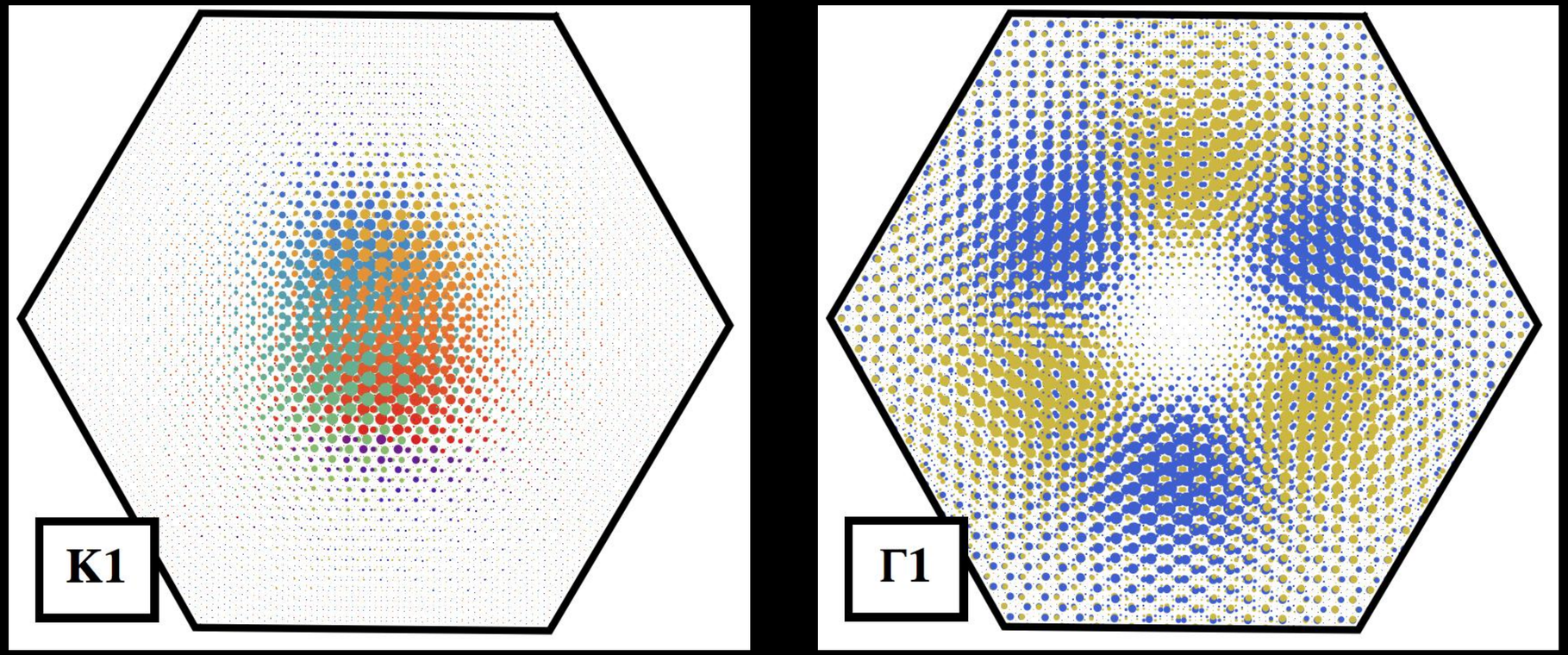
Charge-transfer insulation in Twisted Bilayer Graphene



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Charge-transfer insulation in twisted bilayer graphene

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Charge Smoothening and Band Flattening due to Hartree corrections in Twisted Bilayer Graphene

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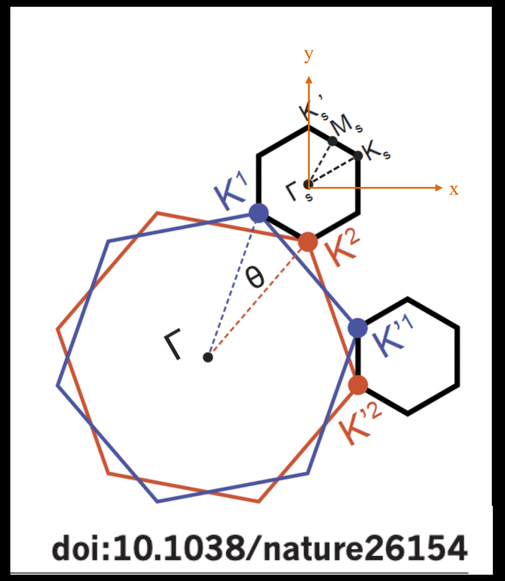
A few facts in Twisted bilayer Graphene (TBG):

- **Flat bands** when θ of TBG is close to "magic".
- **Correlated insulating phase** at half-filling **n = 2.**
- **Superconductivity** when doping the insulating state.
- At **n = 0**, the electronic density of states is **peaked around the "AA"** staking of the Moire super cell.



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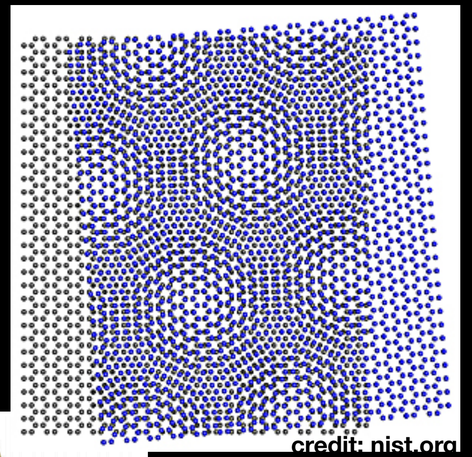

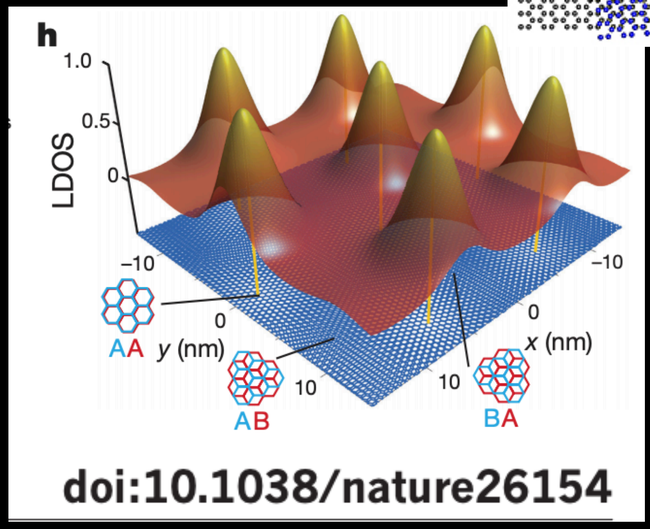

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The mechanism of metal-insulator transition,

The nature of the correlated insulating state,

The superconducting mechanism,

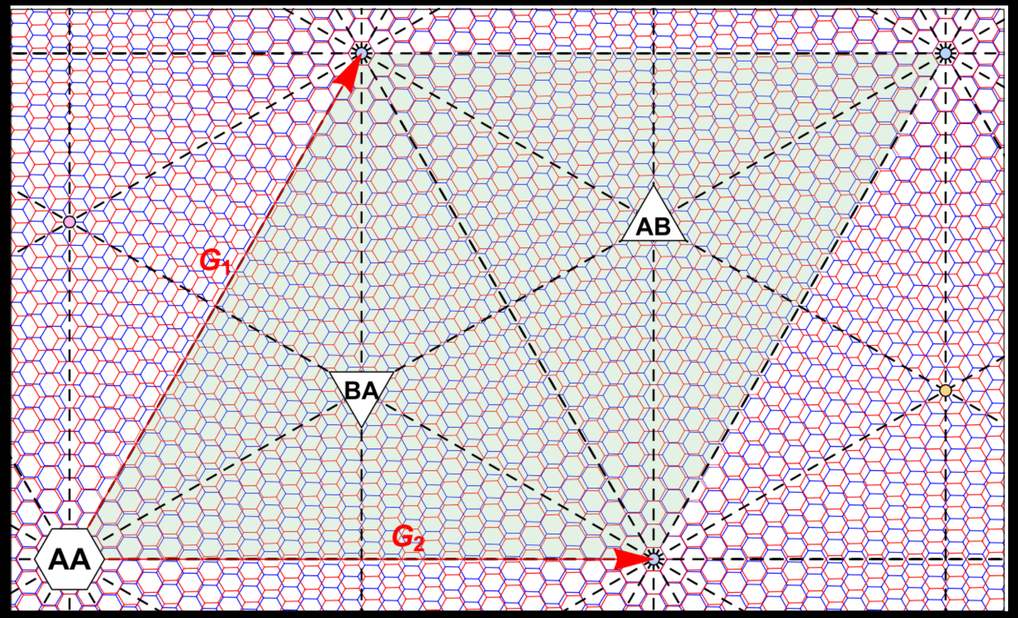
Quantum anomalous Hall effect,

Ferromagnetism

Our approach: To study the real-space structure of states of a full tight binding hamiltonian of TBG at the magic twist angle.

Start with AB stacking and rotate one of the layers around an AB site

Magic angle θ = 1.08, 11 164 atoms in the Moire unit cell.



- Six-fold rotation around AA, 2 three-fold rotations around AB and BA, C2 with axis in the plane
- No mirror symmetries

C_6 (6)			E	C_6	C_3	C_2	C_3^2	C_6^5
x^2+y^2, z^2	R_z, z	A	1	1	1	1	1	1
		B	1	−1	1	−1	1	−1
(xz, yz)	(x, y)	E'	1	ω	ω^2	ω^3	ω^4	ω^5
	(R_x, R_y)		1	ω^5	ω^4	ω^3	ω^2	ω
(x^2-y^2, xy)		E''	1	ω^2	ω^4	1	ω^2	ω^4
			1	ω^4	ω^2	1	ω^4	ω^2

$\omega = e^{2\pi i/6}$

At **K**

K and K' exchanged under six and twofold rotations (z).

The little co-group at the K point is **C_3**.

$C_3(3)$			E	C_3	C_3^2
x^2+y^2, z^2	R_z, z	A	1	1	1
(xz, yz)	(x, y)	E	1	ω	ω^2
(x^2-y^2, xy)	(R_x, R_y)		1	ω^2	ω

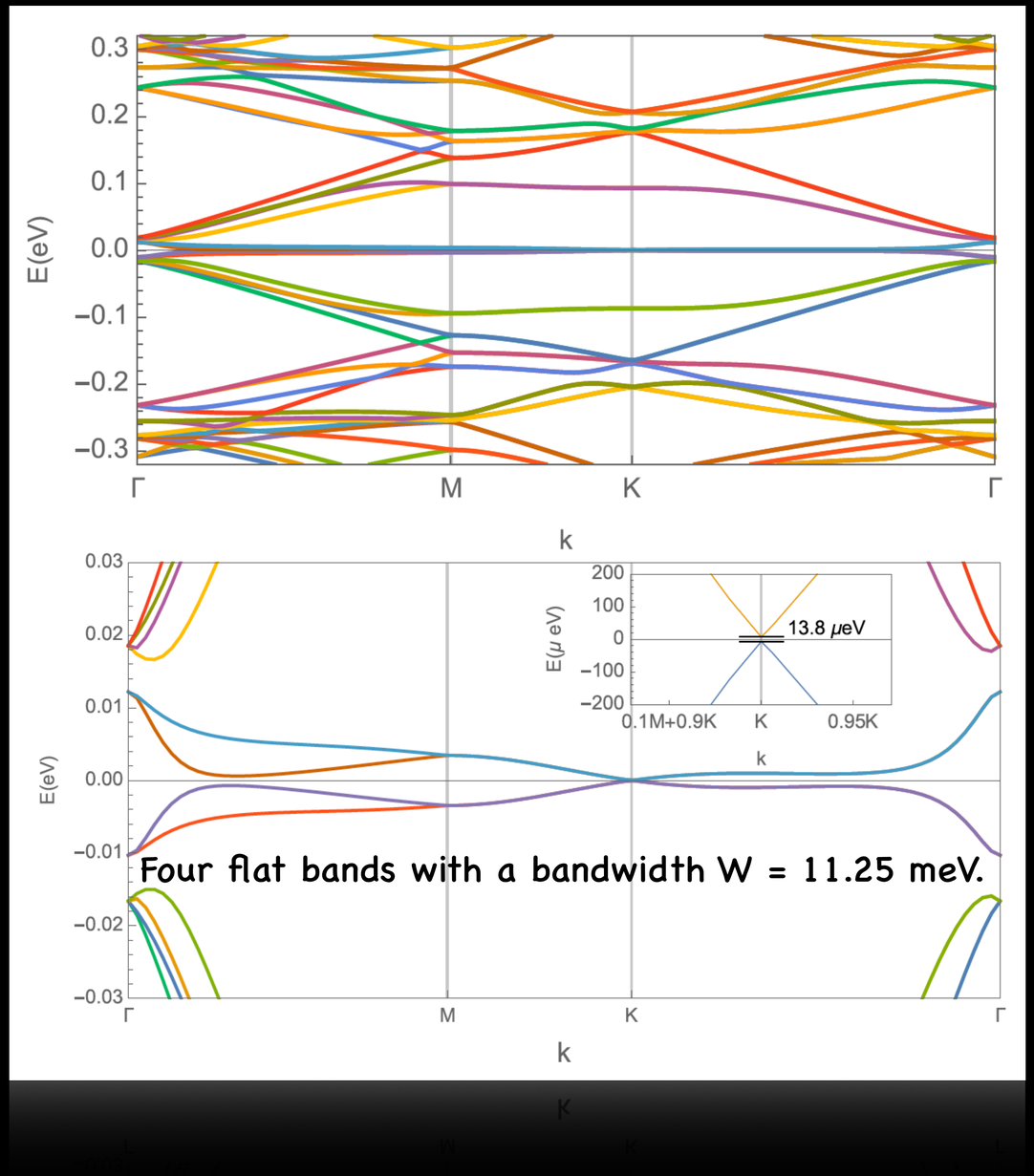
$\omega = e^{2\pi i/3}$

- Orbitals at the K point should be two-fold degenerate.
- Gap neglishable and the K orbitals effective hopping in a honeycomb lattice.

At Γ

- The little group at the Gamma point is isomorphic to **C_6**.
- At the Gamma point the triangular lattice is invariant under 3-fold rotations.
- Orbitals can realize irreps A and B.
- Hopping on a triangular lattice

(Full band) Tight binding model



In-plane n-n hopping t = 2.8 eV.

Interlayer hopping $t_\perp(r) = t_{\perp 0}\, e^{-|r|/\xi}$

r = distance between two atoms including the interlayer distance

d = 0.335 nm,

$\xi = 0.11a$,

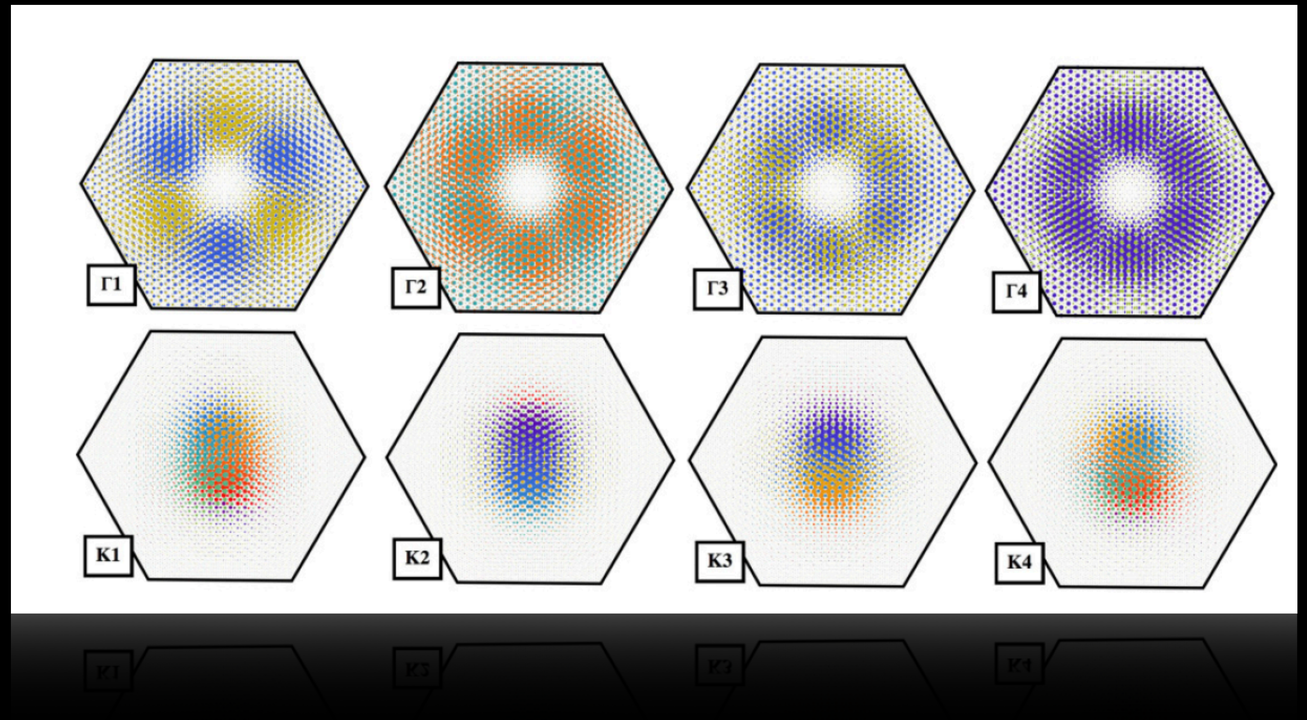
a = 0.246 nm

$t_{\perp 0}$ chosen such that $t_\perp$ = 0.35 eV for the **AA** stacked atoms.

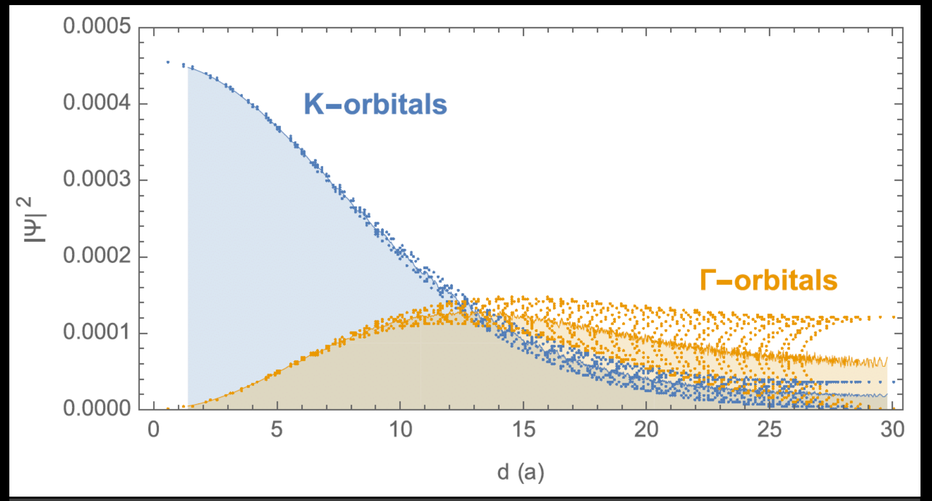
Small gap of 13.8 μeV at K : "approximate" Dirac cones, two flat bands above and two flat bands below them.

Flat bands change orbital character in momentum space

Spatial structure of the low-energy wave functions.



Averaged wave function squared as a function of distance from the AA centers



We propose that the insulating phase is characterized by a charge transfer from the center to the ring orbitals due to long-range Coulomb repulsion.

Orbital nature of the flat band smoothly varies from ring to center orbitals as a function of momentum.

Qualitative difference between the real-space wave function: need more than four localized Wannier orbitals to capture the flat bands.

Including Lattice Relaxation

We use the relaxed atomic positions from F. Gargiulo and O. V. Yazyev, *2D Materials 5, 015019*

Tight-binding with hopping

$$-t(\vec{d}) = V_\pi^0 \left[1 - \left(\frac{\vec{d}\cdot\vec{e}_z}{d}\right)^2\right] e^{-\frac{d-a_0}{r_0}} + V_\sigma^0 \left(\frac{\vec{d}\cdot\vec{e}_z}{d}\right)^2 e^{-\frac{d-d_0}{r_0}}$$

the length of vector connecting two carbon atoms

0.142 nm = intralayer nearest neighbor atomic distance

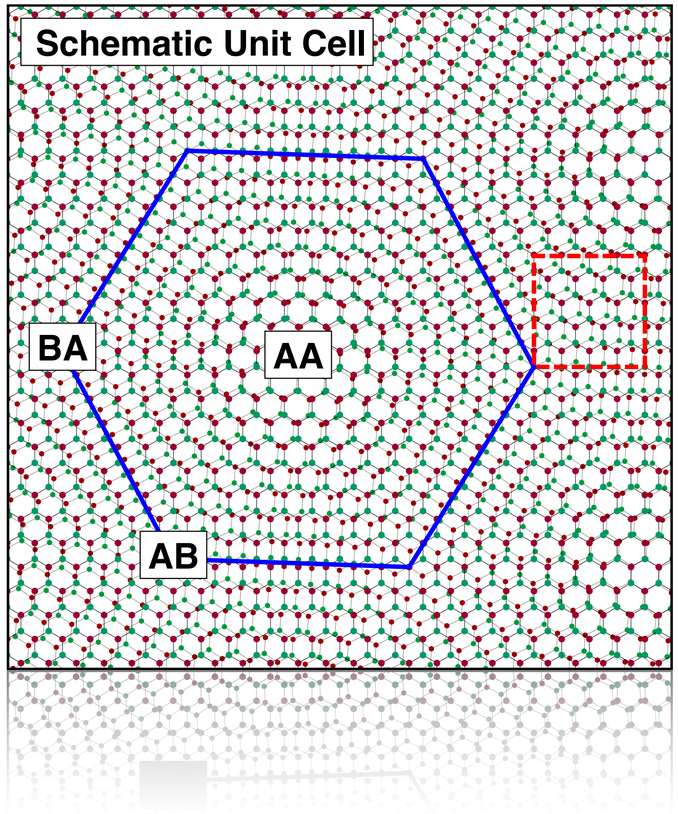
0.335 nm = the unrelaxed interlayer distance

2.7 eV= nearest neighbor intralayer hopping strength

0.48 eV = the inter- layer hopping when two carbon atoms are exactly above each other

p-orbital decay length

Charge distribution at charge neutrality is inhomogeneous



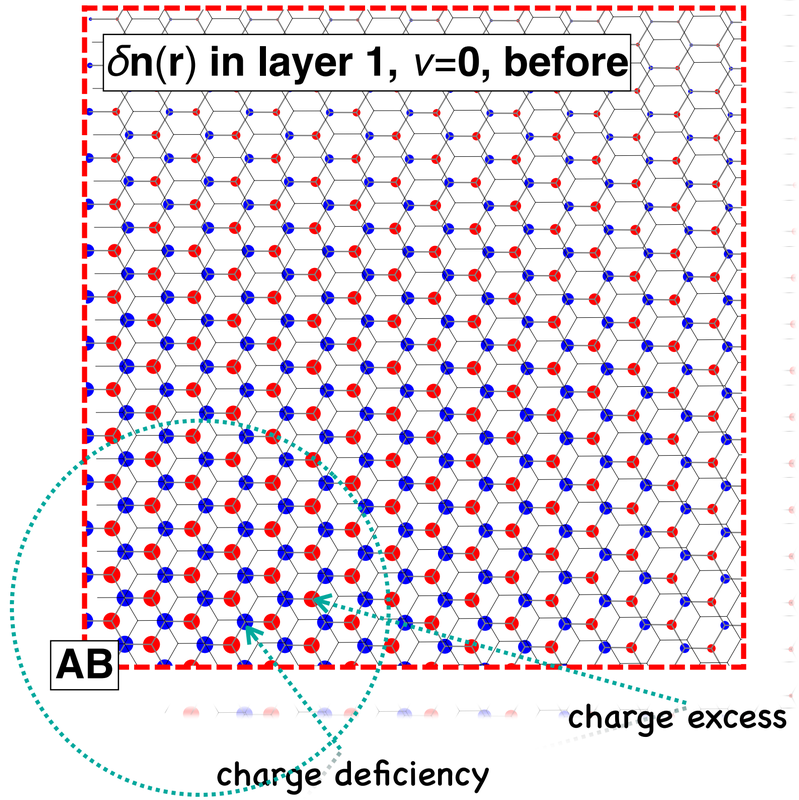
ν : number of electrons per Moire unit cell relative to charge neutrality. +/-4 corresponds to either completely empty or completely filled flat bands

$\bar{n}$: average density per atom = $1 + \frac{\nu}{N}$

N : # atoms in the unit cell.

$$\delta n(\mathbf{r}) \equiv n(\mathbf{r}) - \bar{\mathbf{n}}$$

The size of the circles indicate the magnitude of charge deficiency with a maximum of 0.0025 at the AB point.



Charge inhomogeneity in AB/BA regions is present even at charge neutrality. The distribution of charge at nu = 0, reveals a clear charge deficiency on the atoms that lie on top of an atom from the other layer.

The electrons are subject to Coulomb interactions, which depend on the deviation of the electron density n(r) from the average density.

$$H_V = \frac{1}{2} \sum_{ij} \delta n(\mathbf{r}_i) V(\mathbf{r}_i - \mathbf{r}_j) \delta n(\mathbf{r}_j)$$

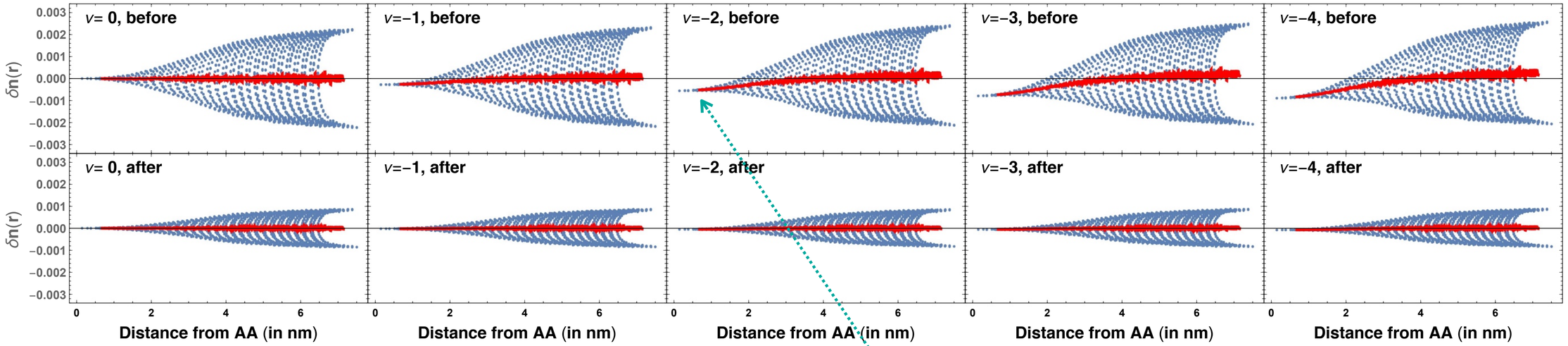
Interpolating between single-layer graphene on-site repulsion V(0) = 9.3 eV and unscreened $\frac{e^2}{|\mathbf{r}|}$ at large distances

$$V(r_i - r_j) = \frac{1.438}{0.116 + |r_i - r_j|} \text{ eV}$$

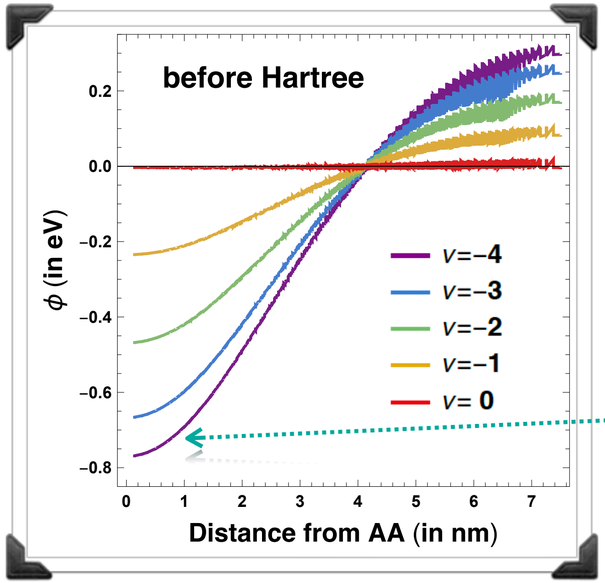
distance between two carbon atoms

Doping TBG away from charge neutrality leads to an enormous buildup of charge inhomogeneities within each Moire unit cell.

Relative charge density between AA and AB sites



Moving average, which shows the charge inhomogeneity between the **AA** and the **AB/BA** regions of the unit cell: **the electronic charge density cluster around AA.**



Charge inhomogeneity leads to a very strong (negative) electric potential at AA sites.

Charge Smoothening due to Hartree corrections in Relaxed TBG

Hartree: the product of two density operators is replaced by the product of an operator and its expectation value.

Interaction replaced by a site-dependent electric potential determined self consistently.

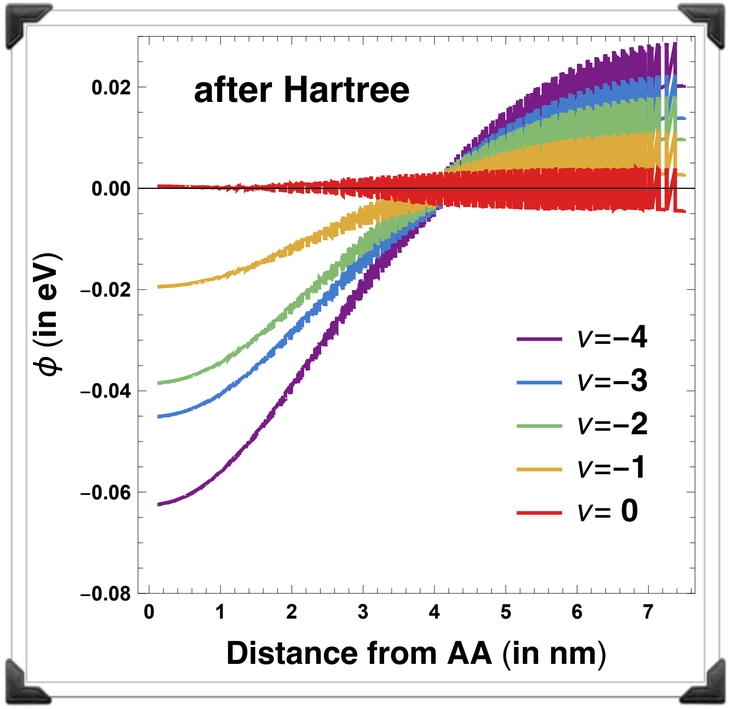
$$H_V = \frac{1}{2}\sum_{ij} \delta n(\mathbf{r}_i) V(\mathbf{r}_i - \mathbf{r}_j) \delta n(\mathbf{r}_j)$$

$$H_H = \sum_i \delta n(\mathbf{r}_i)\phi_i,$$

$$\phi_i = \sum_j V(\mathbf{r}_i - \mathbf{r}_j)\langle \delta n(\mathbf{r}_j)\rangle$$

ν	$\delta n_{\rm AA}$		$\Delta\phi$ (meV)		Reduction
	before	after	before	after	factor
0	-0.026962	-0.0049907	28.8934	8.60985	3 - 5
-1	-0.318072	-0.0285706	339.343	30.5505	11
-2	-0.61295	-0.0515452	660.412	56.6246	12
-3	-0.87059	-0.0641868	935.592	67.5796	14
-4	-1.03138	-0.0850411	1088.83	91.2731	12

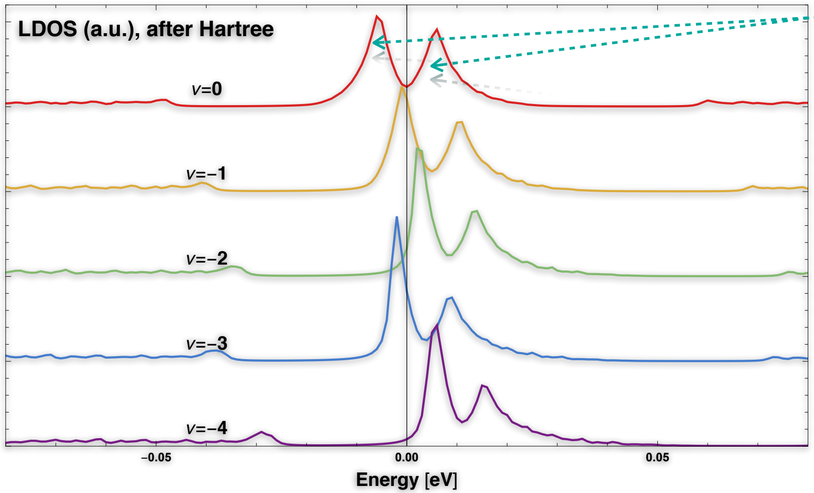
TABLE I: The total accumulated charge in the AA region, $\delta n_{\rm AA}$, and the maximum electric potential difference $\Delta\phi$, as a function of the filling ν. We compare the results before, and after the self-consistent Hartree calculation. Both the electric potential and the charge inhomogeneity are severely reduced by the Hartree potential. The reduction factor (before divided by after) is shown in the last column.



- Electric potential is negative around the AA regions. The potential tends to pull more electrons there.
- Hartree reduce both the charge inhomogeneities and the electric potential one order of magnitude at fillings other than charge neutrality.
- This can be viewed as a charge transfer between AA and AB/BA regions.

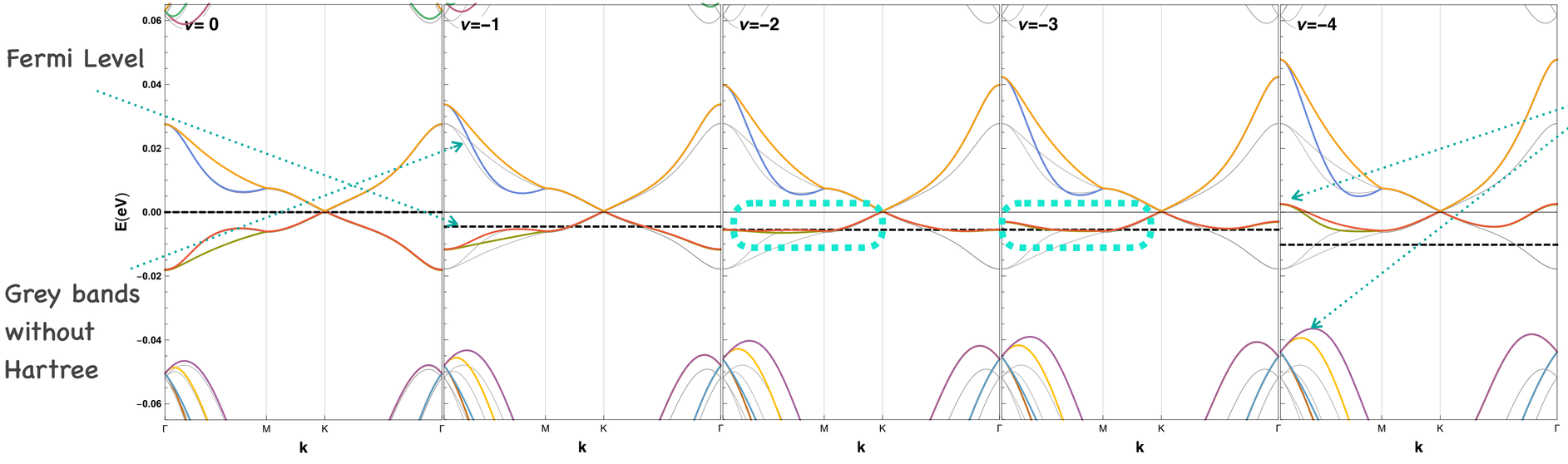
The charge redistribution affects dispersion of the bands, as well as the local density of states (LDOS)

LDOS at the **AA** region of the unit cell



- For hole doping, pinning of the Van Hove singularity (VHS) at the Fermi level consistent with continuum models.
- The change of position of the VHS is a direct consequence of band structure changes.
- Coulomb interactions smoothen the charge imbalance by changing the occupation of `ring' orbitals in the **AB/BA** region and `center' orbitals at the **AA** region.

Band structure after Hartree corrections



- Upon doping away from charge neutrality, the states at Gamma are pushed upwards, a feature of the charge smoothening.
- The increase of the energy of the states at the Gamma point leads to a further flattening of the flat bands.
- For the flat bands, this implies a reduction of the bandwidth.

Synthesis

- Coulomb interactions, as captured by a fully self-consistent Hartree calculation, smoothen the charge inhomogeneities in the unit cell.

- This charge smoothening is achieved by pushing up the energy of the states at the Gamma point, which in turn forces the VHS to remain close the Fermi level.

- Given the strong band flattening and charge smoothening that Hartree gives, any effective low-energy model should start from a Hartree-renormalized band structure.