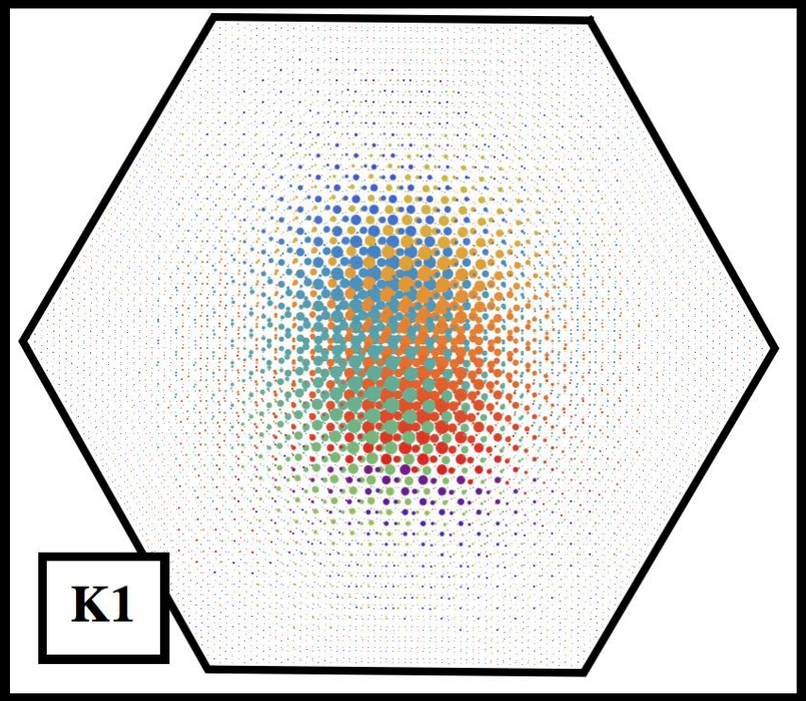
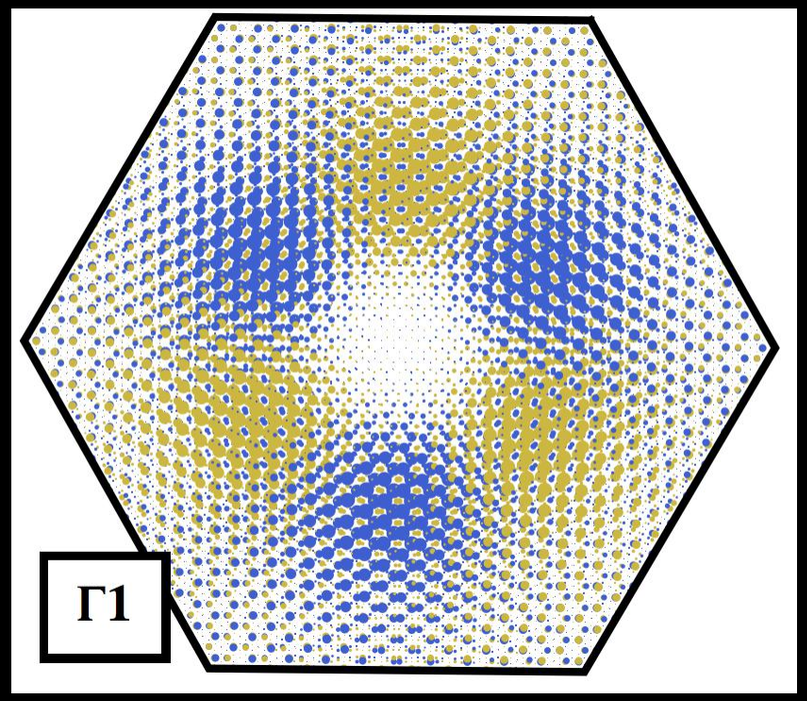
Charge-transfer insulation in twisted bilayer graphene





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KITP, January 2019

In collaboration with

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PHYSICAL REVIEW B **98**, 235158 (2018)

Charge-transfer insulation in twisted bilayer graphene

Louk Rademaker[1,2] and Paula Mellado[2,3]

We study

The real-space structure of states two layers of graphene stacked with a twist angle $\theta = 1.08^\circ$ forming a periodic moiré pattern.

Experiments:

- Flat bands induced by interlayer hybridization when the twist angle of TBG is close to the magic angle.

- Quenching of the quantum kinetic energy leads to a correlated insulating phase at half-filling of these flat bands.

- Electronic phases in TBG: Mott-like insulator behavior in the case of hole-doped bilayer graphene with filling factor n = 2, corresponding to two particles per unit cell of the moiré pattern and Superconductivity when doping the insulating state.

- At charge neutrality n=0, the electronic density of states is peaked around the "AA" staking of the large unit cell.

Plus: The real space structure of the flat bands qualitatively changes when doping away from charge neutrality: Differential conductance maps.

PHYSICAL REVIEW B **96**, 115434 (2017)

Scanning tunneling microscopy and spectroscopy of finite-size twisted bilayer graphene

Wen-Xiao Wang,[1] Hua Jiang,[2] Yu Zhang,[1] Si-Yu Li,[1] Haiwen Liu,[1] Xinqi Li,[3,4] Xiaosong Wu,[3,4] and Lin He[1,*]
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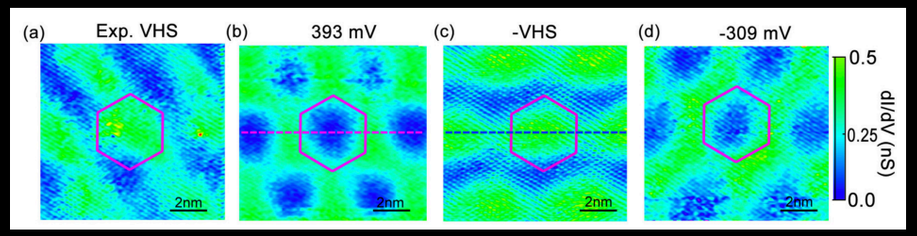

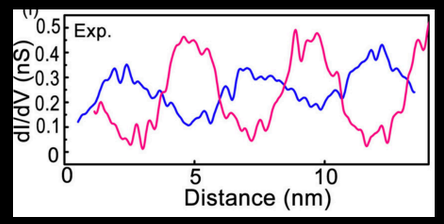

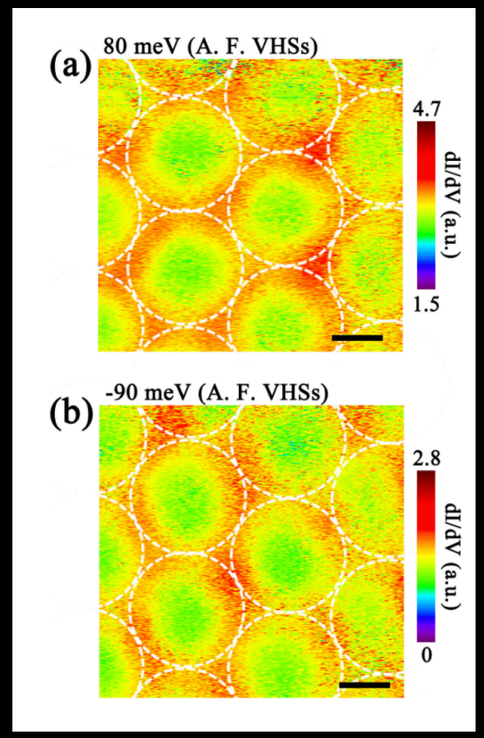

Splitting of Van Hove singularities in slightly twisted bilayer graphene

Si-Yu Li,[1,2] Ke-Qin Liu,[1,2] Long-Jing Yin,[1,2] Wen-Xiao Wang,[1,2] Wei Yan,[1,2] Xu-Qin Yang,[1,2] Jun-Kai Yang,[1,2] Haiwen Liu,[1,2] Hua Jiang,[3,*] and Lin He[1,2,†]
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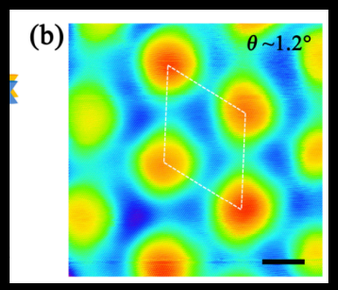
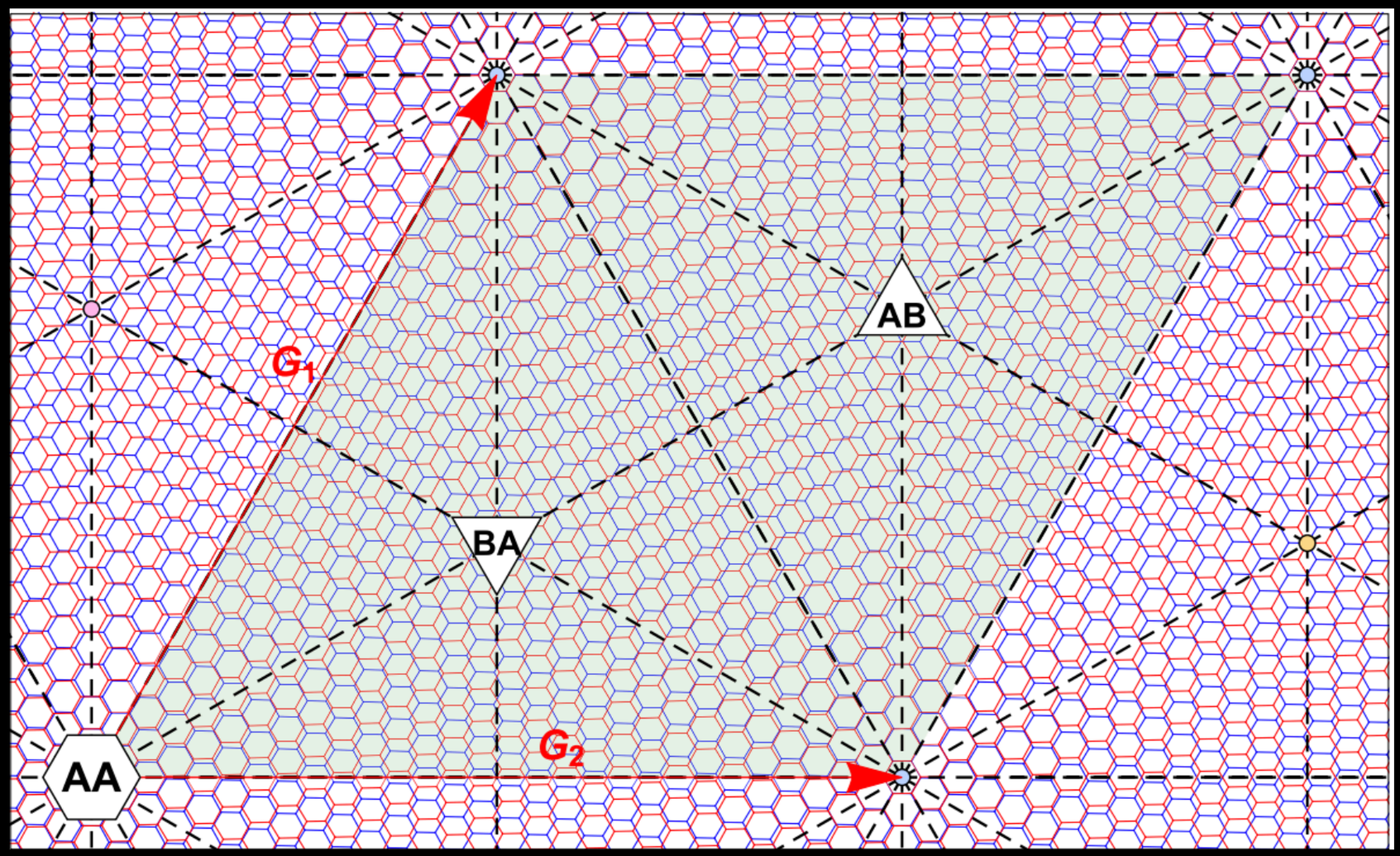
spatial variation of the LDOS. Figures 3(a) and 3(b) show representative STS maps recorded at energies away from the VHSs of the TBG with $\theta \approx 1.2°$. The distribution of the LDOS [Figs. 3(a) and 3(b)] reveals the same period and circular symmetry of the moiré pattern but exhibits the inverted contrast comparing with that shown in the STM image [Fig. 1(b)]. Such a result is reproduced well in our theoretical



We want to understand the nature of its insulating phase by studying the real-space structure of the orbitals that are involved.

We make TBG by starting with AB stacking and rotating one of the layers around an AB site

The new unit cell has unit vectors $\mathbf{G_1 = na_1 + ma_2}$,
$\mathbf{G_2 = -ma_1 + (n + m)a_2}$.

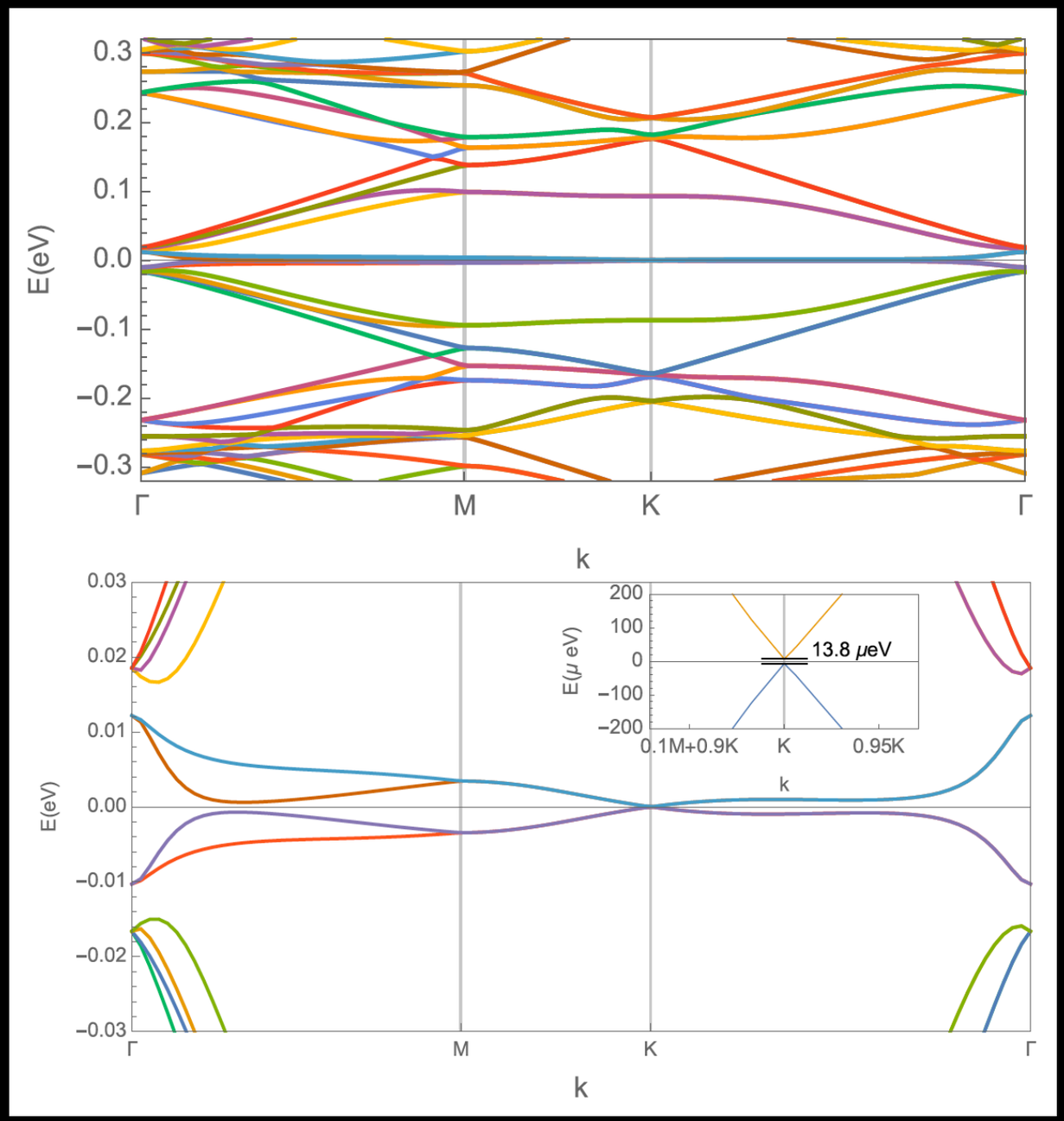


- sixfold rotation around AA centers.
- two threefold rotation around AB and BA centers.
- There are no mirror symmetries

- Magic angle $\theta = 1.08$, generated by choosing $m = 31$ and $n = 30$
- $4(n_2 + nm + m_2) = 11\,164$ atoms in the new unit cell.

Tight binding model

- In-plane n-n hopping t = 2.8 eV. Interlayer hopping $t_\perp(r) = t_{\perp 0}\, e^{-|r|/\xi}$.



r = total distance between two atoms including the interlayer distance d = 0.335 nm,

$\xi = 0.11a$,

a = 0.246 nm

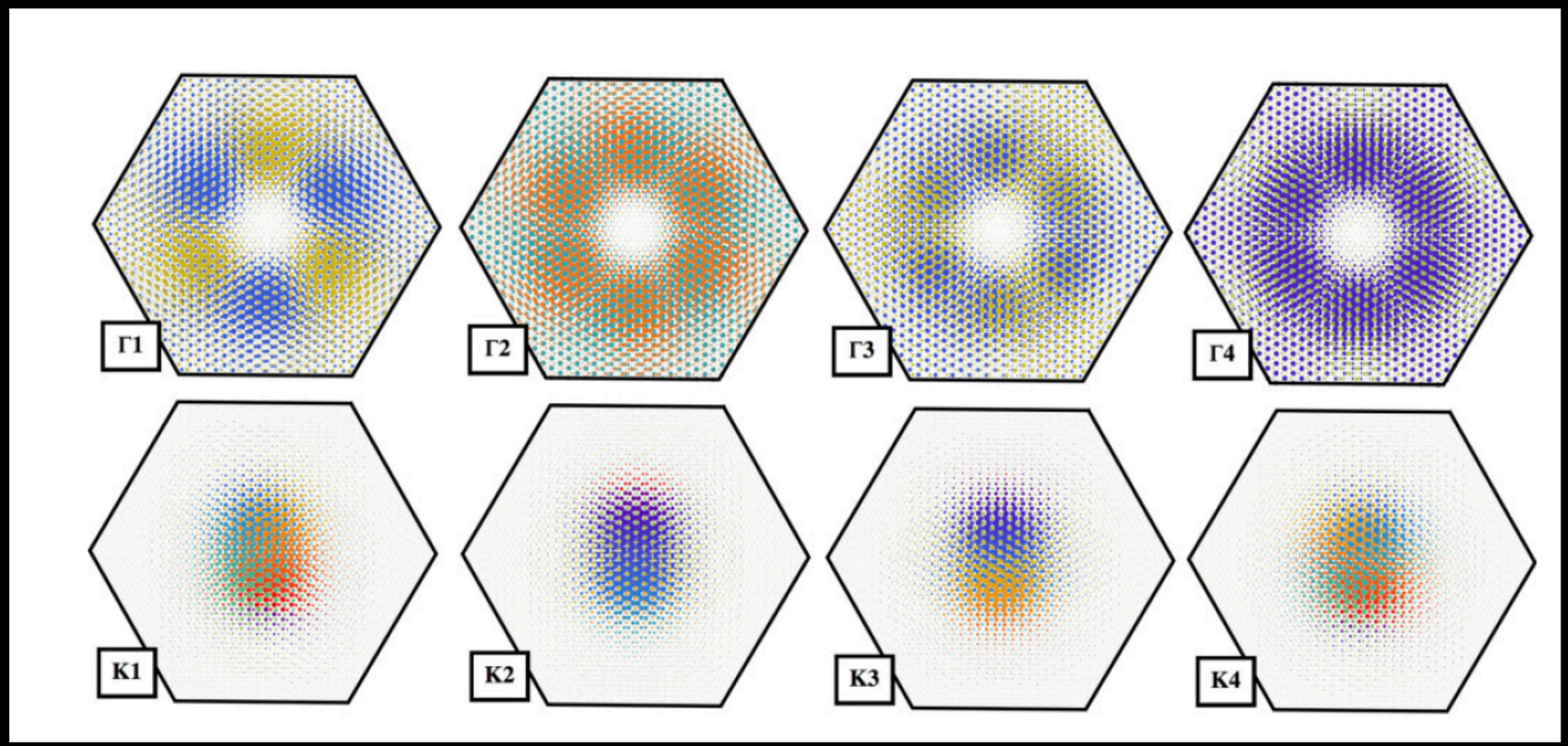
$t_{\perp 0}$ chosen such that $t_\perp$ = 0.35 eV for the AA stacked atoms.

- Four flat bands with a bandwidth W = 11.25 meV around charge neutrality.

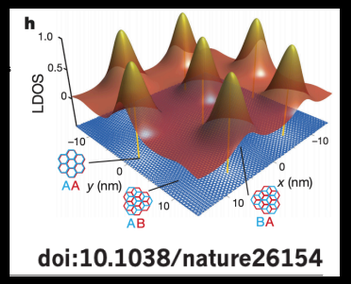
- At the K point there is a small gap of 13.8 μeV, "approximate" Dirac cones, two flat bands above and two flat bands below them.

The real-space change of orbitals

Spatial structure of the low-energy wave functions at K and Γ points



Electrons close to charge neutrality (K point) have large spectral weight at the AA regions consistent with tunneling experiments: we call these “center” orbitals.

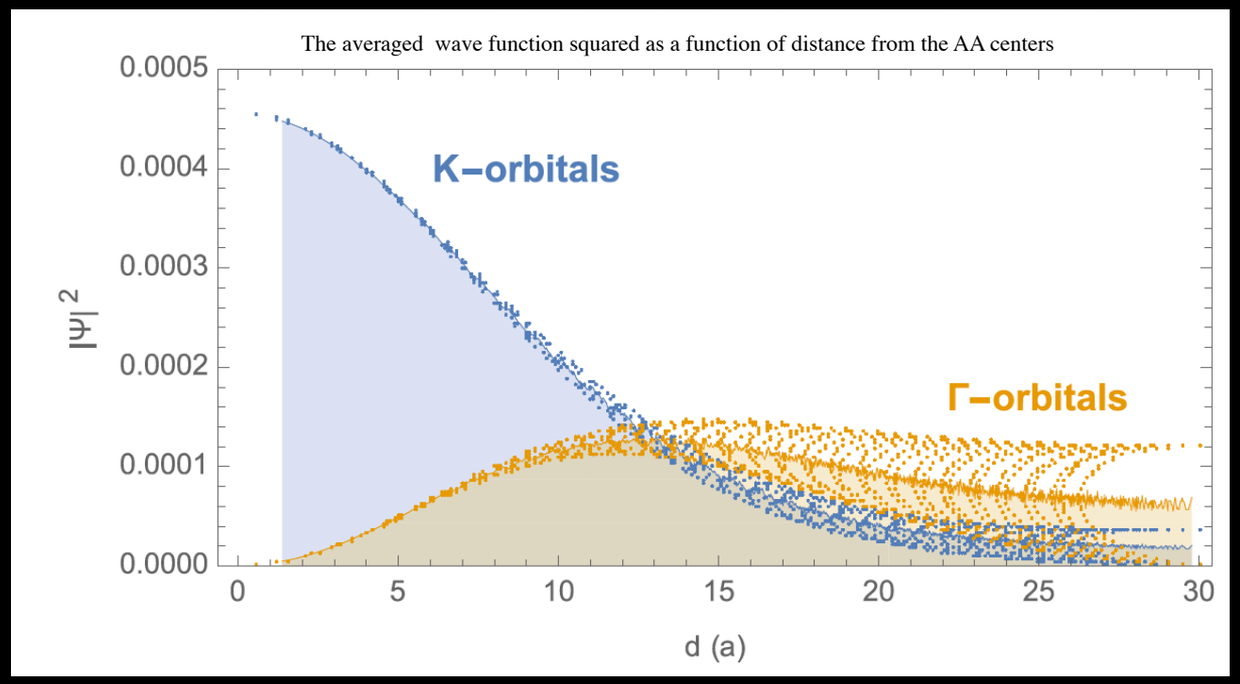


At the Γ point a clear transfer of charge is seen to a ring around the AA center.

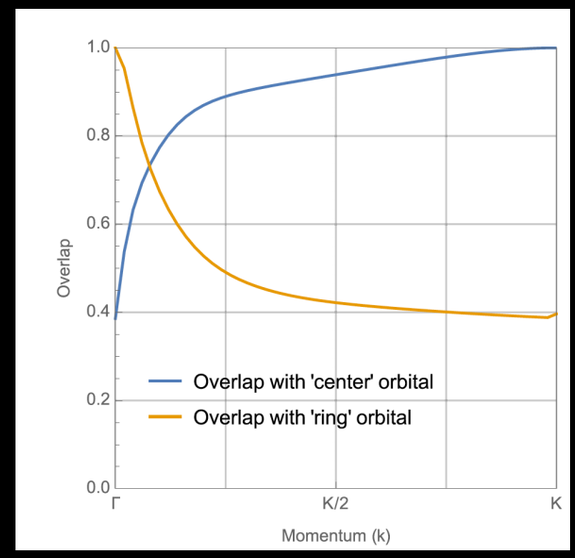
we call these “ring” orbitals.

Flat bands change orbital character between the K and points

In the absence of hybridization between “ring” and “center” orbitals, we have four gapped triangular-lattice orbitals at the Gamma point and fourfold-degenerate Dirac cones.



The orbital character smoothly transforms from ringlike to centerlike. Note that the overlap between ring and center orbitals is ≈0.4.



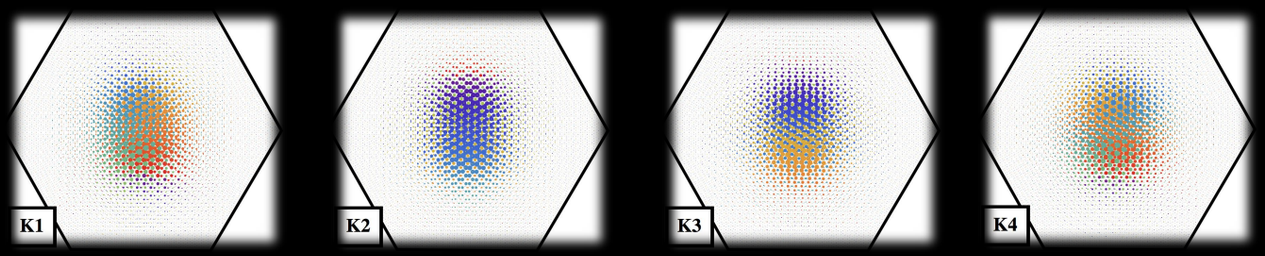
Hybridization between “center” and “ring” orbitals causes the lowest-energy band to be a mix of both orbitals.

Toward an effective model that captures the low energy physics

Hybridization between and K and Gamma orbitals causes the lowest-energy band to be a mix of both orbitals.

Bloch wavefunctions transform under a sum of irreducible representations of the little group; bands at high-symmetry k points will have (non-accidental) degeneracies equal to the dimension of these representations.

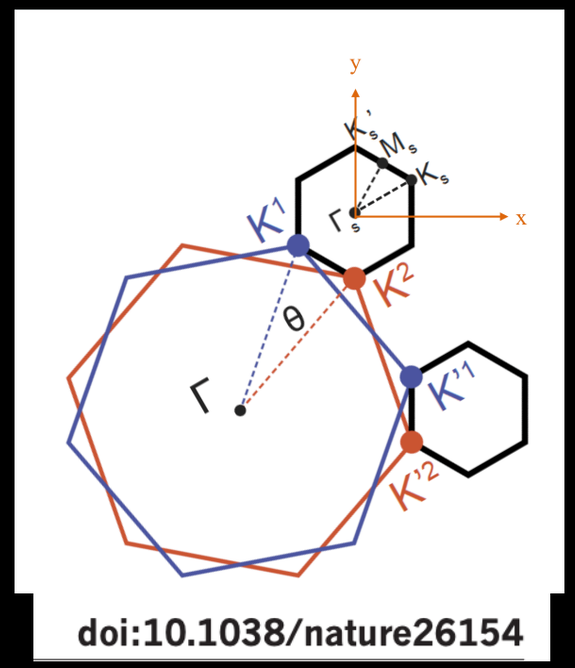
At the K point 'center' orbitals form a triangular lattice



K and K' are exchanged under six- and twofold rotations with respect to the z axis perpendicular to the TBG plane: the little cogroup 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^2-y^2, xy)	(x, y) (R_x, R_y)	E	1 0 0 1	ω 0 0 ω^2	ω^2 0 0 ω

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



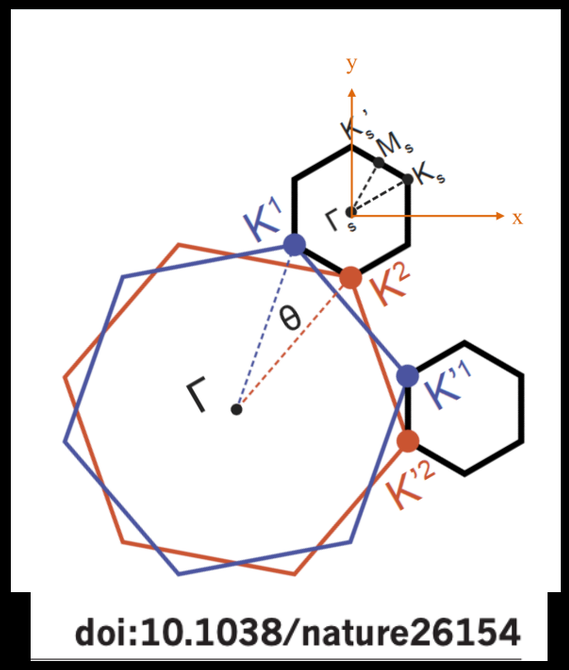
- The reducible (equivalent) representation exp(iKRj) is decomposed into irreps of the group C_3 and is exactly contained in the two-dimensional irrep E of C_3.

- Orbitals at the K point should be two-fold degenerate.

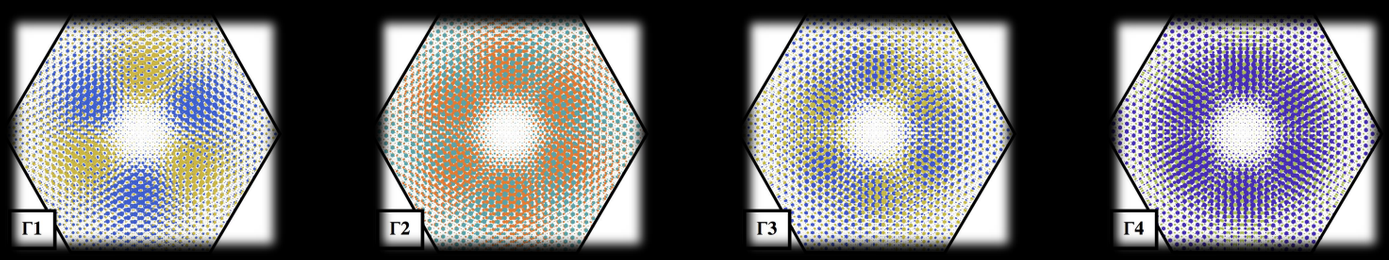
At Γ "ring" orbitals form a triangular lattice.

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) (R_x, R_y)	E'	1	ω	ω^2	ω^3	ω^4	ω^5
			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

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



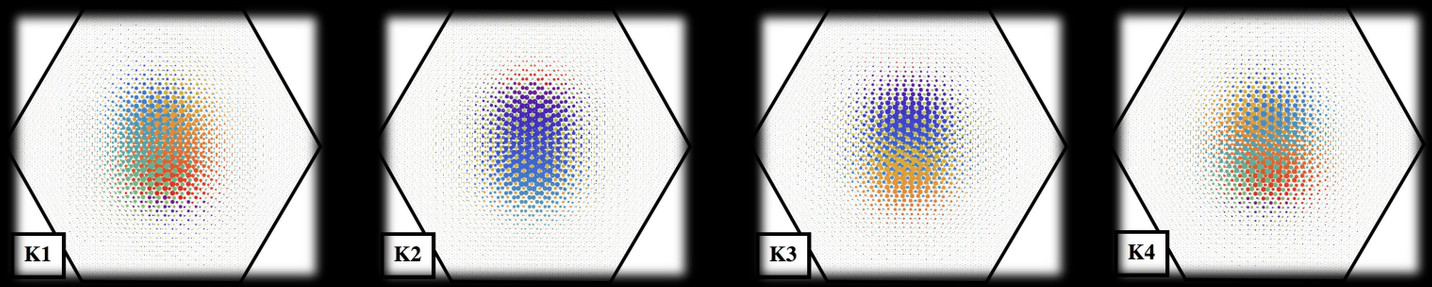
doi:10.1038/nature26154



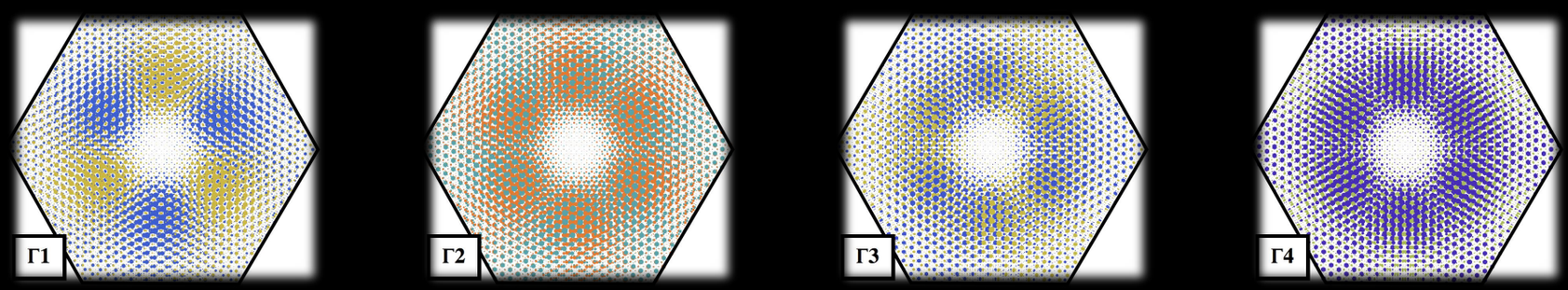
- The little group at the Gamma point is isomorphic to C6.
- At the Gamma point the triangular lattice is invariant under 3-fold rotations.
- Therefore orbitals can realize irreps A and B.

The Hybrid orbital effective model

The “center” orbitals at the K point should be twofold degenerate. However, because the gap at the approximate Dirac cones is practically unobservable, the effective hopping among K (center) orbitals can be approximated by a honeycomb symmetry



The “ring” orbitals, on the other hand, are far away from the approximate Dirac cones and will be treated as having hopping on a triangular lattice.



Effective (non interacting) Model

We split the eight lowest-energy bands in two degenerate valleys. Each valley consists of two “ring” and two “center” orbitals

Effective model consists of two degenerate 4 x 4 blocks

$$H_4(\mathbf{k}) = \begin{pmatrix} 0 & t_K f_K(\mathbf{k}) & t' f_{K\Gamma_1}(\mathbf{k}) & 0 \\ t_K f_K(-\mathbf{k}) & 0 & 0 & -t' f_{K\Gamma_2}(\mathbf{k}) \\ t' f_{K\Gamma_1}(-\mathbf{k}) & 0 & -\Delta_\Gamma + t_\Gamma f_\Gamma(\mathbf{k}) & 0 \\ 0 & -t' f_{K\Gamma_2}(-\mathbf{k}) & 0 & \Delta_\Gamma - t_\Gamma f_\Gamma(\mathbf{k}) \end{pmatrix}$$

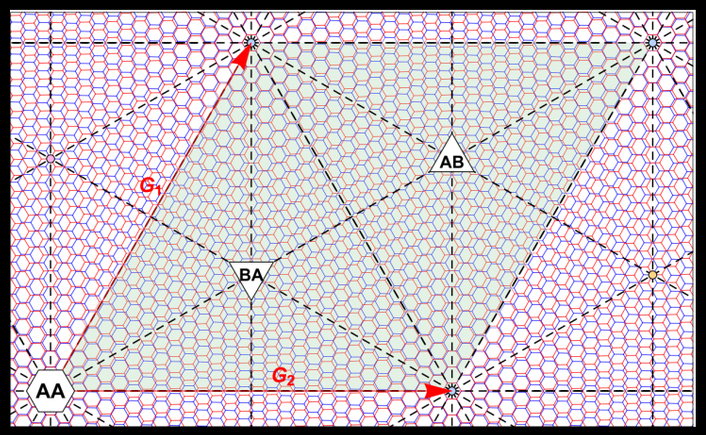
$$f_K(\mathbf{k}) = \mathbf{1} + \mathbf{e}^{\mathbf{i}\mathbf{k}\mathbf{a}_1\cdot\mathbf{k}} + \mathbf{e}^{\mathbf{i}\mathbf{k}\mathbf{a}_2\cdot\mathbf{k}}$$

$$f_\Gamma(\mathbf{k}) = \mathbf{2}\left(\cos \mathbf{k}\mathbf{a}_1 \cdot \mathbf{k} + \cos \mathbf{k}\mathbf{a}_2 \cdot \mathbf{k} + \cos \mathbf{k}\mathbf{a}_3 \cdot \mathbf{k}\right)$$

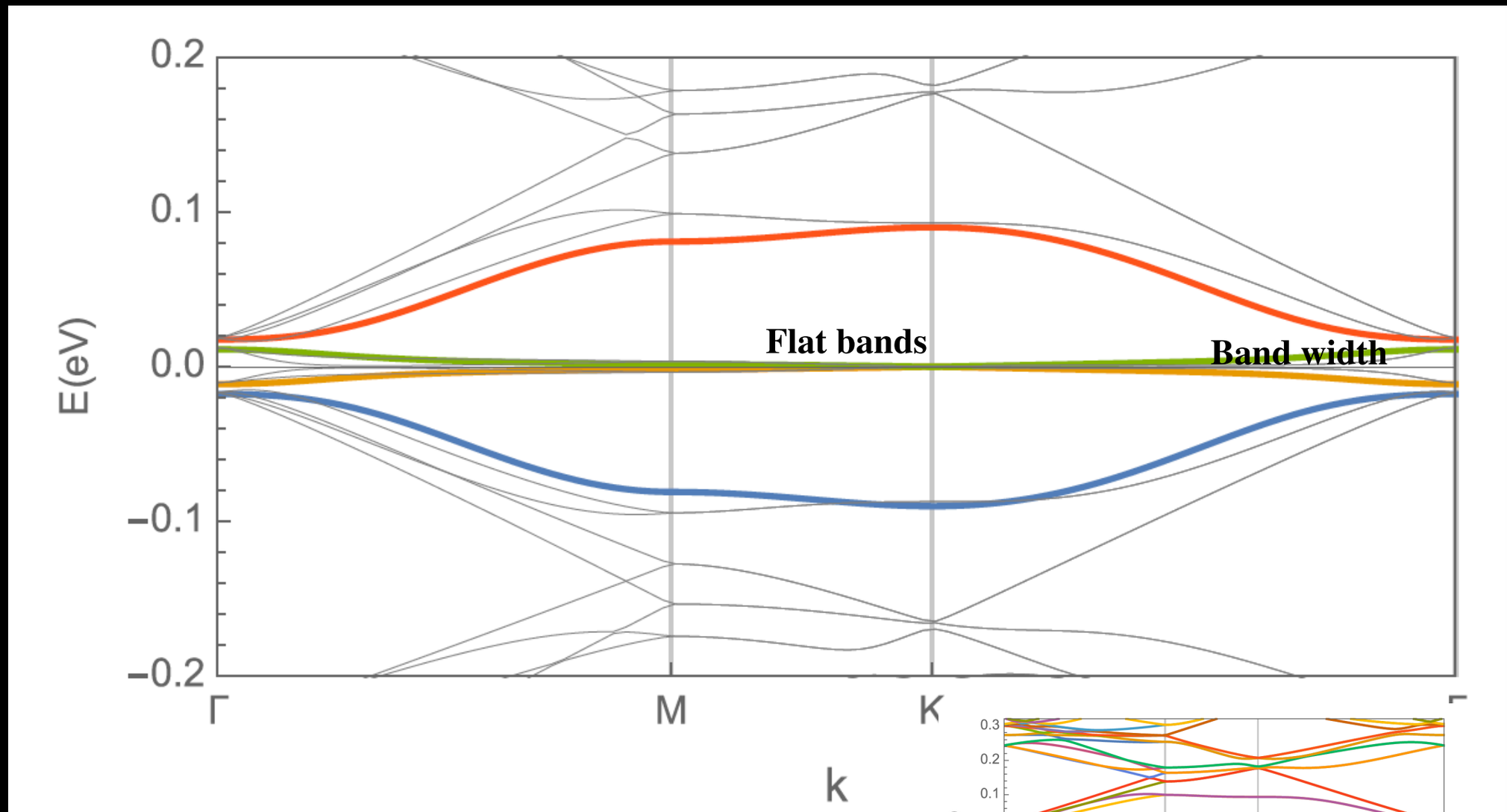
$$f_{K\Gamma_1}(\mathbf{k}) = \mathbf{1} + \mathbf{e}^{-\mathbf{i}\mathbf{a}_1\cdot\mathbf{k}} + \mathbf{e}^{-\mathbf{i}\mathbf{a}_2\cdot\mathbf{k}}$$

$$f_{K\Gamma_2}(\mathbf{k}) = \mathbf{e}^{-\mathbf{i}(\mathbf{a}_1+\mathbf{a}_2)\cdot\mathbf{k}} + \mathbf{e}^{-\mathbf{i}\mathbf{a}_1\cdot\mathbf{k}} + \mathbf{e}^{-\mathbf{i}\mathbf{a}_2\cdot\mathbf{k}},$$

$$t_K = 1.5312\,\text{meV}$$
$$t_\Gamma = 9.5007\,\text{meV}$$
$$t' = 4.6730\,\text{meV}$$
$$\Delta_\Gamma = 61.5978\,\text{meV}$$

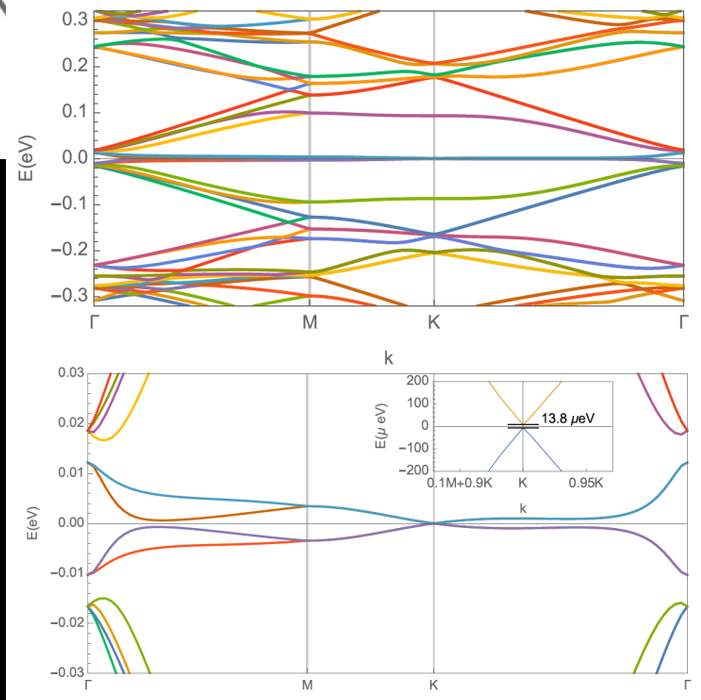


H_4 reproduces the lowest energy bands of the full problem with the right symmetries



It can be improved? Sure!

We are working on it



Consequences of the hybrid orbital picture:

A) The interaction energy is minimized in the **insulating phase** by a charge-transfer from the "center" to the "ring" orbitals.

B) The transfer of the charge picture permits to fix the spin degrees of freedom in the Mott phase, naturally yielding an effective antiferromagnetic Heisenberg Hamiltonian which gives rise to a spin-singlet paramagnet.

C) Doping this paramagnet can lead to superconductivity, with different pairing symmetries for different dopings.

A) Interactions and the Mott phase at n=2 per Moire unit cell

Single-layer graphene has a relatively strong on-site interaction U = 9.3 eV, nearest neighbor Coulomb interaction strength was computed to V = 5.5 eV.

PRL **106,** 236805 (2011) PHYSICAL REVIEW LETTERS

Strength of Effective Coulomb Interactions in Graphene and Graphite

T. O. Wehling,[1] E. Şaşıoğlu,[2] C. Friedrich,[2] A. I. Lichtenstein,[1] M. I. Katsnelson,[3] and S. Blügel[2]

	Graphene		Graphite	
	Bare	cRPA	Bare	cRPA
$U_{00}^{A \text{ or } B}$ (eV)	17.0	9.3	17.5, 17.7	8.0, 8.1
U_{01} (eV)	8.5	5.5	8.6	3.9
$U_{02}^{A \text{ or } B}$ (eV)	5.4	4.1	5.4, 5.4	2.4, 2.4
U_{03} (eV)	4.7	3.6	4.7	1.9

Experiments: upon doping away from charge neutrality the electronic charge density will cluster around the AA centers: charge inhomogeneities.

For orbitals that span thousands of different atoms, the full Coulomb interaction beyond the on-site repulsion plays a central role.

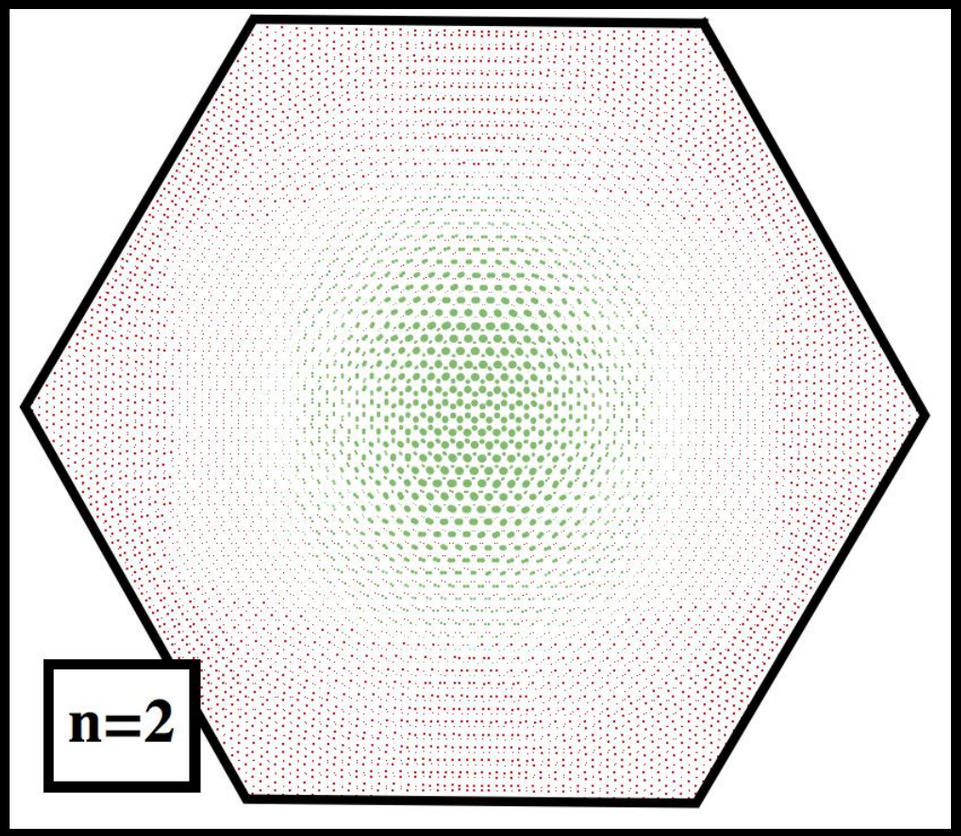
$$E_{\text{int}} = \sum_{r,r'} \delta n(r) \frac{e^2}{4\pi\epsilon(r-r')\,|r-r'|} \delta n(r')$$

The Coulomb energy is nonzero whenever there are macroscopic charge inhomogeneities $\delta n(r)$

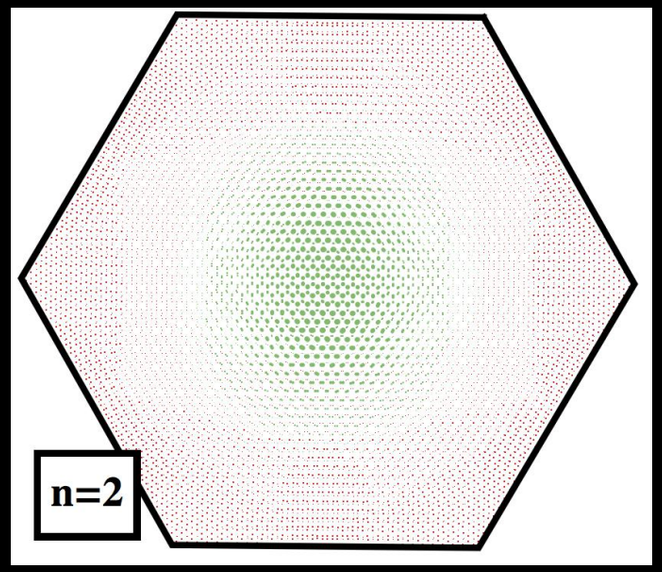
$\delta n(r)$: deviation from the average electron density $\overline{n}$ at position r

Away from charge neutrality (n=0) there will be a large "classical" contribution to the Coulomb energy.

Charge distribution in one unit cell at a density of n=2 holes relative to charge neutrality (n=0) using the full band structure.

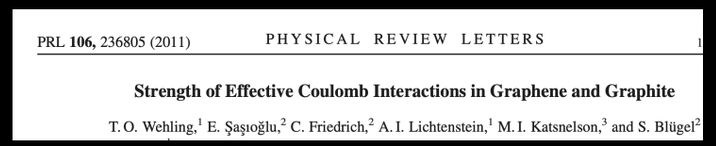


A large concentration of green dots (excess charge) is seen in the AA centers, (91%) whereas around the AB/BA centers there is a depletion of charge (red dots).



$$E_{\text{int}} = \sum_{r,r'} \delta n(r) \frac{e^2}{4\pi\epsilon(r-r')\,|\,r-r'\,|} \delta n(r')$$

The long-wavelength limit of the dielectric constant approaches 1


PRL **106,** 236805 (2011) PHYSICAL REVIEW LETTERS

Strength of Effective Coulomb Interactions in Graphene and Graphite

T. O. Wehling,[1] E. Şaşıoğlu,[2] C. Friedrich,[2] A. I. Lichtenstein,[1] M. I. Katsnelson,[3] and S. Blügel[2]

Effective form of the screened Coulomb interaction in the tight-binding model,

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

Classical Coulomb energy at half-filling of the flat band is large:

$$E_{\text{int}} = 317.6 \quad \text{meV!}$$

Charge-transfer in the Mott phase minimize interaction energy

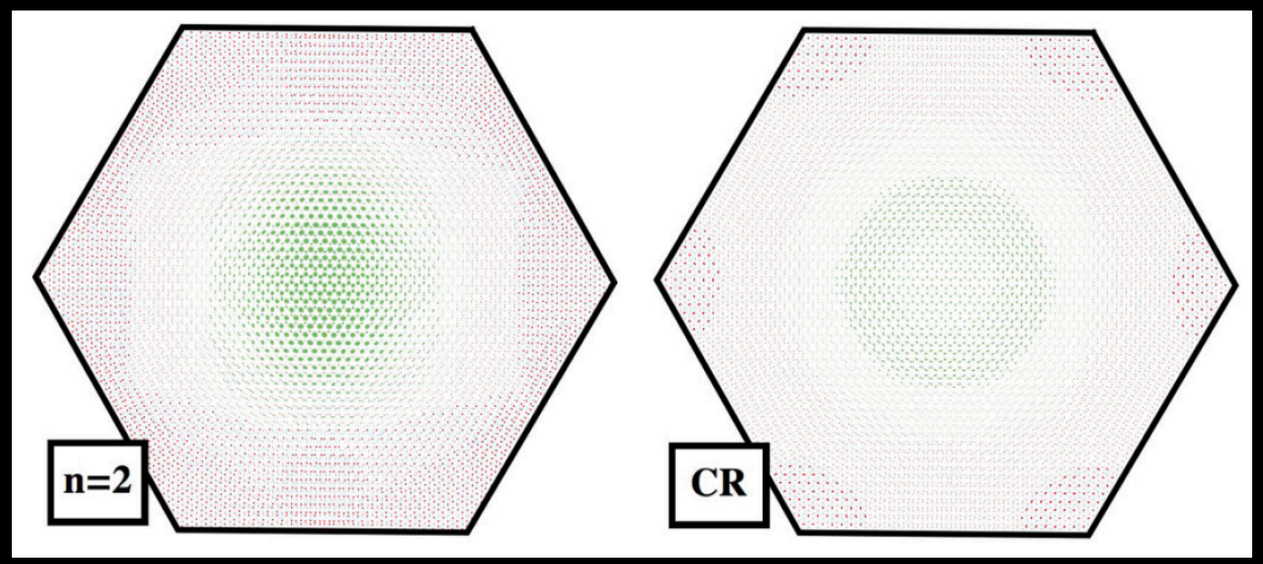
n=2: Charge transfer from the "center" to the "ring" orbitals.

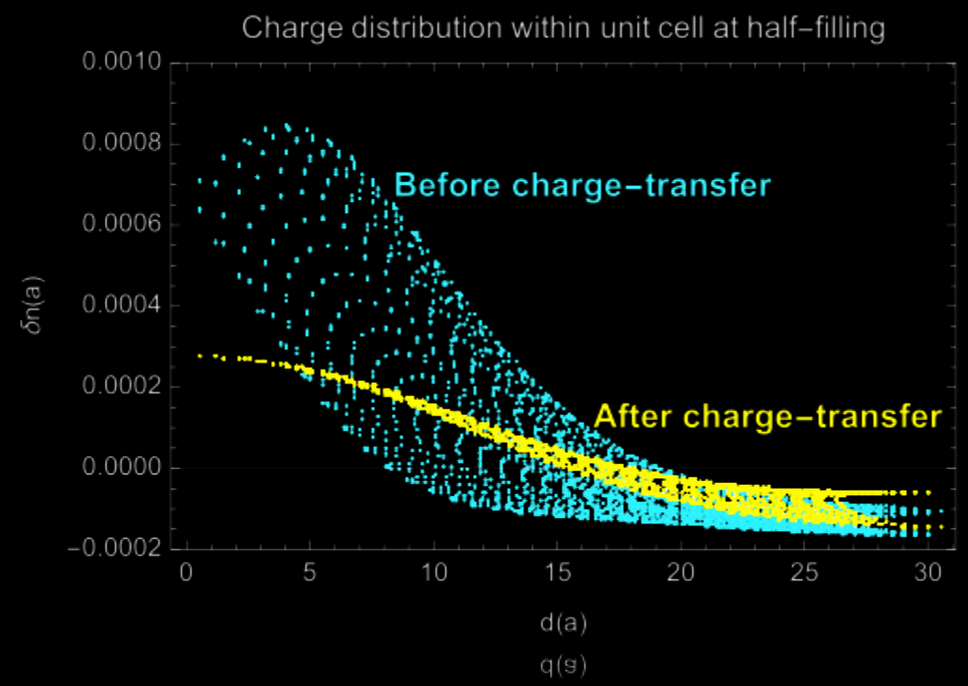
At zeroeth order: one localized hole in the center orbital and one localized hole in the ring orbital.

- Kinetic energy: localization of charges increases the energy per unit cell by 7.86 meV.
- Gap: need to pay the gap 61.6 meV of the ring orbital.
- Interaction energy: drastically reduced to 103.9 meV.

Overall, the average energy gain per unit cell due to the charge transfer is estimated at delta E = 154 meV.

When doping away from charge neutrality a charge-transfer occurs from the center to the ring orbitals because Coulomb repulsion wants to smoothen out charge.





B) The spin degree of freedom in the charge transfer picture is fixed

- One localized charge in a "center" orbital and one in the "ring" orbital,
- Nonzero overlap between "ring" and "center" orbitals, t' = 4.67 meV.
- Localizing one charge in a "ring" and one in a "center" orbital: effective antiferromagnetic Heisenberg coupling between their spins

$$H_{\text{eff}} = J \sum_i \vec{S}_{i\mathbf{K}} \cdot \vec{S}_{i\mathbf{\Gamma}} \qquad J = 2\frac{|t'|^2}{\Delta E} = 0.28\,\text{meV}$$

- The two spins in each unit cell will form a singlet. The Mott phase in TBG is a non-entangled featureless spin-singlet paramagnet.
- J = 3.3 Kelvin, consistent with the energy scale required to break the insulating state.
- Ferromagnetic Heisenberg-like coupling for the orbital degrees of freedom.

C) Dopping away from n=2 (bold claims):

Dynamics of the dopants is described by a t-J model: effective nearest-neighbor attraction between dopants.

Adding electrons to the insulating state will add dopants on the "ring" orbitals: nearest-neighbor attraction on a triangular lattice. The most likely superconducting state would be spin-singlet d+id-wave.

Unconventional superconductivity on the triangular lattice Hubbard model

Kuang Shing Chen, Zi Yang Meng, Unjong Yu, Shuxiang Yang, Mark Jarrell, and Juana Moreno
Phys. Rev. B **88**, 041103(R) – Published 3 July 2013

Hole-doping adds carriers to the "center"-orbitals, which realize an effective honeycomb lattice: spin singlet p+ip-wave superconductivity, as was proposed for single layer graphene away from charge neutrality.

Superconducting States of Pure and Doped Graphene

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(Received 1 December 2006; published 3 April 2007)

We study the superconducting phases of the two-dimensional honeycomb lattice of graphene. We find two spin singlet pairing states; *s* wave and an exotic $p + ip$ that is possible because of the special structure of the honeycomb lattice. At half filling, the $p + ip$ phase is gapless and superconductivity is a hidden order. We discuss the possibility of a superconducting state in metal coated graphene.

A symmetry difference between the electron and hole-doped superconducting phases relative to the Mott state would be a clear proof of the charge-transfer occurring in TBG.

Outlook

- Based on a simple analysis of the real space wavefunctions of an 11,164-bands model, we propose hybrid low-energy bands that are localized within one unit cell and therefore lead to Hubbard-like on-site interactions.

- A direct consequence is that the Mott phase is a featureless spin singlet paramagnet. The lowest energy spin excitations will be propagating triplets, which could be observed using thin film resonant inelastic X-ray scattering.

- The symmetry difference between electron- and hole-doped superconductors is a result of the charge transfer and should be observable in experiments similar to the phase-sensitive experiments in cuprates.

- The effective model is a hybrid mixture of triangular and honeycomb symmetries. Developing such a model and studying it using both analytical and numerical methods might provide key insights towards the understanding of twisted bilayer graphene.