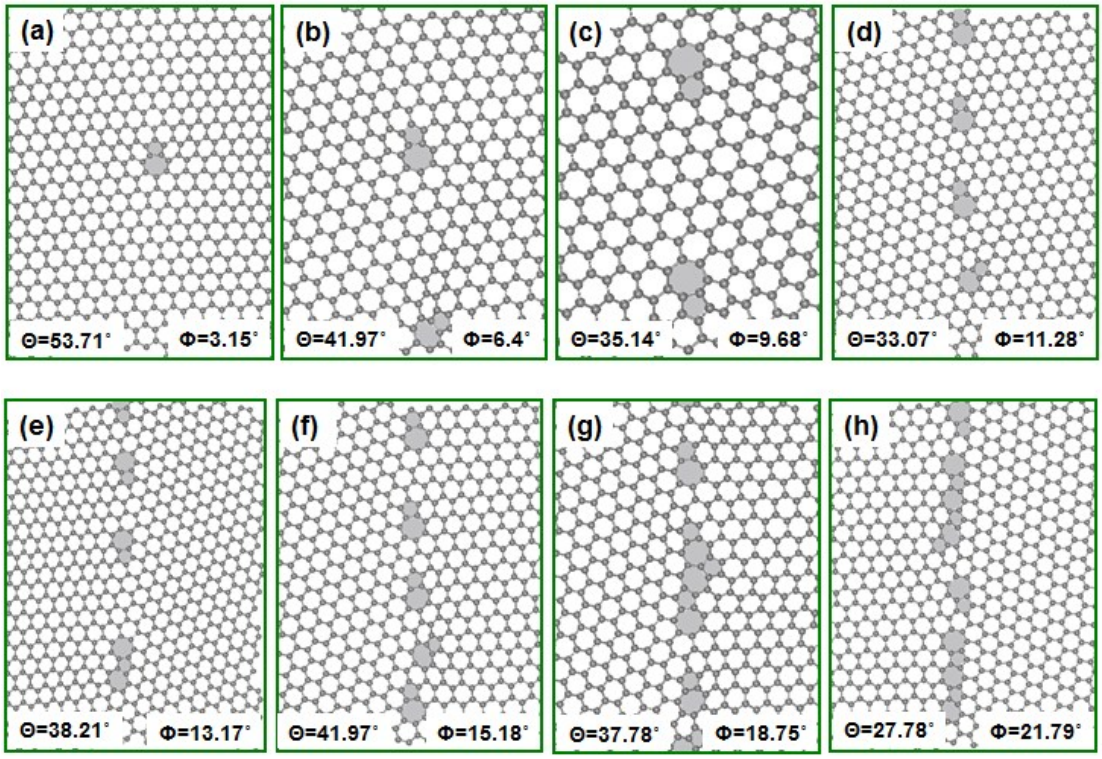

Fig.S8 (a-g) GB-RR structures at misorientation angles of 3.15°, 6.4°, 9.68°, 11.28°, 13.17°, 15.18, 18.75° and 21.79°.

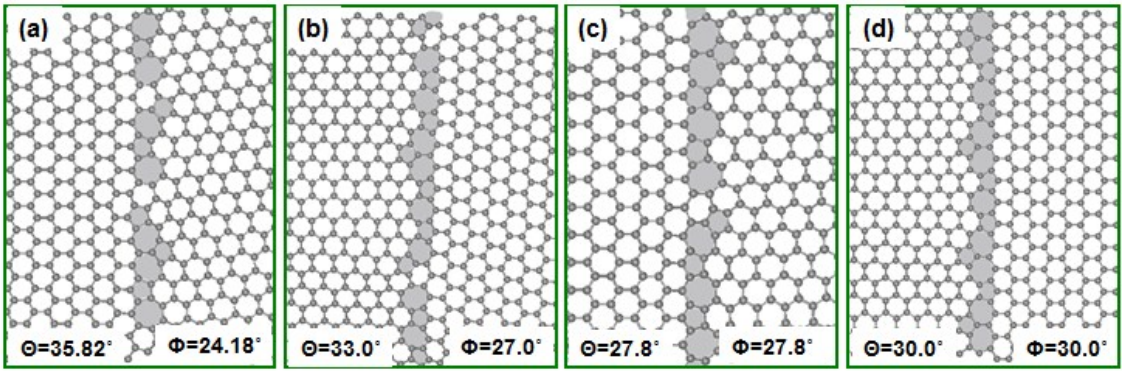

Fig.S9. (a-d) GB-RR structures at misorientation angles of 24.18°, 27.0°, 27.8°and 30°.

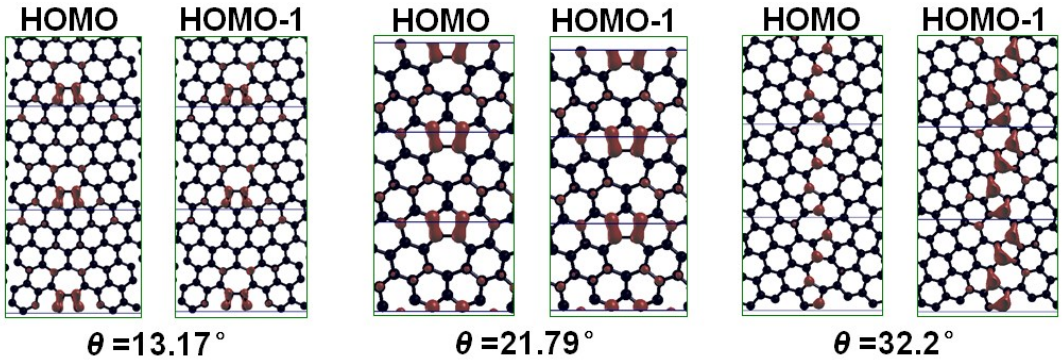

Fig.S10 (a-c) The highest occupied (HOMO) and the second highest occupied (HOMO-1) orbitals of GB-Sswithmisorientation angle of 13.17°, 21.78° and32.2°.

The interaction of two 5|7 dislocations

According to theory of dislocations, the GB energies of these 1D defects can generally be regarded as the summation of linear segment-segment interactions, which interaction energy between two 5|7 dislocations can be expressed as a function of their distance (R),

$$\in$$

$$\varepsilon_{12} = -\frac{\mu(\vec{b}_1 g\vec{\zeta})(\vec{b}_2 g\vec{\zeta})}{2\pi}\ln\frac{R}{R_a} - \frac{\mu}{2\pi(1-v)}[(\vec{b}_1\times\vec{\zeta})g(\vec{b}_2\times\vec{\zeta})]\ln\frac{R}{R_a} - \frac{\mu}{2\pi(1-v)R^2}[(\vec{b}_1\times\vec{\zeta})g\vec{R}][(\vec{b}_2\times\vec{\zeta})g\vec{R}] \quad (1)$$

Where $\vec{b}_1, \vec{b}_2$ are Burgers vectors of two dislocations; $\vec{\zeta}$ represents the dislocation direction; μ is the shear modulus, v is the Poisson' radio, $\vec{R}_a$ is the distance of two dislocations and $\vec{R}$ is a separation between two dislocations. In the 1D GB structures, the Burgers vectors, $\vec{b}_1$ and $\vec{b}_2$ are almost vertical with $\vec{\zeta}$, then equation (1) can be simplified as:

$$\begin{aligned}\varepsilon_{12} &= \frac{\mu}{2\pi(1-v)}|\vec{b}_1||\vec{b}_2|\times(1+\ln R-\ln R_a),(R_a=\frac{1}{\rho_{57}})\\ &= \frac{\mu}{2\pi(1-v)}|\vec{b}_1||\vec{b}_2|\times(1+\ln R+\ln\rho_{57}) \quad (2)\\ &= A+B\ln\rho_{57}\end{aligned}$$

ρ_{57} is the line density of 5|7 dislocations. Such, the GB formation energies per unit length of these 1D GB structures can be described as a function of the line density of 5|7 pairs:

$$E_f = \rho_{57}(A + B\ln\rho_{57}) \quad (3)$$