

Electronic properties and quantum transport behavior of twisted γ -graphdiyne/graphene

Yingcong Liu,^{1,2,#} Haokun Bai,^{1,2,#} Xin Wang,^{1,2} Haochun Sun,^{1,2} Zhaozhao Xiong,^{1,2} Fulong Dai,^{1,2} Zhuojian Liang,^{1,2} Zhuo Kang,^{1,2,*} and Yue Zhang,^{1,2,*}

¹Academy for Advanced Interdisciplinary Science and Technology, Key Laboratory of Advanced Materials and Devices for Post-Moore Chips Ministry of Education, State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing, Beijing 100083, China.

²School of Materials Science and Engineering, Beijing Key Laboratory for Advanced Energy Materials and Technologies, University of Science and Technology Beijing, Beijing 100083, China.

#Contributed equally to this work.

*Corresponding authors (emails: zhuokang@ustb.edu.cn (Zhuo Kang); yuezhang@ustb.edu.cn (Yue Zhang))

KEYWORDS: γ -graphdiyne, moiré superlattice, transport behavior, transmission spectra

1. Relaxed Structure of TGG

In order to satisfy the constructed heterojunction interface matching, we adopted the following theory^[1, 2]. First, the base vector of material A (B_1, B_2) is rotated θ to obtain (C_1, C_2), and a new base vector (V_1, V_2) is obtained by operating the base vector (C_1, C_2) according to the following formula:

$$V = nC_1 + mC_2$$

A new base vector (U_1, U_2) is obtained by $U = nA_1 + mA_2$ operation on the base vector (A_1, A_2) of material B. The (U_1, U_2) and (V_1, V_2) are stretched and compressed, and the A-B heterostructure is obtained by matching. Finally, the structure with small cell strain and orthogonal base vector was selected. According to the above method, we set the origin of GDY and graphene crystal cell as (0, 0), make the six-member ring center are located in the origin, as shown in Figure S1. We defined the lattice vectors L_1 and L_2 of the graphene-graphene superlattice as the least common multiples of the protocell vectors ($a_1(1), a_2(1)$), ($a_3(2), a_4(2)$) of the two single-layer two-dimensional materials, GDY and graphene. L_1 can be expressed as:

$$L_1 = xa_1(1) + ya_2(1) = x'a_3(2) + y'a_4(2)$$

($a_1(1), a_2(1)$) as the original cell monolayer graphene vector, ($a_3(2), a_4(2)$) as the original cell monolayer GDY vector. L_2 is made by rotating L_1 at a twist angle. Then all twisted angles are obtained by matching these two structures.

The twisted γ -graphdiyne/graphene moiré superlattices open a new avenue for quantum state manipulation in twistrionics, highlighting its substantial potential in the future. At present, the mainstream research on twisted γ -graphdiyne/graphene is still at the stage of theoretical analysis, and the experimental synthesis and verification are only just getting underway.

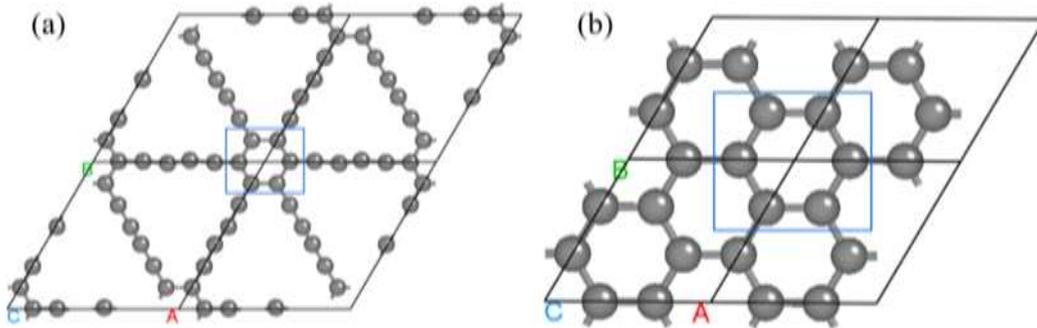


Figure S1. Schematic diagram of GDY and graphene crystal cell. (a) Schematic diagram of GDY protocell. (b) Schematic diagram of graphene protocell.

2. Optimization of Interlayer Spacing for TGG Structure

After the construction of the TGG structures, the models need to be further optimized. In order to optimize the layer spacing^[3], we search for the model that can match the best fit rate. To be specific, we calculate the Hartree energy of the constructed model under different layer spacing models, and find the layer spacing corresponding to the minimum Hartree energy, that is, the layer spacing in an ideal case. Taking the 0° model as an example, the layer spacing of the two materials was set at 2.5 Å, and the self-consistent calculation was carried out every 0.01 Å increase, as shown in Figure S2. Finally, the layer spacing corresponding to the lowest total energy was obtained by spline difference method, namely 2.89 Å. In the calculation of transport properties, the overall structure is divided into the left electrode, the right electrode and the central scattering region.

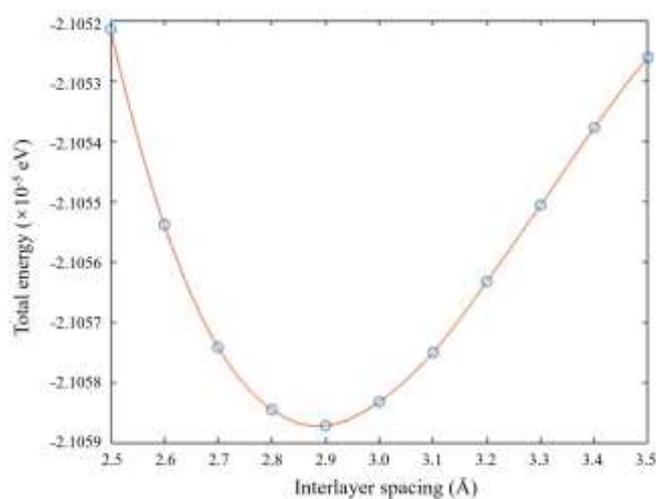


Figure S2. Curve of Hartree energy with layer spacing

3. The Optimized TGG Structure Diagram

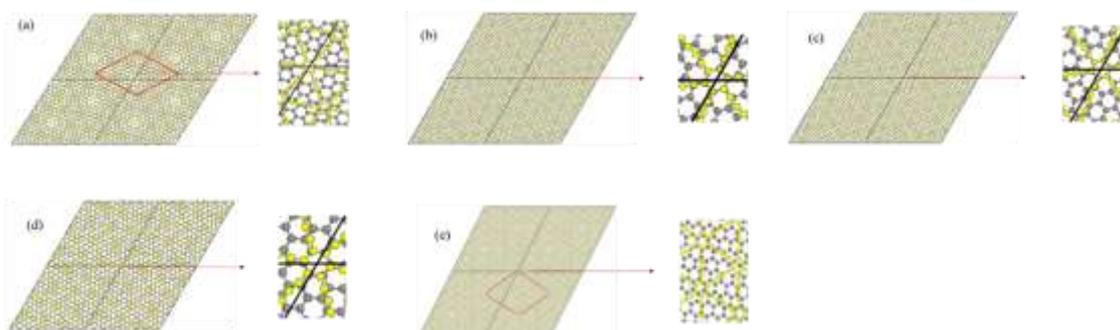


Figure S3. The constructed relaxed crystal cell. (a) with a twist angle of 1° (b) with a twist angle of 2° (c) with a twist angle of 4.5° (d) with a twist angle of 6° (e) with a twist angle of 10° .

4. The Band Structures of TGG

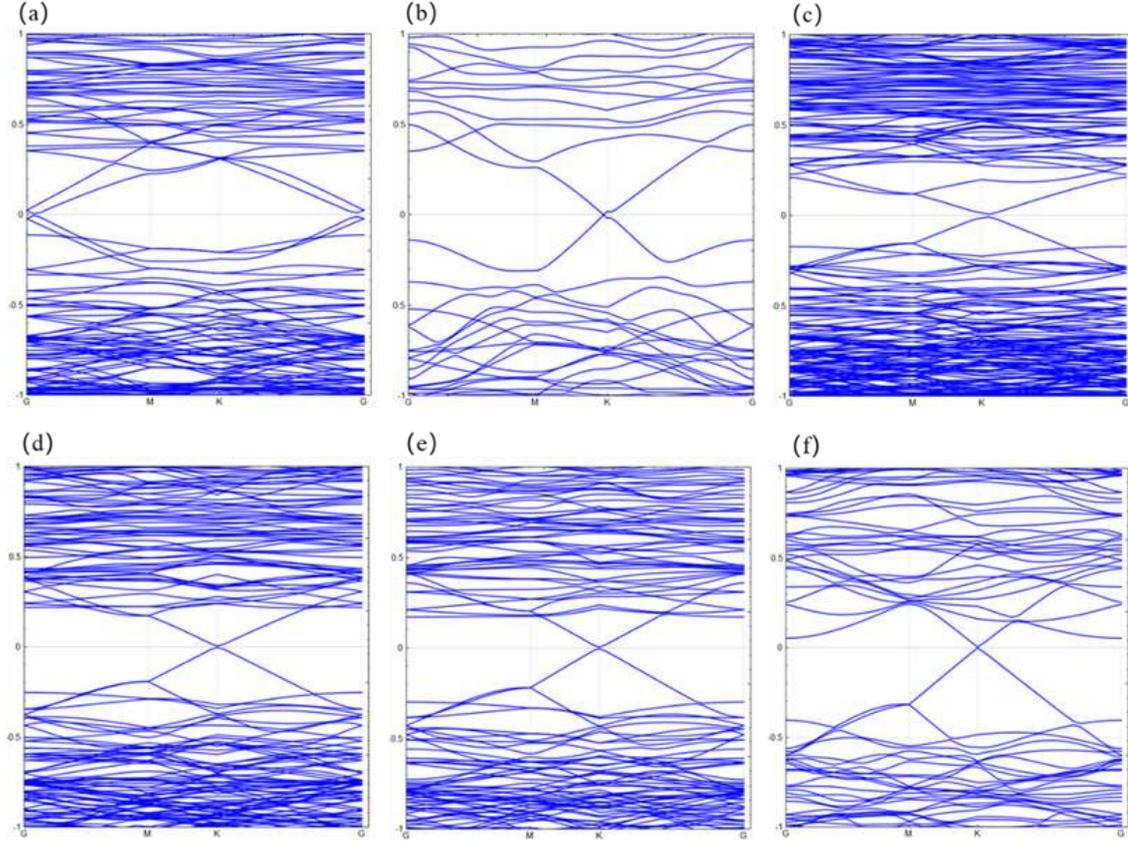


Figure S4. Band structure of TGG structure. (a) twisted structure at 0° (b) twisted structure at 1° (c) twisted structure at 2° (d) twisted structure at 4.5° (e) twisted structure at 6° (f) twisted structure at 10° .

5. Electrostatic Potential Difference of TGG

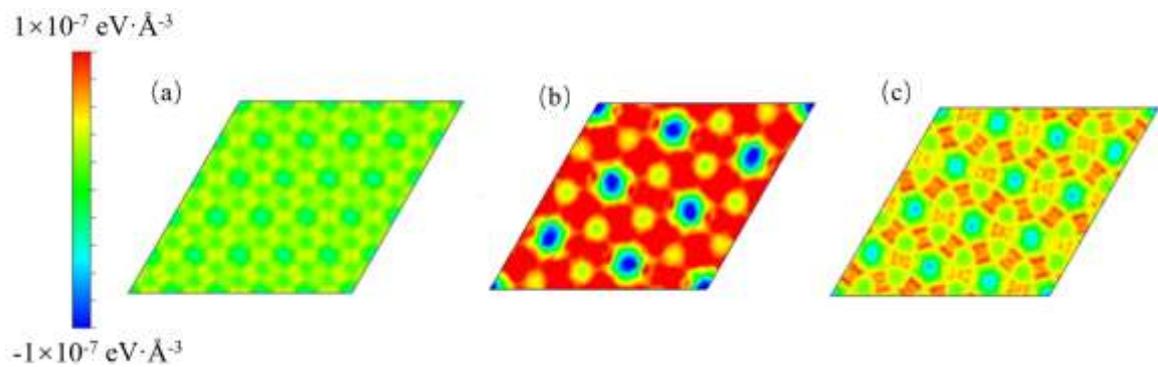


Figure S5. (a) Potential distribution at 0° (b) Potential distribution at 1° (c) Potential distribution at 10° .

6. Electronic Transport Properties of The Moiré Superlattice under Direction Perturbation

In fact, studies have reported that graphdiyne exhibits anisotropic transport properties between the X_1 and X_2 directions, with its electronic transport behavior showing a dependence on directional perturbations.^[4] Based on this observation, a comprehensive characterization of the transport properties along the two perpendicular directions in the γ -graphdiyne/graphene moiré superlattice was carried out, further confirming the variations in electron transport under directional perturbation.

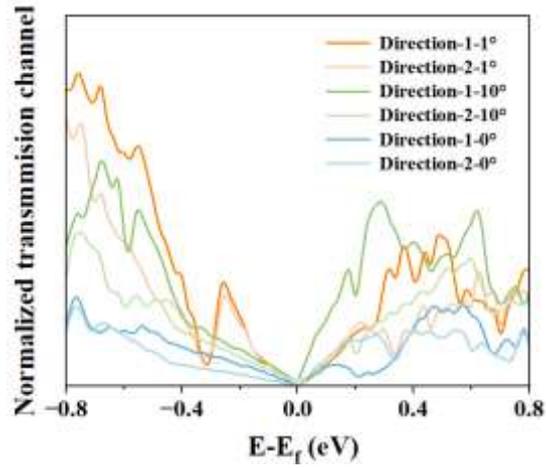


Figure S6. Directional characteristics of electronic transport in twisted γ -graphdiyne/graphene moiré superlattices.

References

- [1] Ishikawa R, Lugg N R, Inoue K, et al. Interfacial atomic structure of twisted few-layer graphene [J]. *Sci Rep* 2016; **6**: 21273.
- [2] Lopes dos Santos J M B, Peres N M R, Castro Neto A H. Continuum model of the twisted graphene bilayer [J]. *Phys Rev B* 2012; **86**: 155449
- [3] Xiao Z, Shupeng S, Huiqi L, et al. First principles study on the electronic structure and optical properties of graphene/MoS₂ heterojunctions with different rotation angles [J]. *Chinese Journal of High Pressure Physics* 2024; **38**: 052201.
- [4] Padilha J E, Fazzio A, da Silva A J R. Directional control of the electronic and transport properties of graphynes [J]. *J Phys Chem C* 2014; **118**: 18793-18798.