

Moire Superlattice-Twisted Bilayer Graphene

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1 Abstract

Lattice of a material is a 3D array of points of the position of atoms,molecules or ions which denote the periodicity of crystal. Now if we take two or more such ultra thin 2D lattices and stack them one above other,then the resultant periodic structure is a superlattice. There are mainly two techniques to create a super lattice i.e 1.Molecular Beam Epitaxy 2.Vanderwaal Structures. Molecular Beam epitaxy is a layer by layer deposition of different materials on a substrate, but it is a slow,labourious process in which we can't change the crystallographic orientation of the material as it totally depends on substrate used. Also if lattice constants of two or more layers are not in a certain range then it creates strain issues which may lead to crystal breaking.

2D materials is the main focus of research since the discovery of graphene in 2004. 2D materials are covalently bonded in 2 dimensions,if we stack them one above the other then they get weakly bonded with the neighbouring layers by Vanderwaal Interactions. Hence it is feasible to isolate,mix and match,introduce rotation/twist in different/same material's atomic 2D layers, to create extremely clean interface of wide range of Vander Waal structures without constraints of lattice mismatching and crystallographic orientation having unique mechanical,optical and electrical properties. As any 2D materials can be easily obtained by just literally scotch taping the 3D bulk materials and can be stacked one on top of other hence there are infinitely many possibilties of Van-Der Waal Structures.

In this paper I will extensively discuss Twisted Bilayer Graphene which at a certain magic angle shows superconductivity,corelated insulating phase,Orbital Magnetism,Anamolous Quantum Hall Effect,Flat Band Structure,Nematicity,Moire Ferro-Electricity etc. Moire Structures combines 2D vanderwaal Materials, strongly corelated materials and Topological Condensed Matter Physics in one platform which can be used to study many body interaction physics.

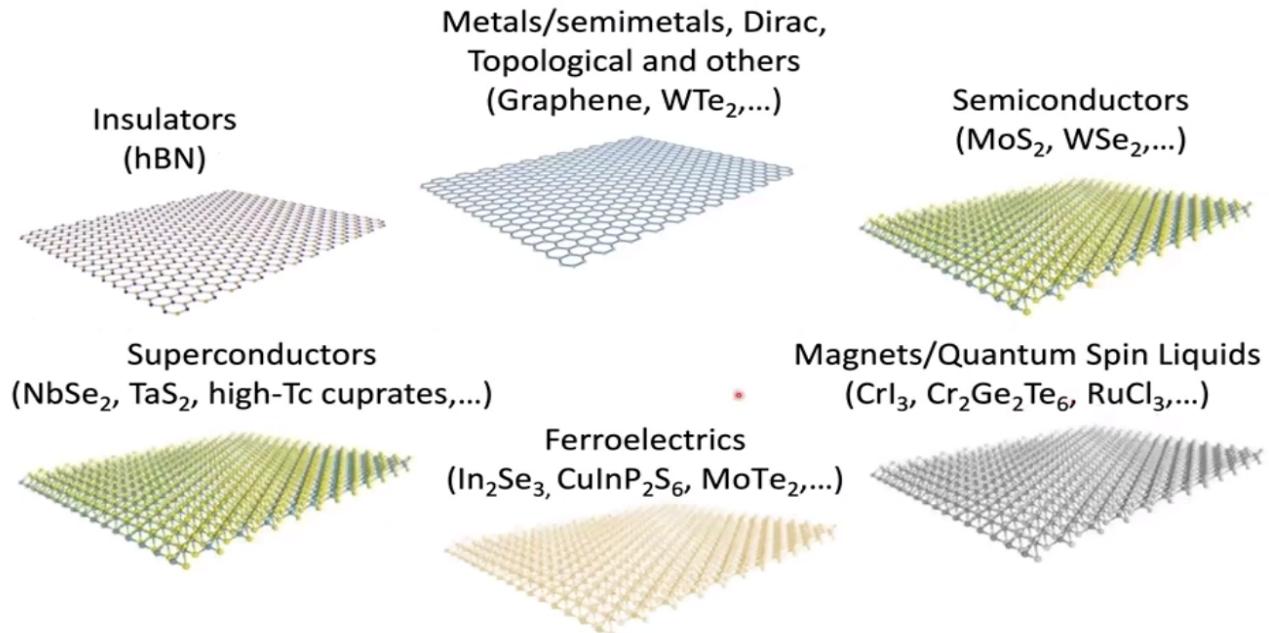


Figure 1: Infinite Possibilities Of Moire Quantum Universe

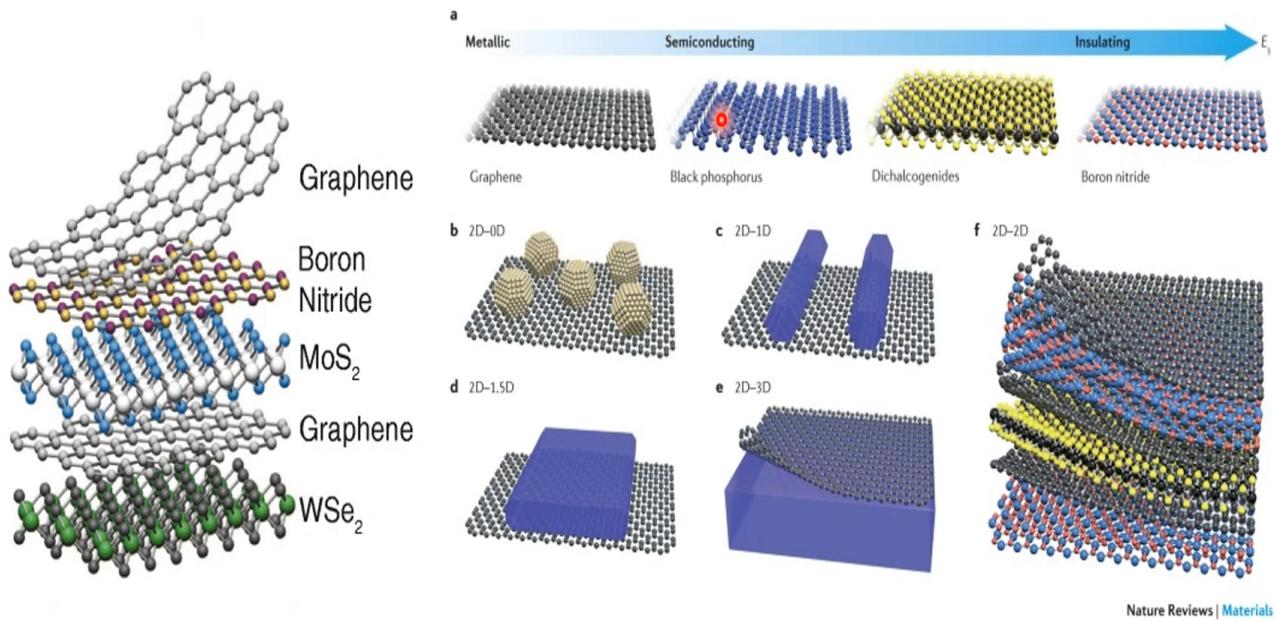


Figure 2: Van-Der Waals Structures-Beyond MBE

2 Introduction

Graphene a 2D material obtained by literally scotch taping 3D Graphite Bulk Material which is bond between hexagonal shaped rings of carbon atoms. Now if we take 2D layer of Graphene and stack it on another 2D layer and twist it as per the process shown in the figure, we get a twisted bilayer Graphene

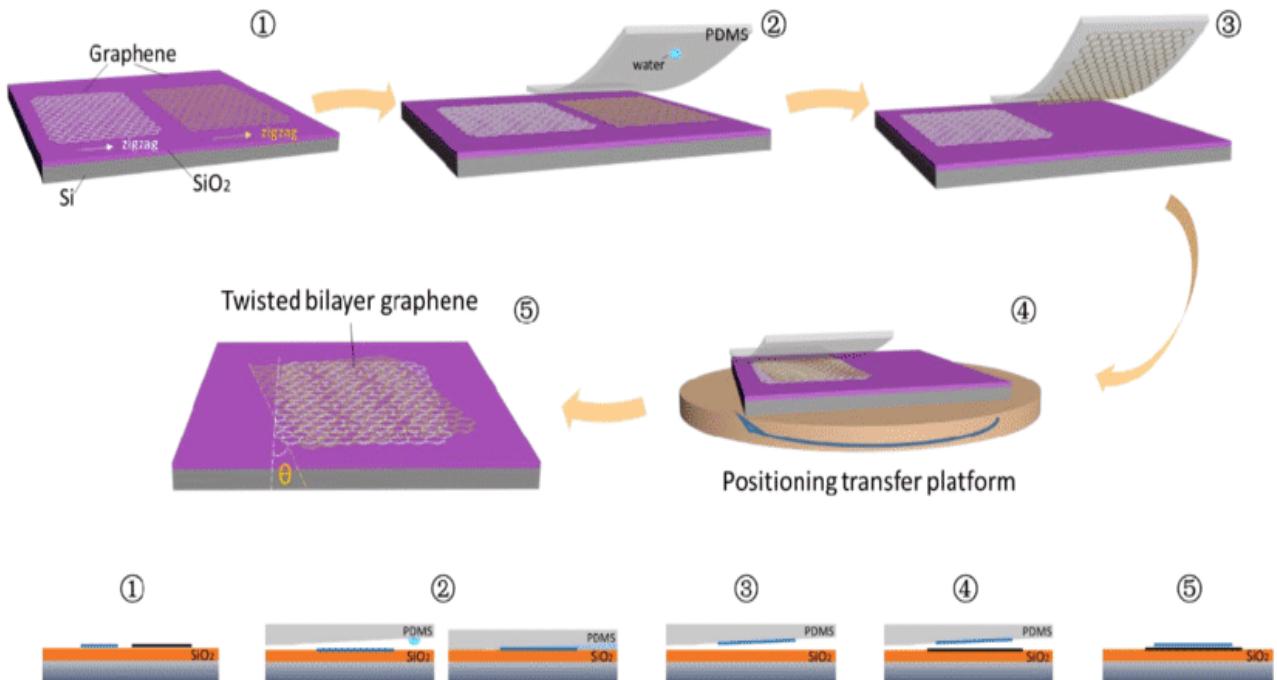


Figure 3: Fabrication Of Twisted Bilayer Graphene

Twisted Bilayer Graphene forms a moire pattern, a large scale visual pattern formed when a pattern with a transparent gap is overlapped on the another similar pattern i.e interference between two or more periodic pattern.

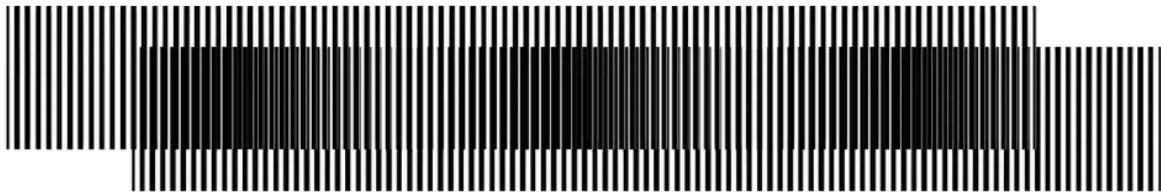


Figure 4: Moire Interference Pattern

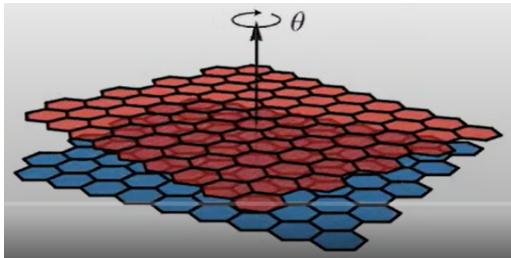


Figure 5: Twisting of Graphene Layer

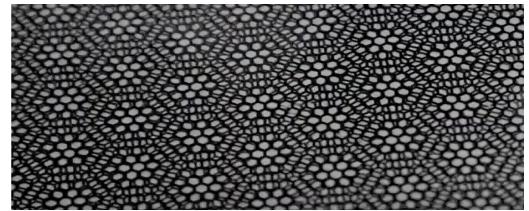


Figure 6: Moire Pattern In Twisted Bilayer Graphene

Now lets deviate from our topic a little to understand the band structure and properties of graphene. We know from special relativity (1905) $E = mc^2$ and therefore a relativistic particle moving with a velocity $V \leq C$ is $E = c\sqrt{p^2 + (mc)^2}$. For $V=C$ and $m=0$, $E=pc$.

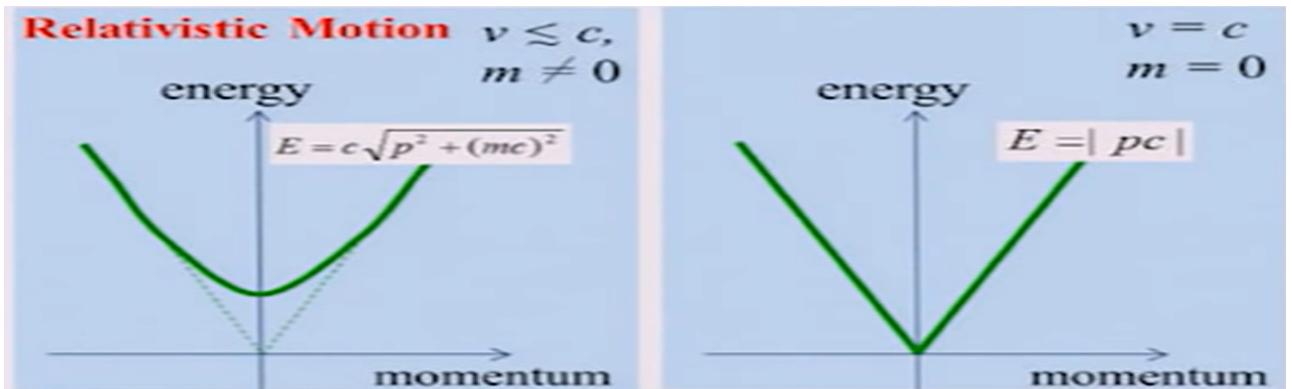


Figure 7: Relativistic Energy vs Momentum Diagram

Now from Schrodinger Equation (1926) we know

$$i\hbar \frac{\partial \psi}{\partial t} = -\frac{\hbar^2}{2m} \nabla^2 \psi + V\psi$$

Paul Dirac combined the Quantum Mechanics given by SE and Relativistic Physics given by Einstein to give Relativistic Quantum Mechanics. Now, Graphene's band structure follows relativistic Quantum Mechanics mathematics despite not having velocity of electrons in graphene equal to speed of light (2-3 percent of speed of light) as the effective mass of electron is considered to be zero in graphene. Hence the term used when Relativistic Quantum Mechanics is applied to charge carriers in Graphene is Quasi Relativistic Quantum Mechanics. Now the dirac equation is

$$(i\gamma^\mu \frac{\partial}{\partial \mu} - m)\psi = 0$$

Dirac's Equation in 2D for massless particles is,

$$H\psi = E\psi$$

$$\hbar v_f \vec{k} \cdot \vec{\sigma} \begin{bmatrix} \alpha_k \\ \beta_k \end{bmatrix} = E(\vec{k}) \begin{bmatrix} \alpha_k \\ \beta_k \end{bmatrix}$$

$E > 0$ are solutions of Conduction Band and $E < 0$ are solutions of Valence Band.

Relativistic Electrons (or Fermions) are chiral i.e electron spin becomes parallel to momentum as shown in the figure 8. Note-The chirality of electron wavefunctions in graphene reduces the back scattering of electrons when incident on a barrier to zero i.e the tunnelling transmission coefficient is 100 % for electrons in Graphene. The solutions of Dirac equations follows a linear dispersion relation with zero band gap, such a cone formed is known as Dirac Cone. Graphene as follows the same maths as relativistic Quantum Mechanics has the

same band structure with no band gap with conduction band and valence band intersecting at dirac point at 6 different point at zero/ground energy level as shown in figure 8.

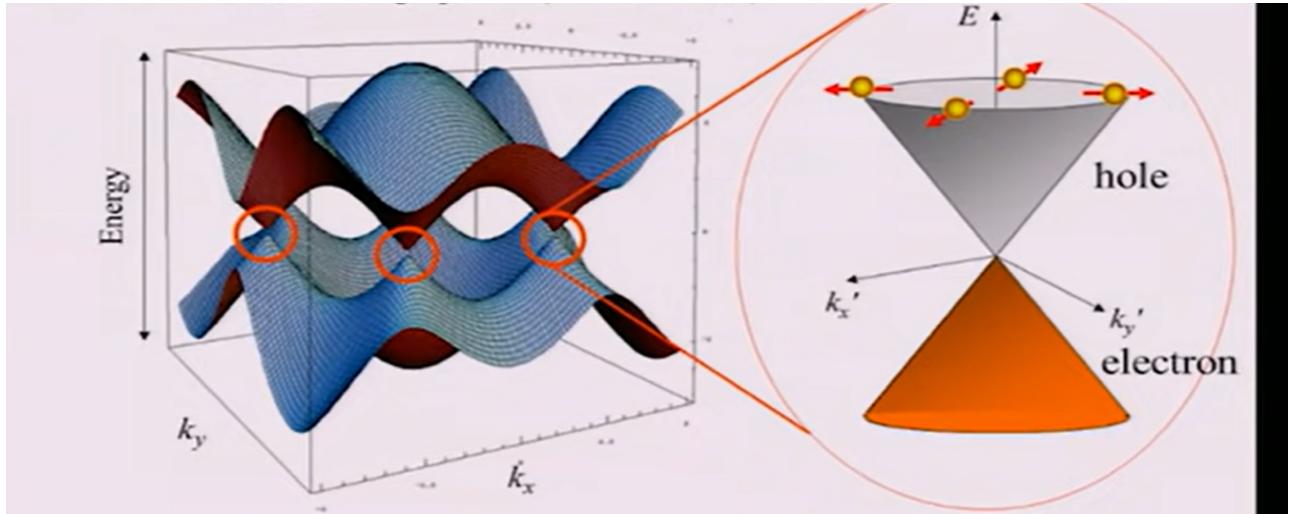


Figure 8: Graphene Band Structure And Dirac Cone

Graphite is a 3D bulk material having parabolic dispersion relation whereas Graphene is a 2D bulk material having linear dispersion relation.

Fermions (electrons) in Graphene has to turn twice in the orbit before it comes back to its own original spin as the orbit the electron follows is like a mobius strip.



Figure 9: Mobiüs strip

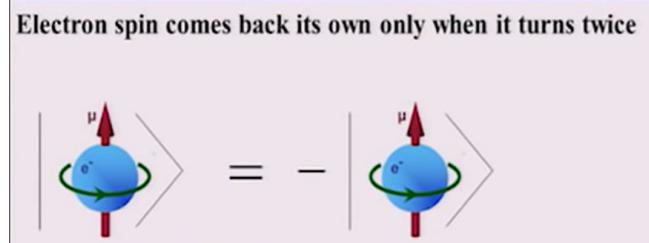


Figure 10: Negative Global Phase Factor After 1 rotation of electron in Mobiüs Strip

There are two spin degeneracies (spin up and spin down) and two valley degeneracies (k and k') in Graphene. The unit cell of Graphene is hexagonal in shape i.e Lattice structure is hexagonal with 3 corners having spin up configuration of electron i.e the sublattice A and remaining 3 corners having spin down configuration of electron i.e the sublattices B. Hence the maximum doping/removal of electrons allowed in Graphene is 4 electrons.

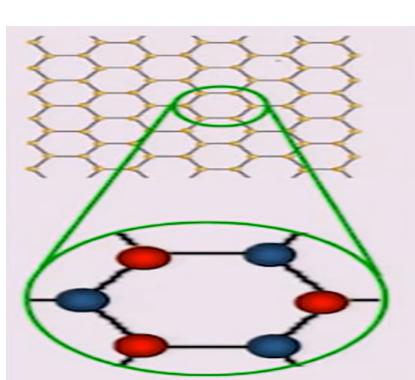


Figure 11: Graphene Hexagonal Lattice

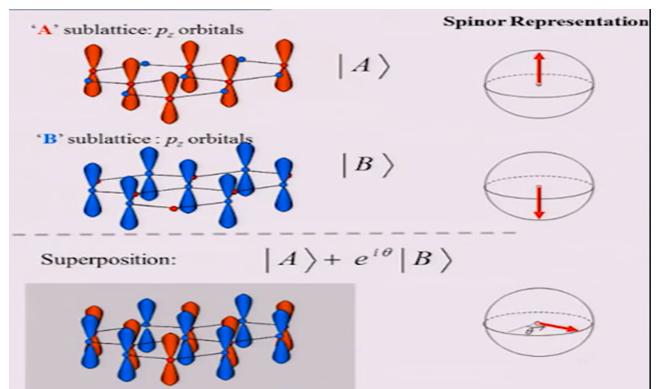


Figure 12: A and B sublattices in Graphene

Now let us consider some important properties of Graphene

1.High Speed Graphene Electrons-

Graphene is a semiconductor that gives the best electron mobility of around $70000 \text{ cm}^2 \text{ per sec}$

2.Optically Almost Transparent-

Graphene is optically almost transparent as it is one atomic layer thick i.e $T \approx 100\%$

3.Mechanical Properties of Graphene-

1.Young's Modulus of Graphene = 1 TPa (very high stiffness as steel has Young's Modulus nearly equal to 0.2 TPa)

2.Lateral Strain = 25%

Graphene has many applications in Semiconductor Industries,Printable Inks,Composites,Heat Discipation,etc with a main one being transparent screen mobile or laptop which is yet to be launched in the market.

Now that basics of Graphene have been discussed lets go back to the discussion on Twisted Bilayer Graphene.

Twisting of two layers of Graphene on top of each other breaks the translation symmetry but results in much more superlattice translation symmetry with its length known as Moire Wavelength or Moire superlattice constant (a_m or L_m)

Let's say the twist angle θ and the lattice constant of Graphene is $a = 0.246 \text{ nm}$ then the Moire Super-lattice Constant or Moire Wavelength is given by a_m or $L_m = \frac{a_0}{\sqrt{2(1-\cos\theta)}} = \frac{a_0}{2\sin(\theta/2)}$

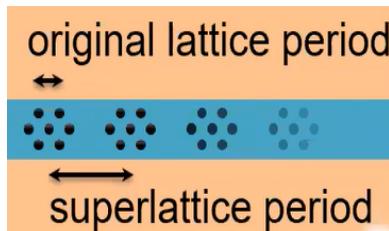


Figure 13: Superlattice Period



Figure 14: Additional lattice period

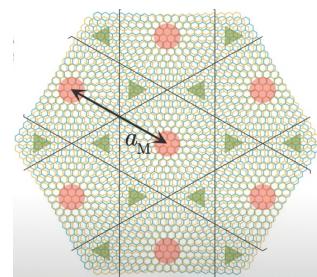


Figure 15: Moire Superlattices

In General for conventional 2D materials $a_0 \approx 0.1$ to 1 nm and therefore the Moire Superlattice Period $a_m \approx 1$ to 100 nm For small angles $a_m = a_0/\theta$ hence the moire wavelength varies as shown in the figures.

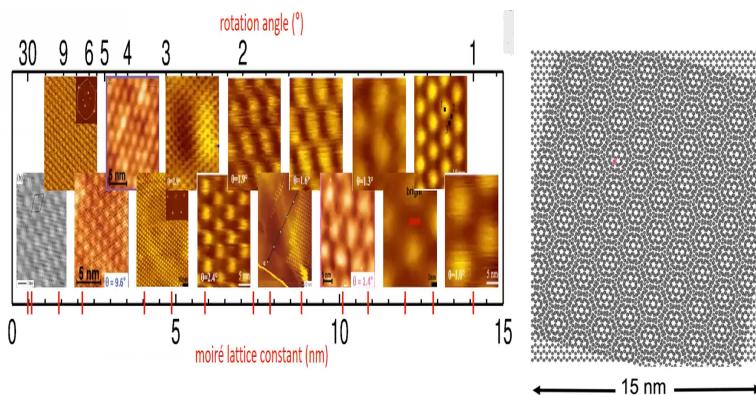


Figure 16: Variation of Crystal Structure of TBG with Twist Angle

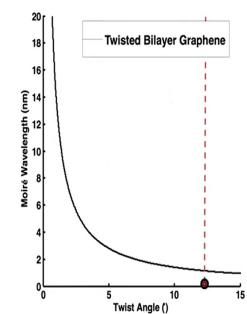


Figure 17: Moire Wavelength vs Twist Angle

When we twist the Graphene layer on top of another Graphene Layer in real space then the dirac cone in the momentum space overlap with each other i.e. as the twisting angle increases the separation between dirac cones in momentum space increases. Now in monolayer Graphene the distance between the two adjacent dirac cones in momentum space is given by $2\pi/a$ and hence in energy scale it is

$E_{mono} = \hbar v_f 2\pi/a$ for a fermi velocity i.e the velocity of electron in graphene at 0k of the order of 10^6 the Energy required for electron to move through the graphene lattice from one dirac cone to other is $\approx 16 \text{ eV}$.

In twisted bilayer the two dirac cones gets overlapped with each other hence the distance between the cone centres in momentum space is $\Delta k = 2k \sin(\theta/2) \approx k\theta$ for small θ and the distance between the two adjacent dirac point of two Graphene layers in momentum space is $2\pi\theta/a$ and hence the dirac energy scale i.e the intralayer tunnelling energy required for electron to go from one dirac cone of one layer to adjacent dirac cone of other layer is,

$$E_{twistedBLG} = \hbar v_f 2\pi\theta/a \approx 16\theta \text{eV}.$$

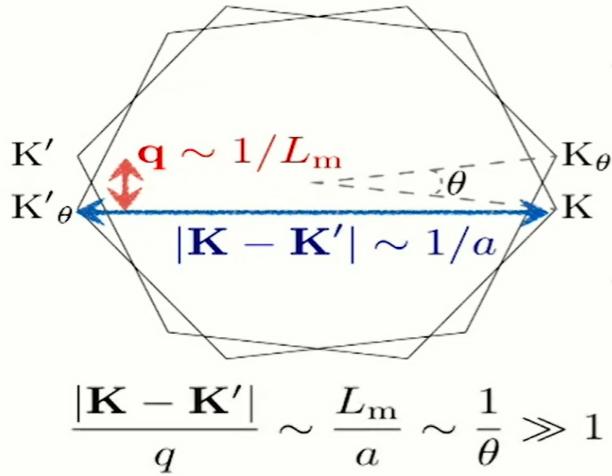


Figure 18: Momentum Space Hexagon in Brillouin Zone of TBLG

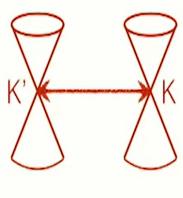


Figure 19: Monolayer Graphene Distance

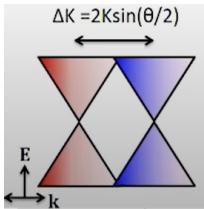


Figure 20: Overlapping in between dirac cones due to twisting

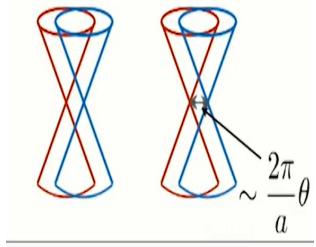


Figure 21: Twisted Bilayer Graphene Distance between the Dirac Cones

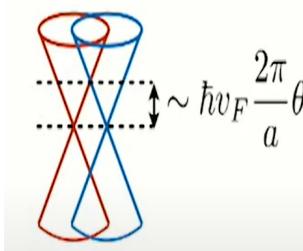


Figure 22: Dirac Energy Scale

Now the coupling strength between the top layers of dirac cone of the conduction band and the bottom layers of the dirac cone of the valence band in T.B.L.G = 0.3 eV i.e energy required for electrons to tunnel in between the two graphene layers Initially at large twist angles the energy required for interlayer tunnelling of electrons is very small compared to energy required for intralayer movement of electrons i.e $2w \ll 16\theta$.

Now, if we decrease the twist angle to smaller values there comes an angle when the interlayer tunnelling energy is equal to the intralayer tunnelling energy required for electrons. At such an angle of twist the band at dirac point of TBLG becomes completely flat i.e a type of band structure with constant energy independent of the momentum, such an angle is called magic angle.

$$2w \approx \hbar v_f 2\pi\theta/a$$

$$0.3 \text{eV} = 16\theta$$

$$\theta = 1.07^\circ \dots \text{Magic Angle i.e band structure at dirac point becomes completely flat.}$$

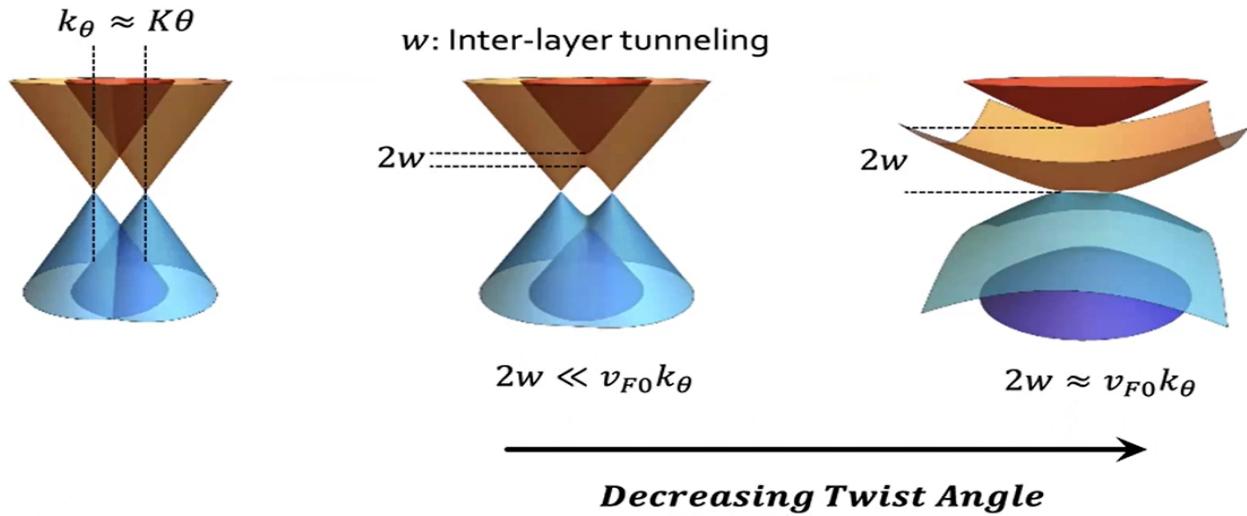


Figure 23: Magic Angle and Flattening of Bands at Dirac Points

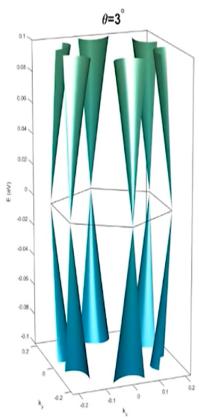


Figure 24: Simulation of Angle of Twist other than magic angle

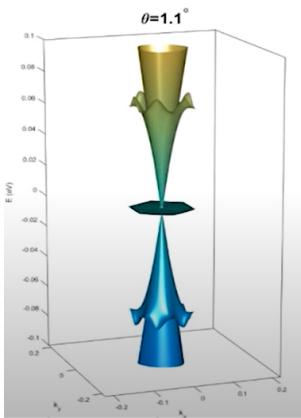


Figure 25: Simulation of Perfect Flat Band at Magic Angle

For a band to be characterized as flat ,charge carriers in it should have zero group velocity i.e $dE/dk=V_g = 0$.

Flat bands are considered ideal to study many body interaction phenomenon in physics such as superconductivity,orbital magnetism,etc as carriers in the flat bands are dominated by electron electron interaction when compared to other platforms of cold atomic optical lattices and quantum materials i.e. material chemistry.

Electron-Electron Interaction for Flat band is given by

$$U = e^2 \theta / (4\pi \epsilon k a_m) \approx 15 \text{ meV}$$

Bandwidth(W)/Kinetic Energy(t) of Hopping of Electrons of the flat band (pratically not zero)is given by
 $W = (\hbar^2 k^2) / 2m^* = (\hbar^2 \pi^2) / (2m^*(a_m)^2) \approx 10 \text{ meV}$

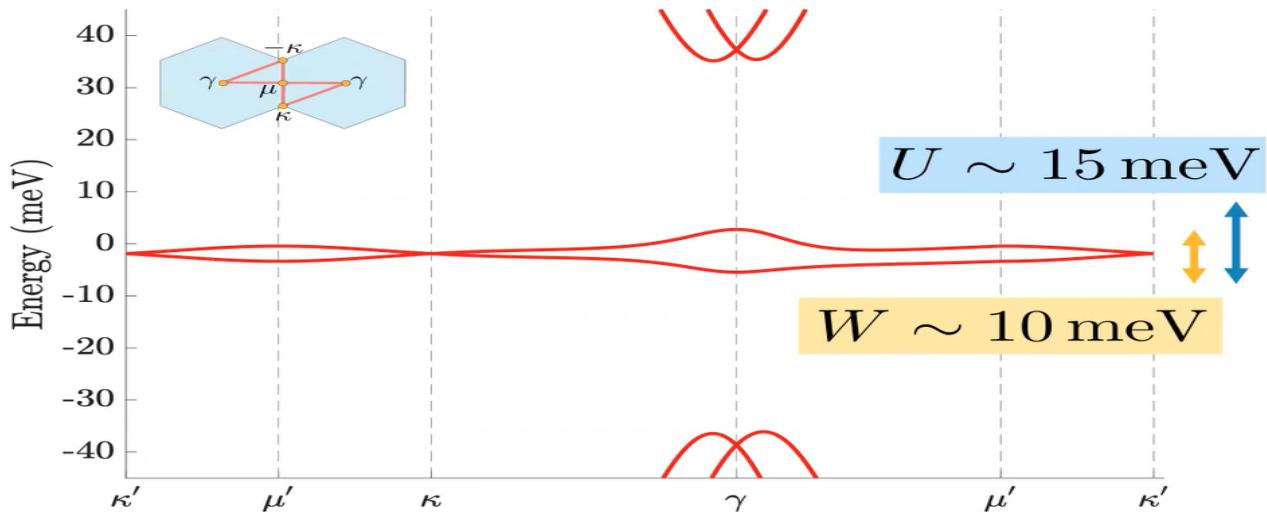


Figure 26: Strong Correlation Exists for Flat Bands

$U/W = 1.5 > 1$ hence we get a new platform i.e Flat Band to model the many body Interaction Physics.

As already discussed Graphene has two sublattices A and B having spin up and spin down state of electron respectively. Now when we stack and twist two layers of Graphene atop each other we get A-A stacking, A-B Stacking and B-A stacking as shown in the figure. If we join the four neighbouring A-A lattices we get Moire Unit Cell as shown in the figure. As Graphene has two spin and valley degeneracies in total Moire Unit cell can occupy max of 4 electrons or 4 holes. Since there are two bands in Twisted Bilayer Graphene we get a total of 8 degrees of freedom per moire unit cell.

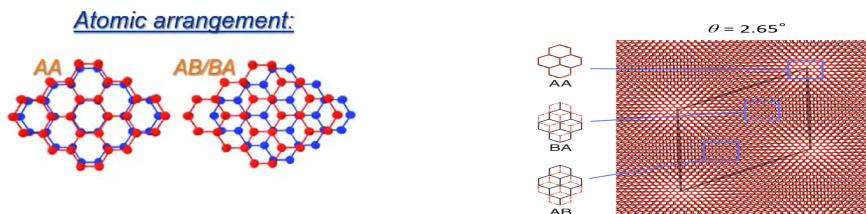


Figure 27: AA and AB/BA Stacking In Twisted Bilayer Graphene

Figure 28: Moire Unit Cell

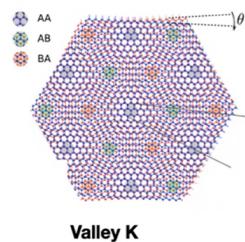


Figure 29: AA and AB/BA stacking in Hexagonal Lattice of TBLG

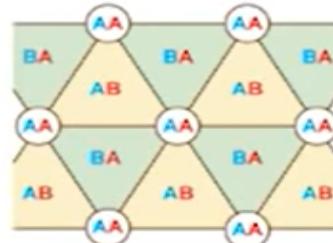


Figure 30: Stacking + Moire Unit Cell

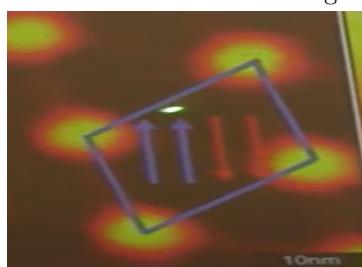


Figure 31: Max. Electron Filling Of Moire Unit Cell(STM image)

Now let's discuss Moire Brillouin Zone. As we already know from above discussion the moire superlattice

period is much greater than lattice period in real and hence the moire brillouin zone is much smaller than Brillouin zone of Monolayer Graphene in momentum space. Moire Brillouin zone for twisted bilayer graphene is nothing but the mismatch of two brillouin zone of Monolayer Graphenes as shown in the figures.

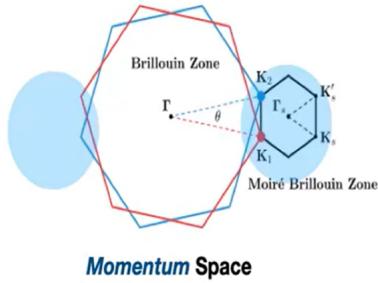


Figure 32: Moire Brillouin Zone

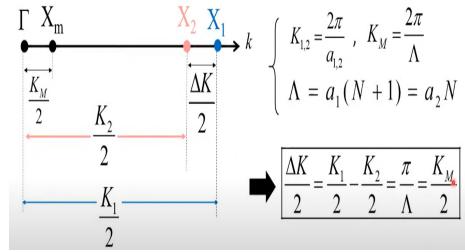


Figure 33: Moire Brillouin zone of TBLG as Mismatch Between two B.Z of MLG

Flat Band Structure In Twisted Bilayer Graphene and its comparison with the monolayer graphene is shown in the below figures.

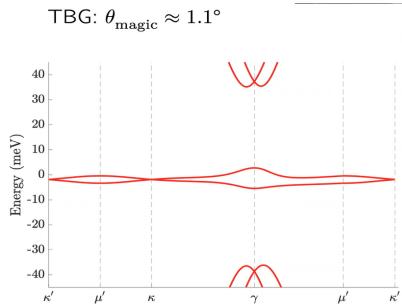


Figure 34: Band Structure of Twisted Bi-layer graphene at magic angle

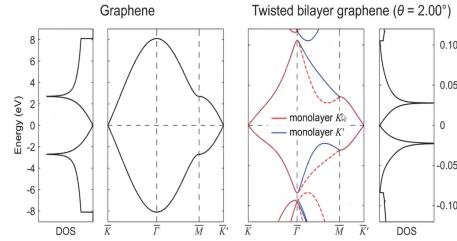


Figure 35: Band Structure Of Monolayer And Twisted Bilayer Graphene

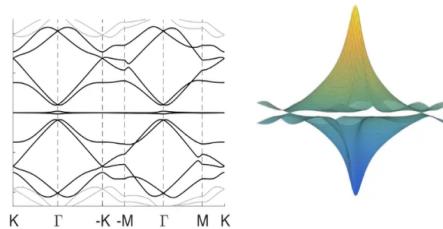


Figure 36: Flat Band Structure in 2D and corresponding 3D diagram in momentum space

3 Quantum Phenomenons Observed in Moire Superlattices

Unfortunately due to short time frame of submission I will not be able to discuss in detail the Quantum Phenomenons Experimentally Observed In Twisted Bilayer Graphene but I will just mention their names for the sake of Completion of Concept.

- 1.Superconductivity
 - 2.Corelated/Mott/Chern Insulator
 - 3.Topological System
 - 4.Orbital Magnetism
 - 5.Anamolous Quantum Hall Effect
 - 6.Creating Flat Band and Flat Band Physics (Many Body Interactions)
 - 7.Nematicity
- and more and more coming on the way owing to future research.

4 Outlook and Future Applications in Quantum Technologies

1.Superconductivity at Room Temperature is a dream physicists have been dreaming since 19th century.Moire Superlattices provides a new platform for theoritically as well as experimentally studying the many body interactions physics with superconductivity at room temperature being the crown prize.Superconducting Circuits are used for creation and measurement of transmon qubits but the problem of cooling Helium to 4.1 K by Dilution Refrigerator still remains,so Moire Superlattices or Van derwaal materials which have theoritically property wise infinite possibilities can be studied and researched extensively to provide alternatives in Quantum Circuits required for quantum Computing.

2.Quantum Physics though practically realized is still not understood/solved completely.Doing Extensive Experiments on such Vanderwaal materials can provide us new insights on how the quantum mechanics really ticks!! And we can hopefully solve all the models in quantum physics both theoretically and practically .For example Ashwin Vishwanath in his talk raises the question of what is fundamental an electron or a ferromagnet at least in topological sense.We know conventionally that Ferromagnet is struture formed when the spin of electrons arrange in some same direction.Now if we start with a ferromagnet and make a skrymions arrangement then this arrangment essentially has the same properties of an electron.So is electron a derived concept? This may sound bizzare but so is quantum physics.

3.Study of New Kind of Magnetism i.e Orbital Magnetism which is an extremely rare case can be done to gain new insights.

4.Conventional Quantum Materials i.e Material Chemistry changing Approach to achieve a transition from superconducting to insulating phase we have to essentially change the material each time we want the material to achieve a certain point in the phase diagram but in twisted bilayer graphene we can tune the doping only to change from a insulting phase to a superconducting phase in the same material itself just by varying the gate voltage which can be leterally done by rotating a knob!!

5.As tunability of Moire Superlattices is very easy we can create strong and tunable quantum interactions which can be used in quantum gates/circuits to achieve particular exact locations of quantum states.

6.In Twisted Bilayer Graphene we can flip magnetism with a small DC current which suggests that we can create a quantum memory out of this concept.But why do we require a quantum memory that works only at few kelvins? As from this system we require a current density that is 10^4 times lower than the conventional used spin orbital torque memory i.e we can get a power efficient quantum memory which can act as an control and read out interface for quantum computer

References

Youtube Talks-

- 1.Abbay Pasupathy - “Twisty fun in 2D materials”
- 2.Alan MAcdonald’s Electrical and Optical Properties of MATBG
- 3.Alex Thomson’s (CalTech) Magic-angle multilayer moiré materials
- 4.Ashvin Vishwanath (Harvard) Topology on graphene moiré materials (3rd talk)
- 5.Ashvin Vishwanath (Harvard), Quantum Magic in Moiré Lattices
- 6.Band Free Approach To Twistorronics by Stepann Carr
- 7.Bilayer Graphene - Superconductors, Orbital Magnets, Correlated States and beyond by Dmitri Efetov
- 8.Competing Orders, Nematicity and Novel Josephson Effects in Superconducting Graphene by Yuan Cao
- 9.Daniel Leykam’s Flat bands, sharp physics
- 10.David Goldhaber Gordan’s Ordered and maybe topological states in flat band 2D moire systems
- 11.Electrons in Moiré Superlattices A playground for correlation and topology by Ali Yazdani
- 12.Eslam Khalaf’s Flat bands, correlated insulators, and superconductivity in Moire graphene
- 13.Feng Wang’s Correlated Topological Phenomena in Trilayer GrapheneBN Moiré Superlattices
- 14.Graphene Superlattices -Electrostatic Superlattices and Moiré Superlattices
- 15.Hai Son Nguyen’s Magic configurations in Moiré Superlattice of Bilayer Photonic crystal
- 16.Jie Shan’s Electrons in 2D semiconductor moiré superlattices
- 17.Jun-Won Rhim’s Novel geometric aspects in flat band systems
- 18.Magic-Angle Graphene Superlattices by Pablo Jarillo-Herrero(Discoverer of Superconductivity in Graphene)
- 19.New Physics In Flat bands by Erez Berg
- 20.Orbital Magnetism in Graphene Hetrostructures by Andrea Young
- 21.Pablo Jarillo Herrero, MIT - “The magic of moire quantum matter”
- 22.Philip Kim’s Relativity quantum Physics And Graphene
- 23.The Facinating Quantum World of Two-dimensional Materials by Steven G. Louie
- 24.Zoe Zhu’s Modelling the structural and electronic properties of Moire