Properties of tri-layer and quad-layer graphene systems

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BACKGROUND

Twistronics is the study of how changing the angle between layers in a two-dimensional system changes its electronic properties. Materials such as twisted bilayer graphene were shown both theoretically and experimentally to exhibit superconductive and non conductive states at a 'magic angle' of 1.16°(depending on temperature)[1].

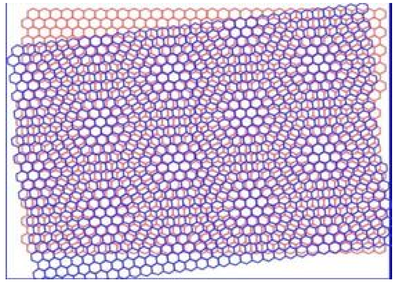

Figure 1: Moire pattern arising from twisting two graphene layers relative to each other. Image: Eva Andrei/Rutgers University-New Brunswick.

This project encompasses the study of two novel twisted graphene systemss - AtAB (A twisted on AB bi-layer graphene) and AtABC (A twisted on ABC tri-layer graphene).

SYSTEM

AB and ABC are graphene layers placed on each other are shown in figure 2. A further layer is then twisted and placed onto the A layer (in phase) to give AtAB and AtABC respectively (shown in figure 3 and 4). This is the repeating unit of the two twisted graphene systems.

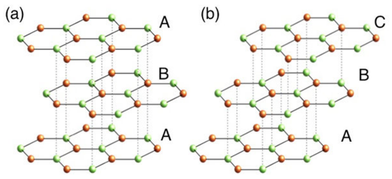

Figure 2: Different stacking patterns for graphene. Credit: Katsuaki Sugawara

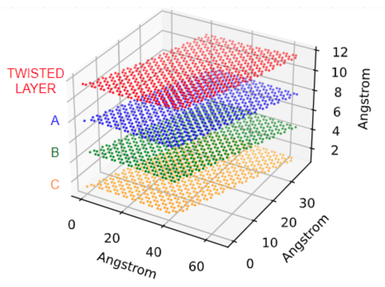

Figure 3: AtABC at an angle of 3.15°. This is the unit cell of this system, having 2648 Carbon atoms in it.

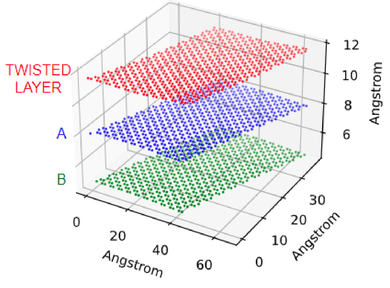

Figure 4: AtAB at an angle of 3.15°. This is the unit cell of this system, having 1986 Carbon atoms in it.

MOTIVATION

The exotic properties exhibited by twisted bilayer graphene has caused twisted graphene systems to garner interest, with researchers motivated by the prospect of finding interesting and unexpected electronic properties like superconductivity, topological states, insulating states etc. While such properties can be found in other materials, twisted systems allow for their tunability.

This has led us to study the AtAB and AtABC systems not only to search for interesting properties, but also to find key differences between these systems and twisted bilayer graphene and hopefully understand the fundamental physical cause of these differences.

METHOD

Tight binding is a method in solid state physics to calculate electronic structures of systems; it provides single electron states using the assumption that the wavefunction of the electron at a point is a superposition of the wavefunctions of an electron at isolated atoms at each atomic site (does not account for electron correlation). The Hamiltonian for our system is then simply that for isolated atoms adjusted for wavefunction overlap between atoms(Fig. 5).

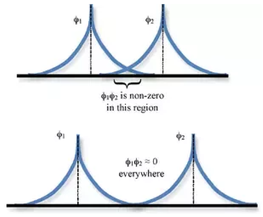

Figure 5: Tight binding takes into accounting the overlap between the orbitals of two separate atoms. Credit: Gautam Shine

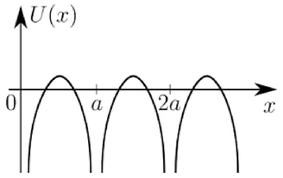

Figure 6: In tight binding, a crystal is modelled as having a periodic potential, with the potentials centered at every lattice point. Credit: TU Delft, Open Solid State Notes

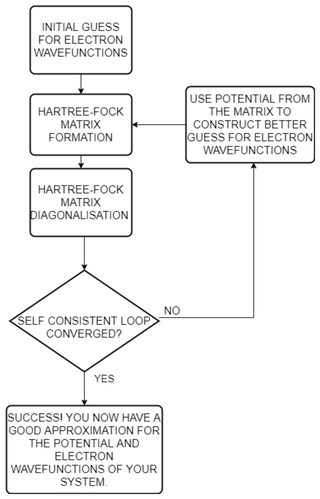

Figure 7: Flowchart explaining the working of the Hartree-Fock self consistent loop

The term accounting for wavefunction overlap is the hopping integral, expressing the energy interaction term between the mth and lth orbital of two different atoms.

$$\gamma_{m,l} = \int \psi_m(\vec{r}) \Delta U(\vec{r}) \psi_l(\vec{r} - \vec{R_n}) \tag{1}$$

For our model, these tight binding parameters are calculated in advance using a semi-empirical approach by fitting these to more precise DFT calculations.

To accurately describe the electronic properties, we cannot ignore the effective potential from the electrons and how that affects the on-site energies. Therefore we integrate into this a self consistent potential calculation, this is called **Hartree-Fock theory**. The Hartree potential at a point is the effective potential created by the sea of electrons at that point.

RESULTS

An interesting find was that AtAB and AtABC have nearly identical properties, in-spite of the extra layer in the case of ABC. Due to their similarity, the subsequent results are true for both systems.
Unlike tBLG (Twisted Bilayer Graphene), the initial results show very little redistribution of electrons and therefore a small magnitude in Hartree potential.

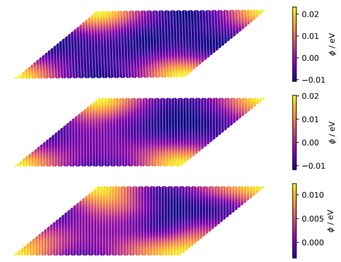

Figure 8: Hartree Potential at the Magic angle

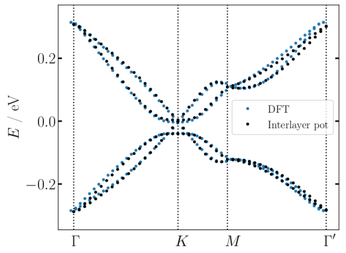

Figure 9: Band structure for DFT vs with 50 meV interlayer potential

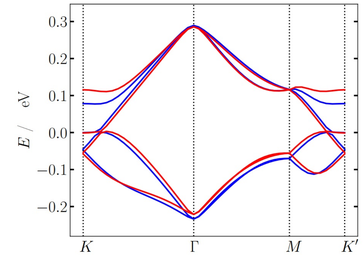

Figure 10: Band Structure with Hartree (blue) and without Hartree(red) with 100 meV/layer applied potential

To make our band structures fit that given by DFT, we must insert an inter-layer potential (adding potential to inner layers) of 50 meV. Therefore, DFT predicts a redistribution of electrons away from the inner layer. This graph also illustrates that an interlayer potential gives rise to a splitting at the K-point with this electron distribution.

The application of an external potential is expected to cause a similar redistribution of electrons. Indeed we again observe a splitting at the K point caused by an the external potential applied across the layers.

This separation at K varies linearly in magnitude with respect to the applied potential. The magnitude of the split is measured with respect to the Dirac Cone, hence the presence of negative values.

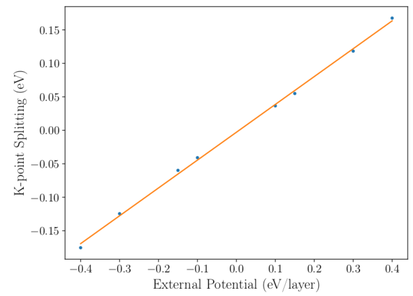

Figure 11: Linear behaviour of K-Point splitting under External Potential.

REFERENCES

[1] Oh M. Wong D. et al. Nuckolls, K.P.
Unconventional superconductivity in magic-angle graphene superlattices.
Nature, 588:610–615, 2020.