The favorite of large misorientation angle grain 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-S structures 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°.
The highest occupied (HOMO) and the second highest occupied (HOMO-1) orbitals of GB-S with misorientation angle of 13.17°, 21.78° and 32.2°.

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),

\[ \varepsilon_{12} = -\frac{\mu(b_1 \times \zeta)(b_2 \times \zeta)}{2\pi} \ln \frac{R}{R_a} - \frac{\mu}{2\pi(1-\nu)}[(b_1 \times \zeta)g(b_2 \times \zeta)]\ln \frac{R}{R_a} \\
- \frac{\mu}{2\pi(1-\nu)R^2}[(b_1 \times \zeta)gR][(b_2 \times \zeta)gR] \quad (1) \]

Where \( b_1, b_2 \) are Burgers vectors of two dislocations; \( \zeta \) represents the dislocation direction; \( \mu \) is the shear modulus, \( \nu \) is the Poisson’ radio, \( R_a \) is the distance of two dislocations and \( R \) is a separation between two dislocations. In the 1D GB structures, the Burgers vectors, \( b_1 \) and \( b_2 \) are almost vertical with \( \zeta \), then equation (1) can be simplified as:

\[ \varepsilon_{12} = \frac{\mu}{2\pi(1-\nu)} \left| b_1 \right| \left| b_2 \right| \times (1 + \ln R - \ln R_a) \left( R_a = \frac{1}{\rho_{57}} \right) \\
= \frac{\mu}{2\pi(1-\nu)} \left| b_1 \right| \left| b_2 \right| \times (1 + \ln R + \ln \rho_{57}) \quad (2) \]

\[ = A + B \ln \rho_{57} \]
\( \rho_{57} \) 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:

\[
E_f = \rho_{57}(A + B \ln \rho_{57})
\]

(3)