Supporting Information

Electronic and transport properties of graphene with grain boundaries

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1. The transmission spectra of (2,1)|(2,1) GB using both the LDA and GGA functionals.

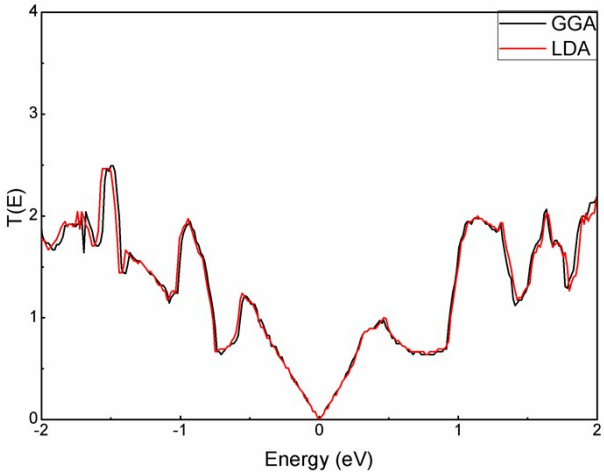

Figure S1. The transmission spectra of (2,1)|(2,1)GB calculated with GGA and LDA

2. The crossed GB derived from the singe GB

Take the most complex (3,1)|(3,1) GB as an example, as see form Figure S2 that the supercell lattice constant d and L are 8.9776 Å and 31.1452 Å respectively, A and B are two points of the located at the core of the GB. If we want to obtain a new supercell ACEF also satisfied with period conditions by rotating AB around A point with an angle θ, the θ has some relationship with d and L. Here point C and E can be

obtained from B and A by making period of md and (m+n)d (m and n are integer). For (3,1)|(3,1)GB, we can get θ=16.07°, 29.96°, 40.85°, 49.06°, 59.96°, 73.87°, namely, if we rotate the GB with one of these angles, we can create a new supercell also satisfied with period. After the new supercell is obtained by rotating 59.96°, (3,1)|(3,1)D can be obtained by doing some other translations and rotations based on the new supercell. The whole process is illustrated in Figure S3-FigureS8.

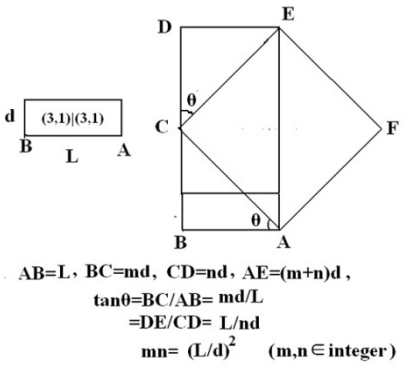

Figure S2.

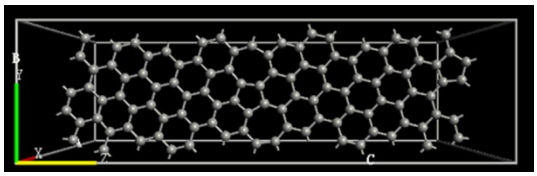

Figure S3. The supercell of (3,1)|(3,1) GB

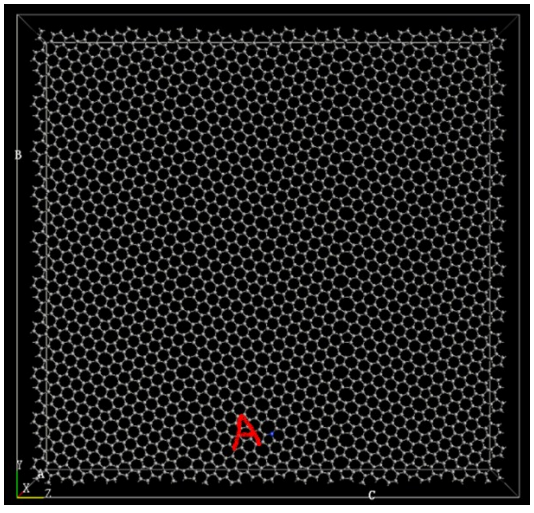

Figure S4. Cleave the supercell with 1*10*3 and select A (blue atom) as the reference point

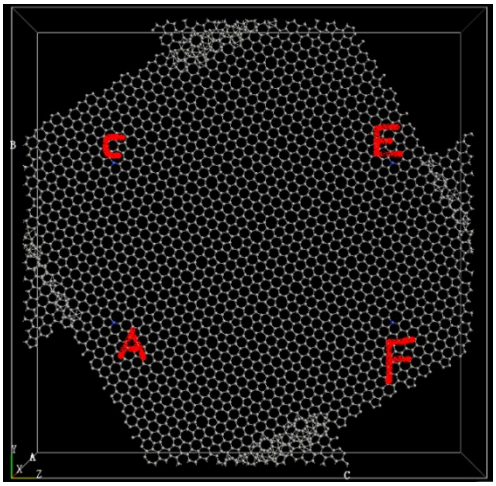

Figure S5. Rotate the above supercell with 59.96° and find the equal points C, E and F (blue atoms)

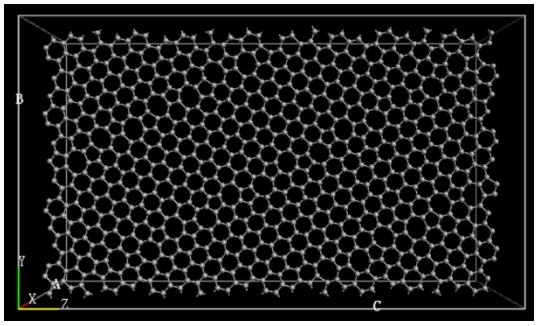

Figure S6. The new supercell created with the lattice constant AC and AF

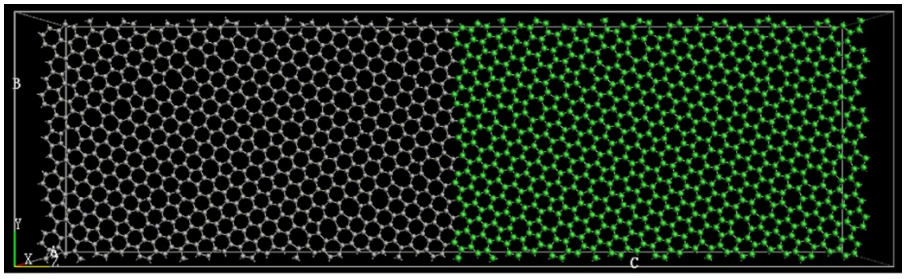

Figure S7. Copy and translate the new supercell with period AF and get the green part

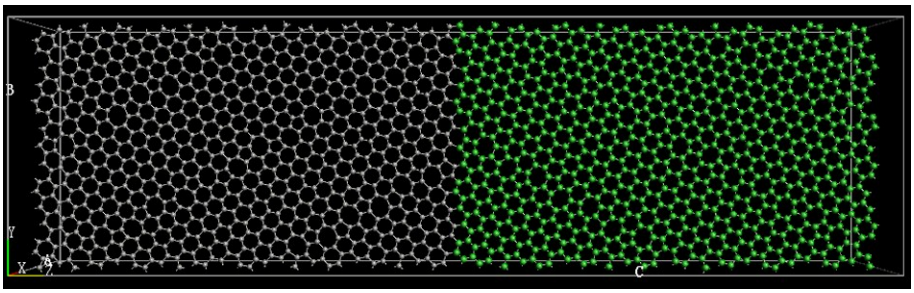

Figure S8. Rotate the green part along AF direction with 180°and then obtain (3,1)|(3,1)D GB

3. **Some benchmark tests of the formation energy of grain boundary to avoid the coupling effects between the neighbour grain boundaries**

We constructed (2,1)|(2,1) GBs with different length between the neighbor grain boundaries, the atom number (N), and the distance (d) between the neighbor grain boundaries. The formation energies are shown in Table S1. We can see that the formation energy has converged with the distance 14.67 Å between two neighbor grain boundaries. Moreover, that distance in other GBs investigated in our manuscript is larger than 14.67 Å, which indicates the coupling effects between the neighbor grain boundaries can be neglected.

Table S1. Different types of (2,1)|(2,1)GBs with the different atom number (N), the distance (d) between the neighbor grain boundaries, and the corresponding formation energies.

GB	5621	6421	7221	8021
N	56	64	72	80
d(Å)	11.42	13.04	14.67	16.30
E_{form} (eV/nm)	3.2868	3.2473	3.2368	3.2348

4. The detail procedures to calculate the transport properties.

The transport properties calculation is a self-consistent iteration process combined Density functional theory (DFT) with nonequilibrium Green's function (NEGF) method. The process is as follows:

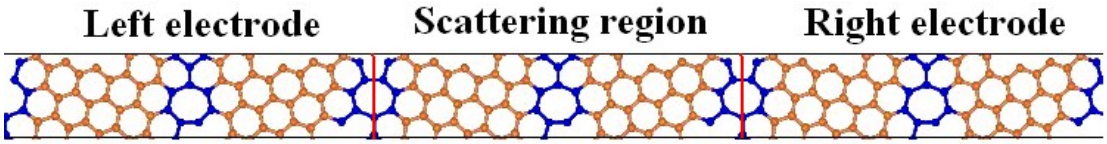

Figure S9. Two-probe systems model

Figure S9 shows a typical non-equilibrium system, which can be separated into three parts, the left electrode, the scattering region and the right electrode. The Hamiltonian of two electrode H_L and H_R can be obtained by calculated the electrode bulk system with period conditions. The coupling of the electrodes with the scattering region is taken into account using self-energies, Σ_L and Σ_R. The Hamiltonian of the scattering region H can be obtained by self-consistent DFT calculations,

$$H_{i,j} = \left\langle i \left| -\frac{\hbar^2}{2m}\nabla^2 + U_{KS}(\rho) \right| j \right\rangle \qquad (1)$$

From the Hamiltonian H, we can get the retarded Greens function G,

$$G(E) = \left[E - H - \Sigma_L - \Sigma_R\right] \qquad (2)$$

Using the retarded Greens function G, we can obtain the lesser Green's function $G^<$,

$$G^{<}(E) = iG(E)\left[\Gamma_L f\left(E-\mu_L\right)+\Gamma_R f\left(E-\mu_R\right)\right]G^{\dagger}(E) \qquad (3)$$

where Γ_L and Γ_R is the contact broadening functions associated with the left and right electrodes.

The non-equilibrium density matrix can be constructed by the lesser Green's function,

$$\rho = \frac{1}{2\pi i}\int dE G_M^{<}(E) \qquad (4)$$

Then, taking the non-equilibrium density matrix ρ into the equation (1), doing self-consistent cycles until the density matrix ρ of the system reaches its convergence criteria. The whole process is shown in figure S10.

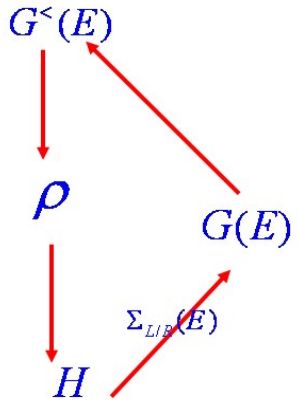

Figure S10. The self-consistent cycle.

After we obtained the convergent density matrix ρ, we can obtain H, G(E), Γ_L and Γ_R and transmission coefficient $T(E,V) = Tr\left[\Gamma_L G^{\dagger}(E)\Gamma_R G(E)\right]$,

Thus, the current can be obtained based on the Landauer-Büttiker formula:

$$I = G_0\int T(\mathrm{E},V)[f_L(E)-f_R(E)]\,\mathrm{d}\,E \qquad (5)$$