Atomic defects in topological and correlated twisted van der Waals heterostructures

Jose Lado

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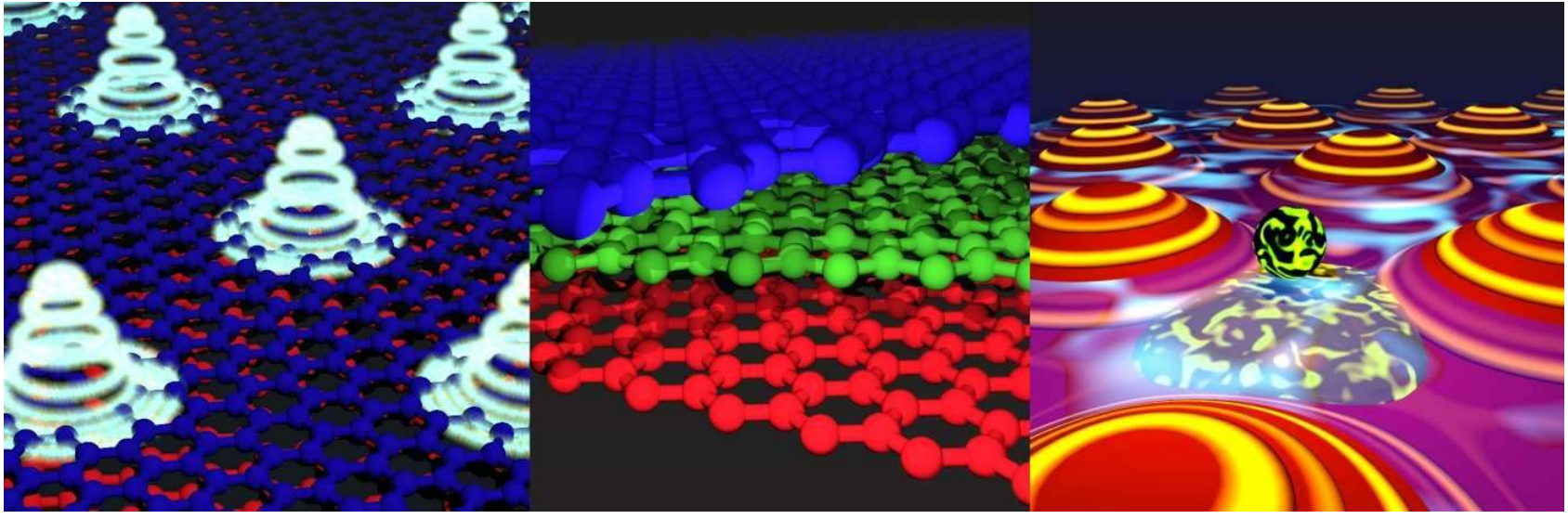


Phys. Rev. B 99, 245118 (2019)
Phys. Rev. Materials 3, 084003 (2019)

Phys. Rev. Research 2, 033357 (2020)

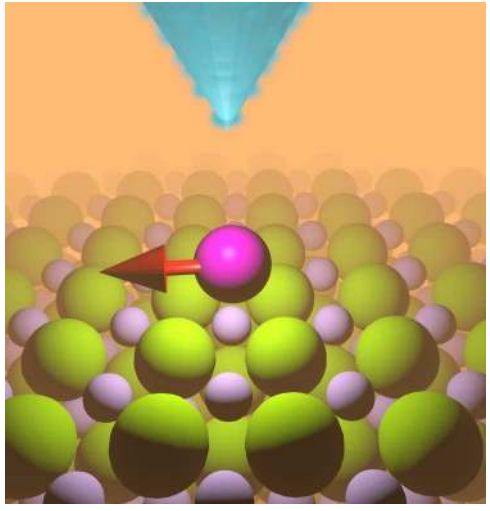
arXiv:2202.11003 (2022)

First-principles modeling of defects in solids, ETH Zürich, Switzerland

June 15th 2022

Building quantum matter with artificial lattices

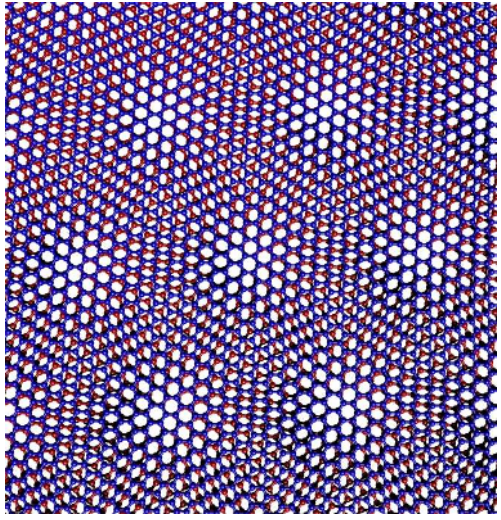
Atomic lattices



a = 0.5 nm

Nature Physics 12 (7), 656 (2016)
Nature Physics 13 (7) 668 (2017)
Science 335 (6065), 196-199 (2012)

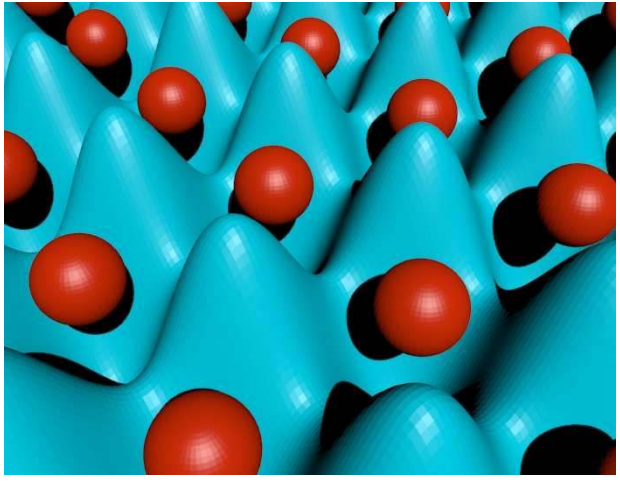
Twisted 2D moire materials



a = 30 nm

Phys. Rev. Lett. 99, 256802 (2007)
Nature Physics 6, 109–113 (2010)
PNAS 108 (30) 12233-12237 (2011)
Nature 556, 80–84 (2018)

Cold atoms



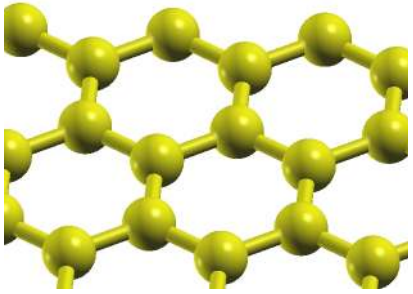
a = 1000 nm

Nature 545, 462–466 (2017)
Nature 455, 204–207 (2008)
Phys. Rev. Lett. 111, 185307 (2013)

The two-dimensional materials world

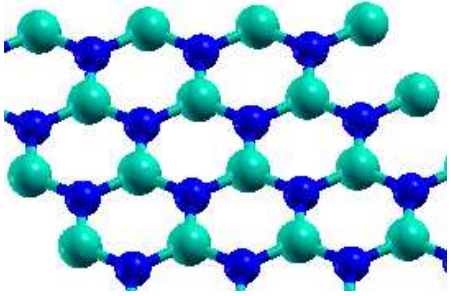
Semimetal

Graphene



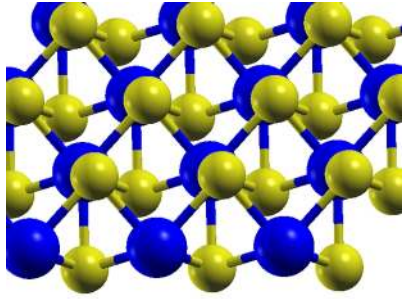
Insulator

BN



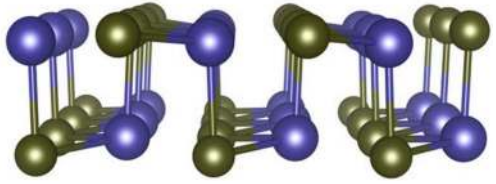
Superconductor

$NbSe_2$



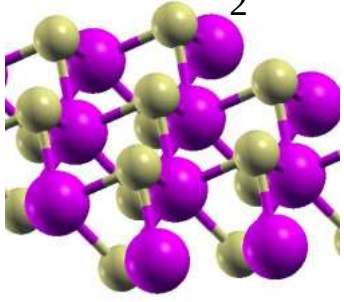
Ferroelectric

SnTe



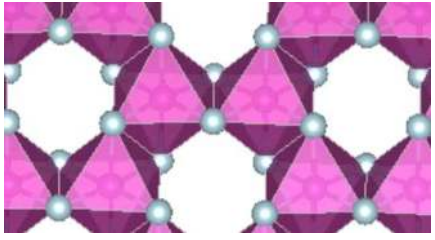
Semiconductor

WSe_2



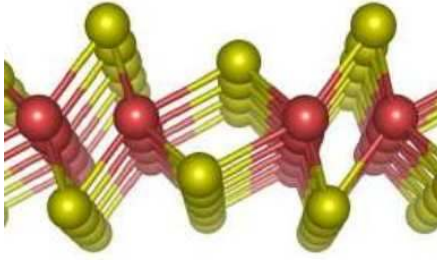
Ferromagnet

CrI_3



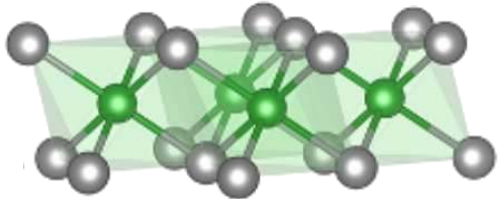
Quantum spin Hall insulator

WTe_2



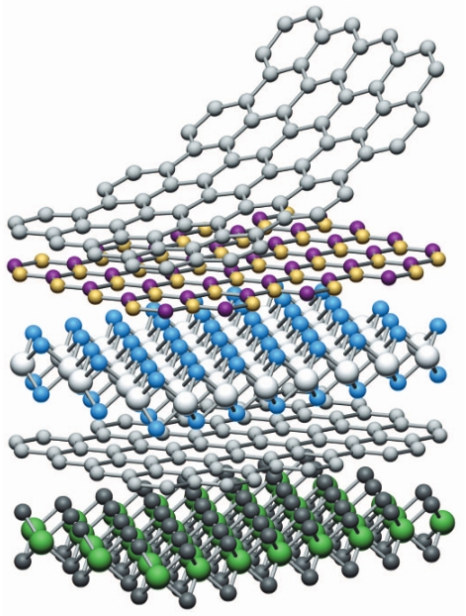
Multiferroic

NiI_2



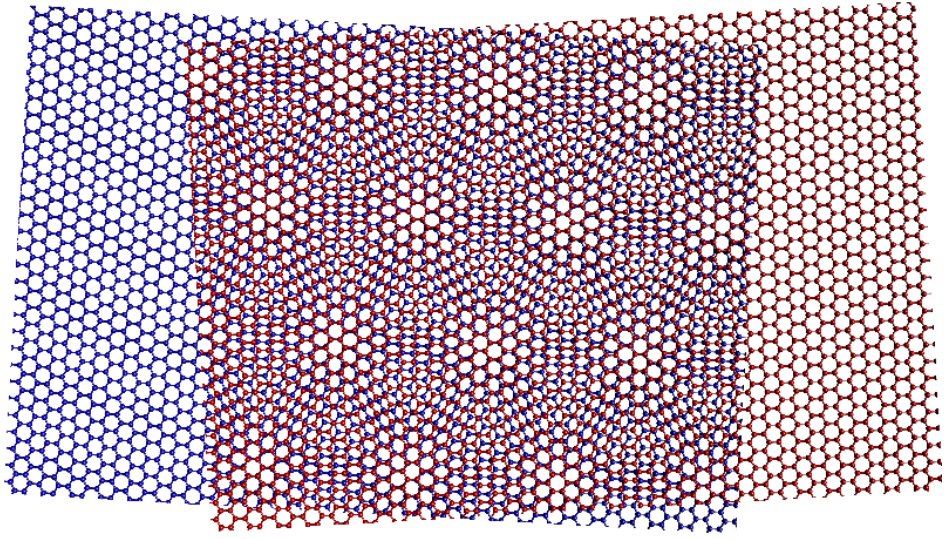
The flexibility of two-dimensional materials

They can be stacked



Nature 499.7459 419. (2013)

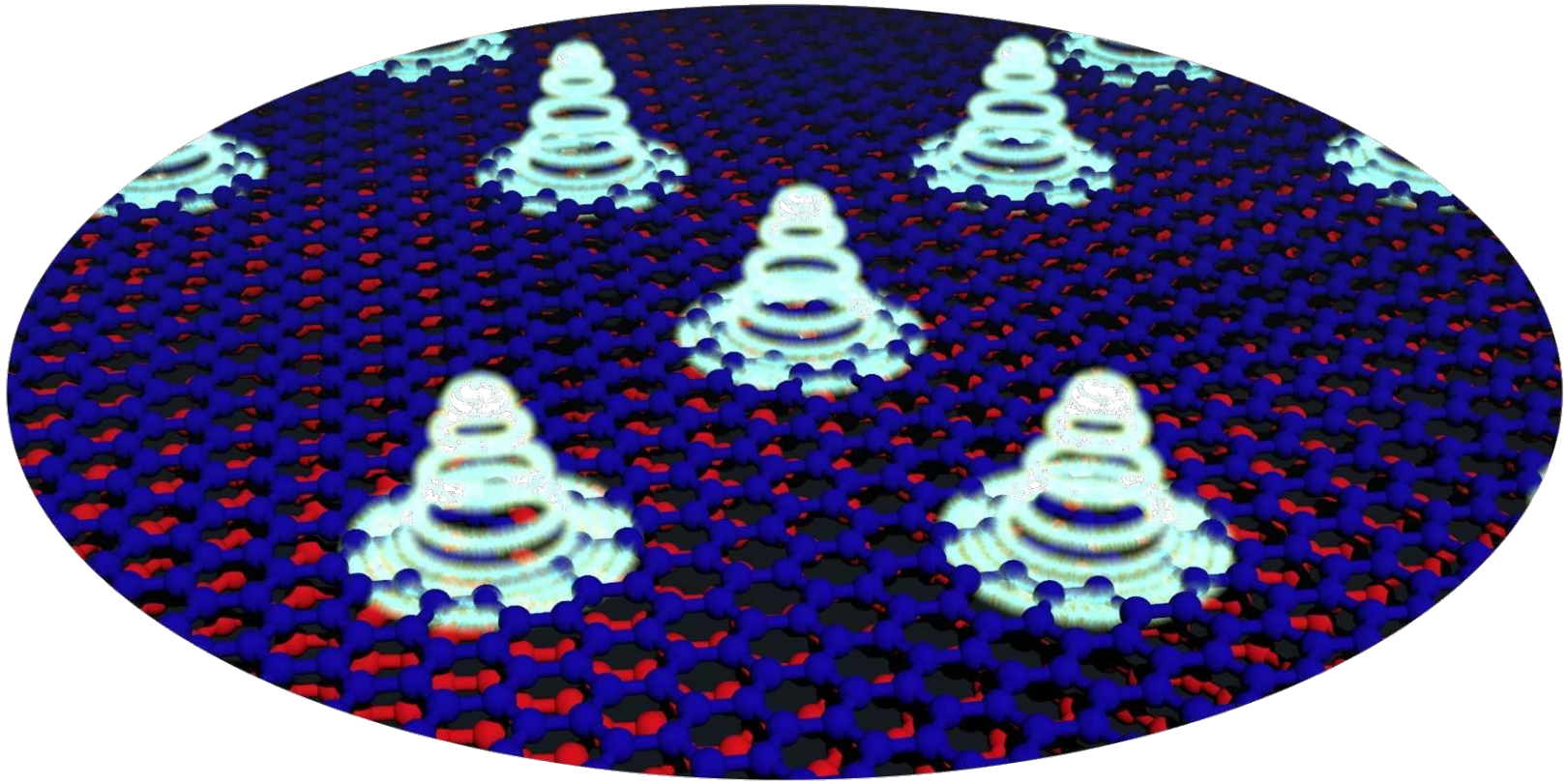
They can be rotated



Science 361.6403 690-693. (2018)

These are unique features of two-dimensional materials

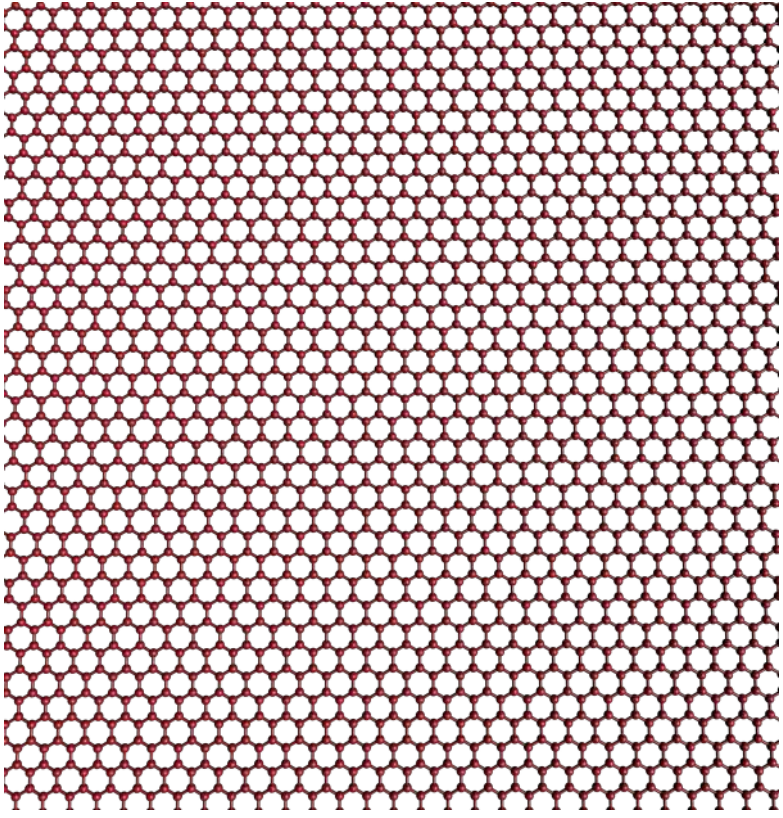
A new universe in each new twisted material



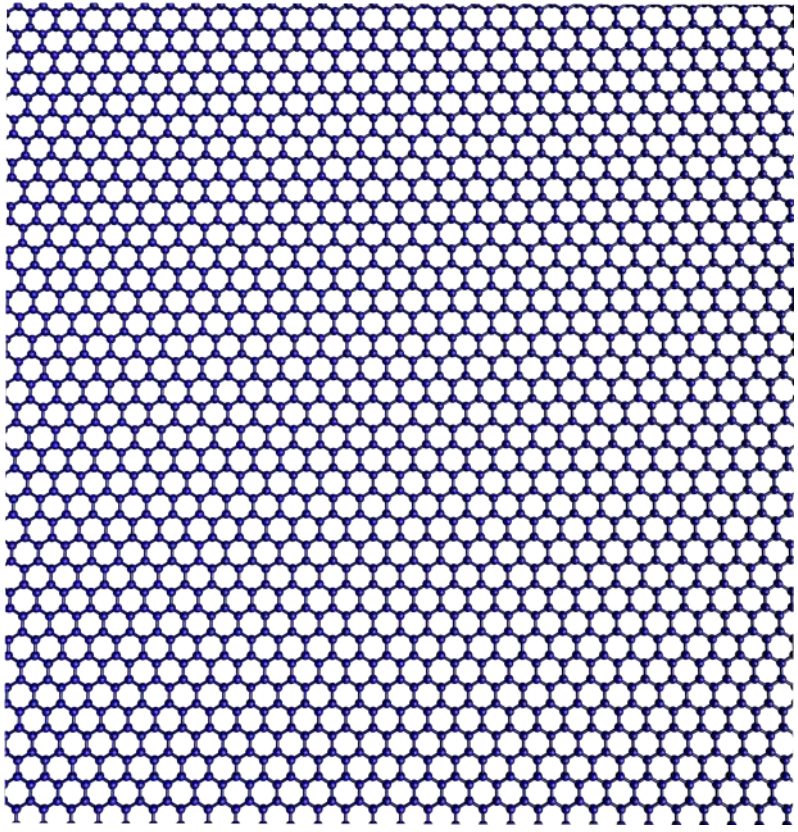
Each twisted material is a new universe for electrons, with laws that change from twist to twist

Moire in twisted bilayer graphene

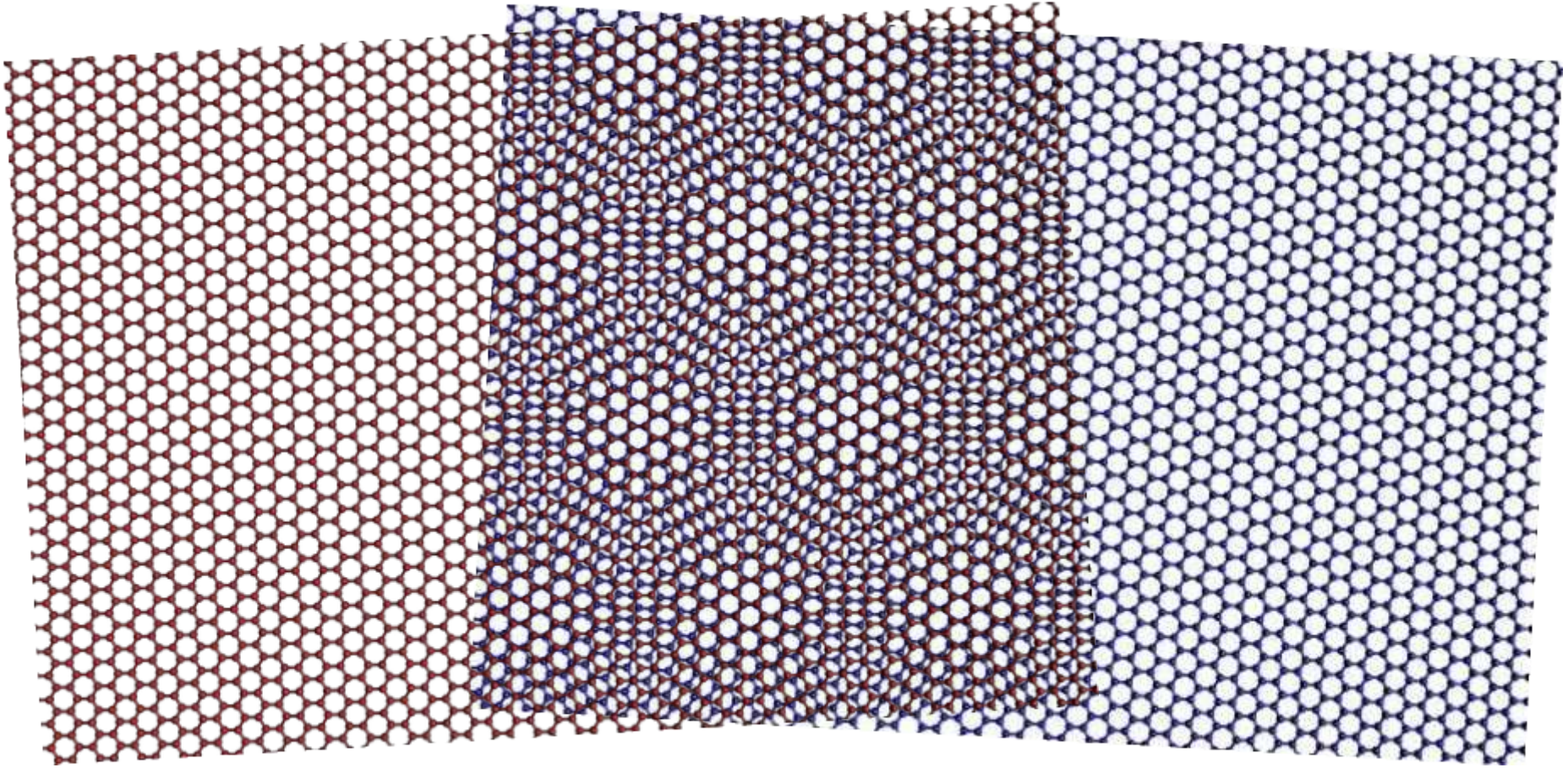
Upper graphene layer



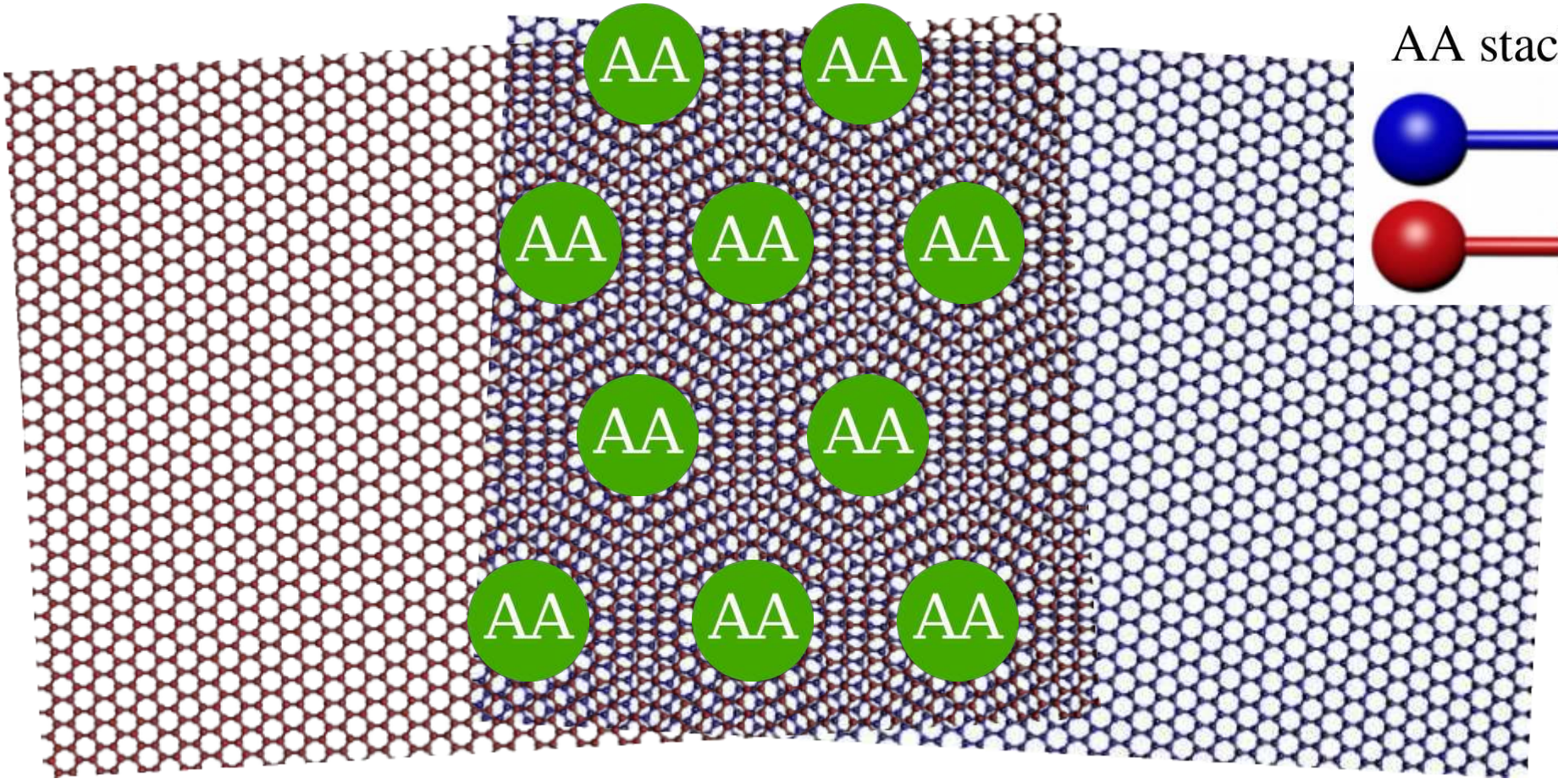
Lower graphene layer



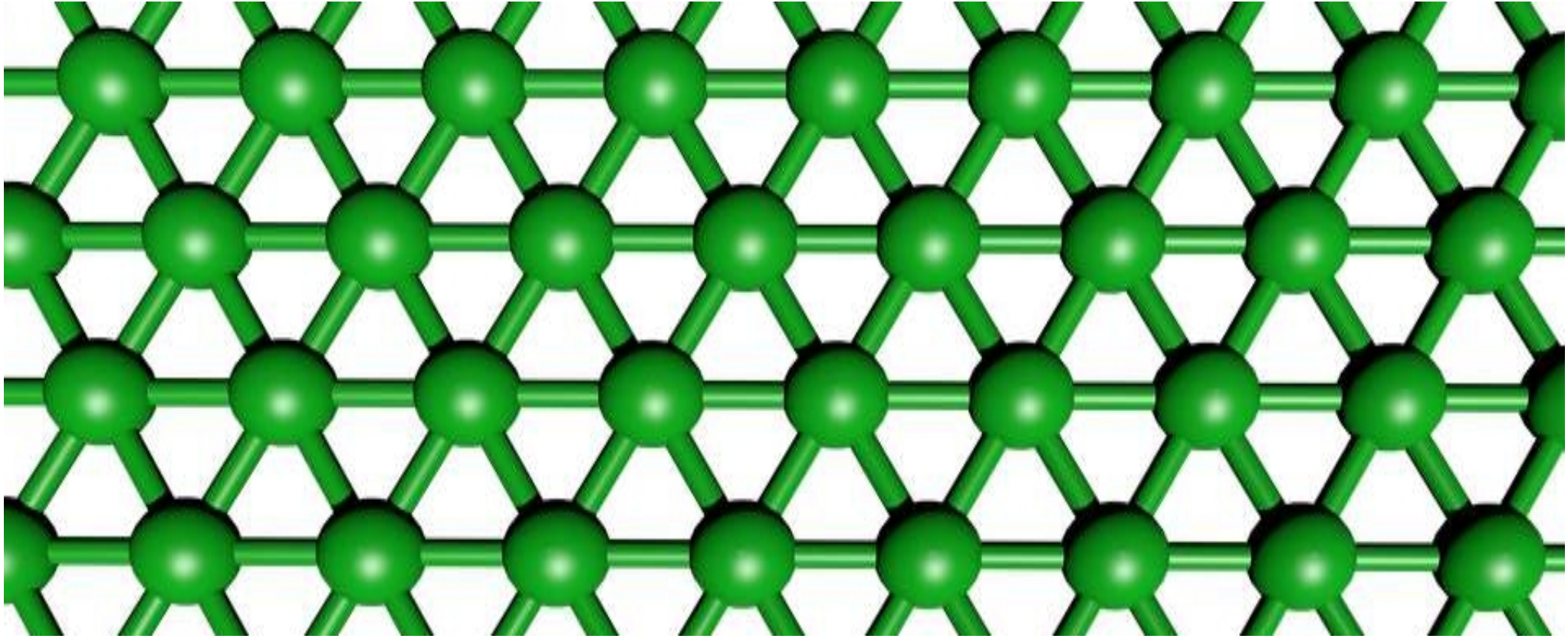
Moire in twisted bilayer graphene



Moire in twisted bilayer graphene

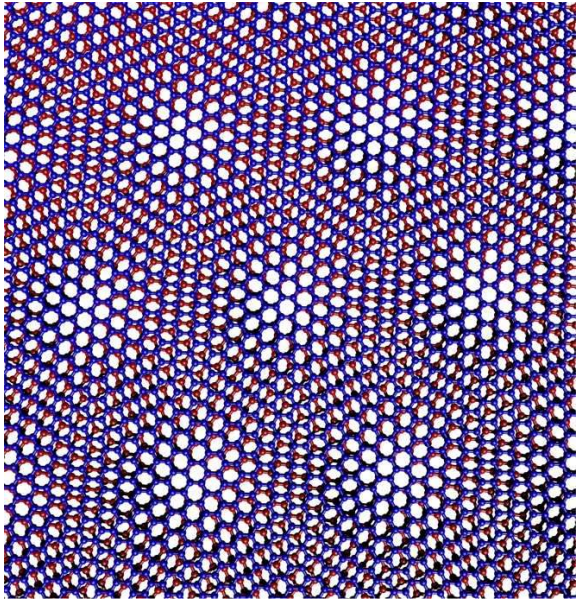


Moire in twisted bilayer graphene

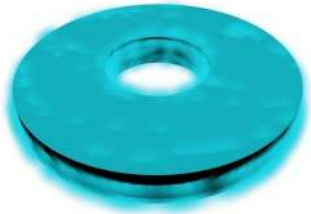


One material, a zoo of electronic phases

Twisted bilayer graphene



Superconductivity

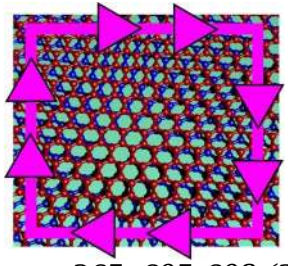


Nature 556, 43–50 (2018)

Topological networks

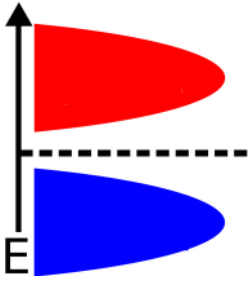
Nano Lett. 18, 11, 6725-6730 (2018)

Chern insulators



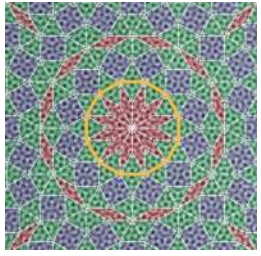
Science 365, 605-608 (2019)

Correlated insulators



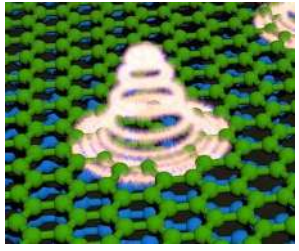
Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782-786 (2018)

Fractional Chern insulators



Nature 600, 439–443 (2021)

Twisted graphene multilayers provide a powerful platform for emergent phenomena

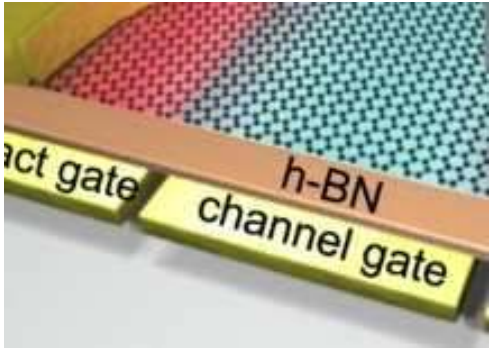
Controlling electronic states in twisted van der Waals materials

Of course, we can use the typical knobs of bulk compounds

(pressure, chemical doping, etc)

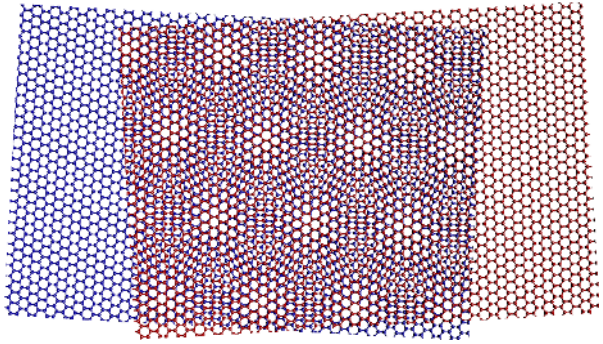
Most importantly, we can exploit the full tunability of van der Waals materials

Gating



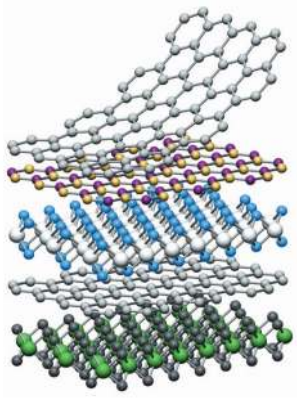
Science 306, 5696, 666-669 (2004)

Twist engineering



Nature Reviews Materials 6, 201–206 (2021)

Materials engineering



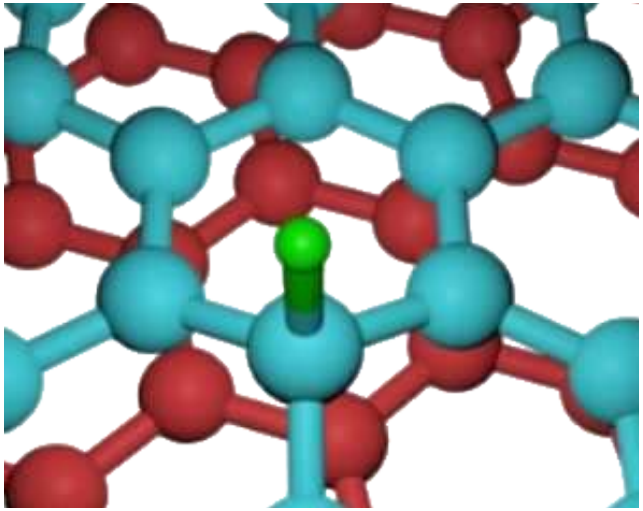
Nature 499, 419–425 (2013)

Allowing to independently control different features of the electronic structure

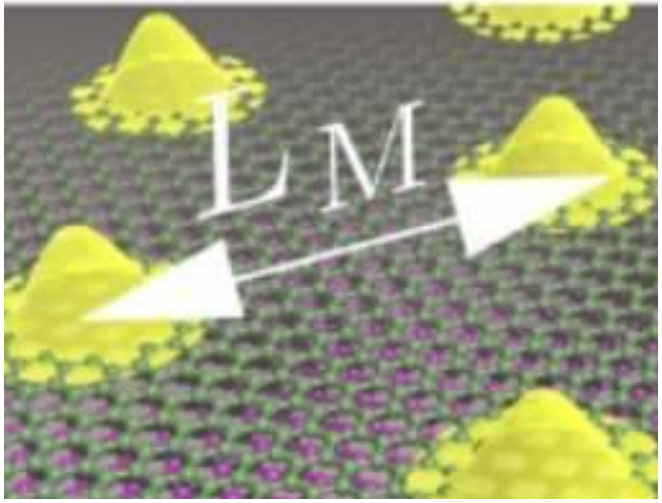
Defects in twisted van der Waals materials

Two length scales in moire systems

The original atomic scale



The emergent moire scale

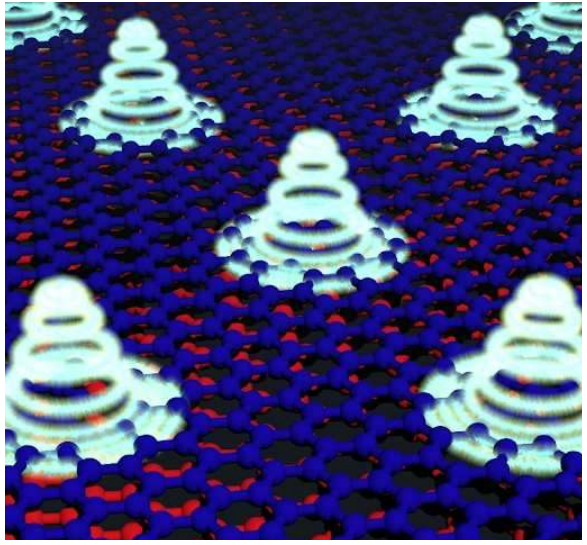


The coexistence of these two different scales is unique to moire systems

How do atomic impurities impact the physics emerging from much bigger length scales?

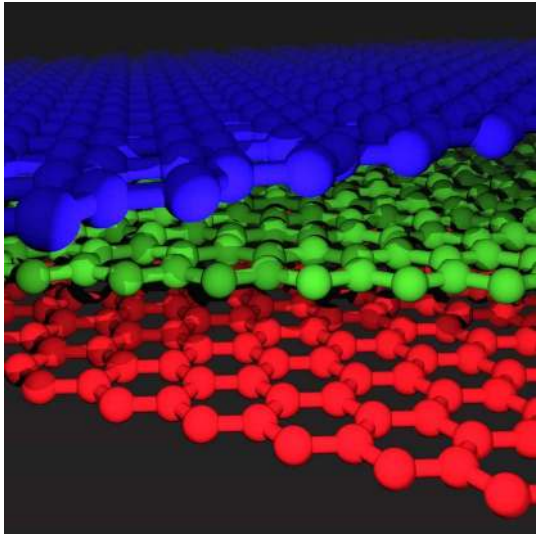
Today's plan

Impurities in twisted graphene bilayers



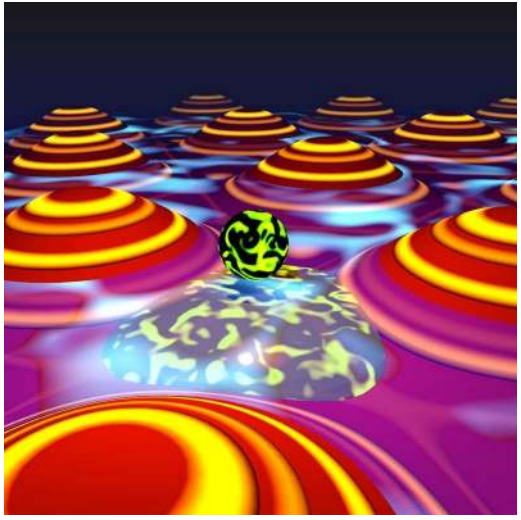
Phys. Rev. B 99, 245118 (2019)
Phys. Rev. Materials 3, 084003 (2019)

Impurities in twisted graphene trilayers



Phys. Rev. Research 2, 033357 (2020)

Impurities in twisted moire topological superconductors



arXiv:2202.11003 (2022)

Behind the scenes

Impurities in twisted graphene bilayers

Aline Ramires

Phys. Rev. B 99, 245118 (2019)

Impurities in twisted graphene trilayers

Alejandro Lopez Bezanilla

Phys. Rev. Research 2, 033357 (2020)
Phys. Rev. Materials 3, 084003 (2019)

Impurities in twisted moire topological superconductors

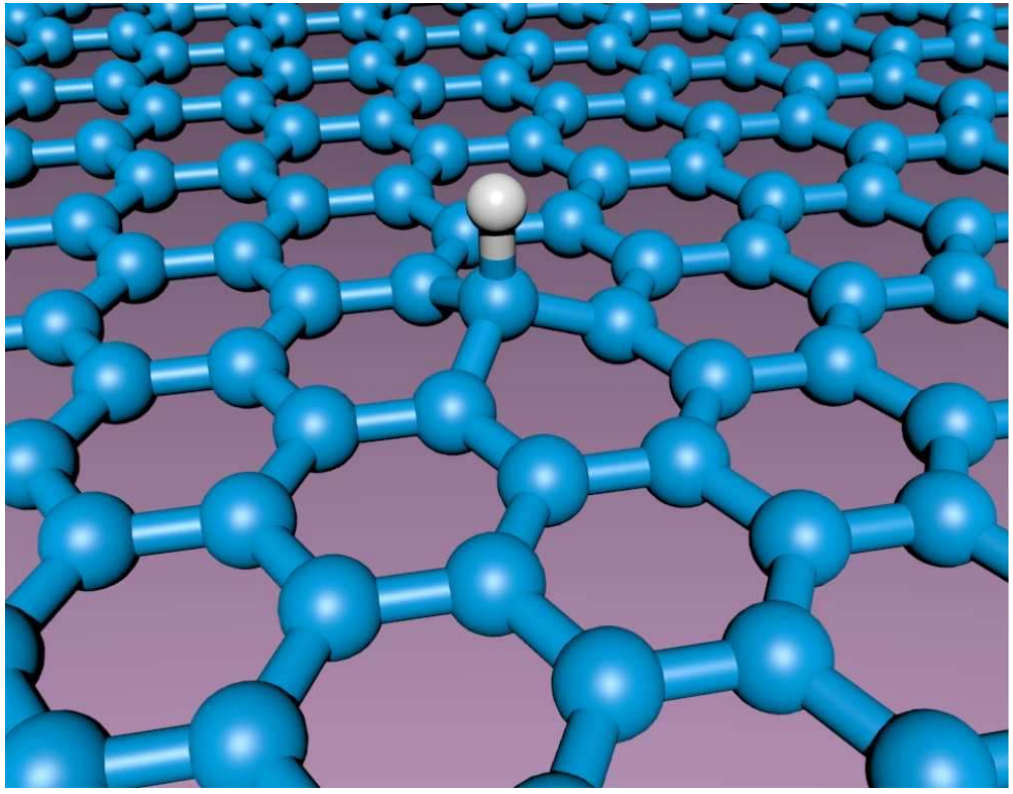
Maryam Khosravian

arXiv:2202.11003 (2022)

Impurities in twisted bilayer graphene

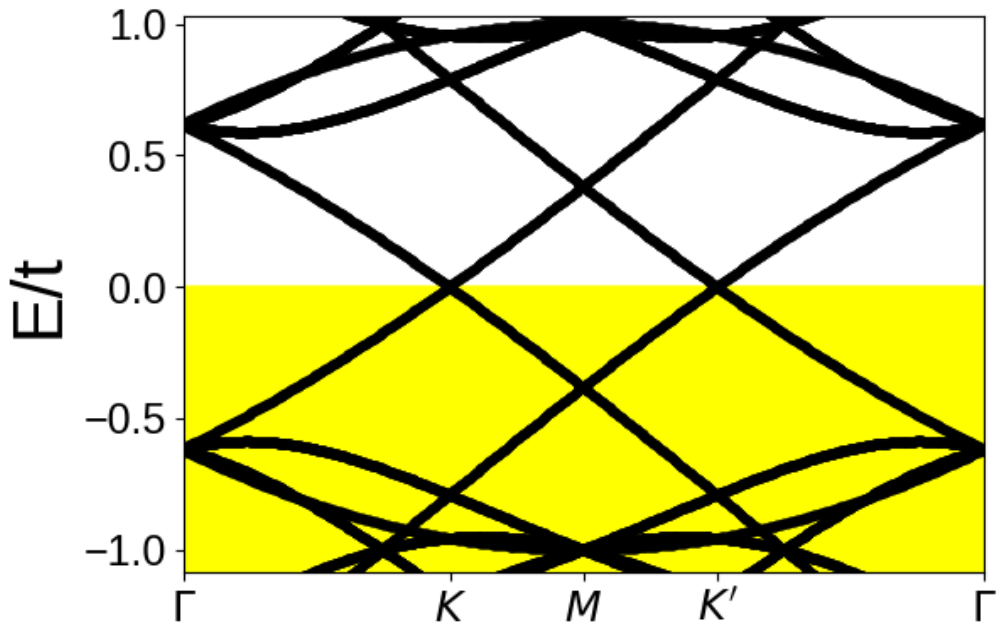
Impurities in monolayer graphene

Hydrogen adsorption in graphene

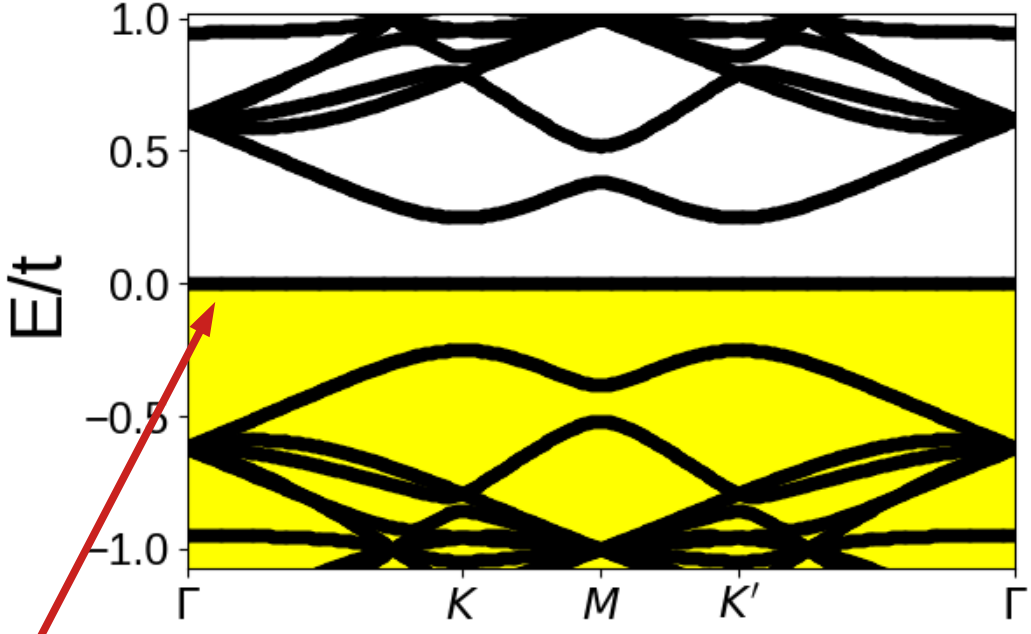


Impurities in monolayer graphene

Pristine 5x5 graphene supercell



Defect in a 5x5 graphene supercell

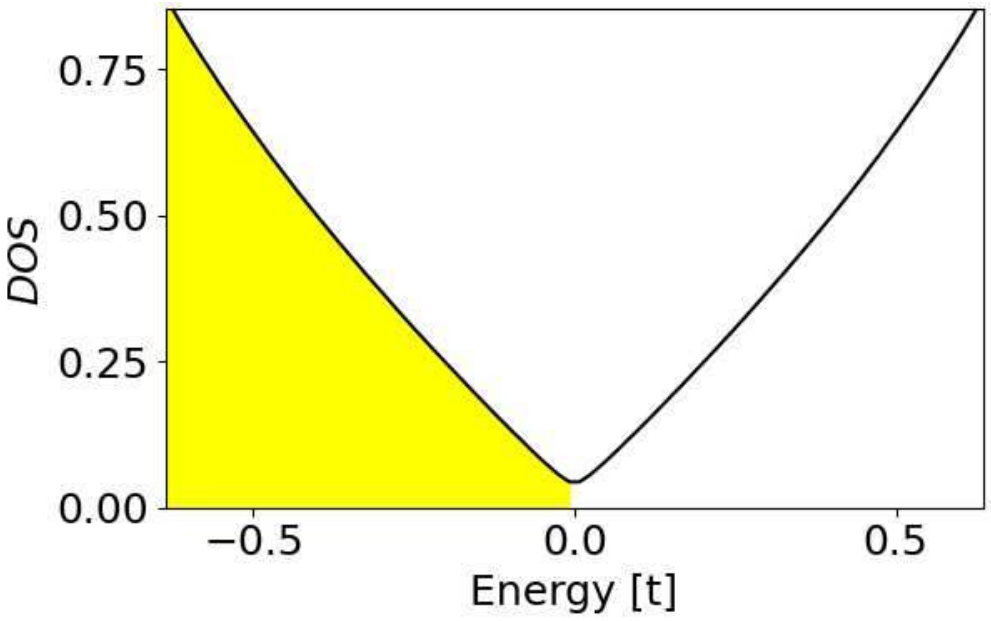


Phys. Rev. B 75, 125408 (2008)
Rep. Prog. Phys. 73 056501 (2010)

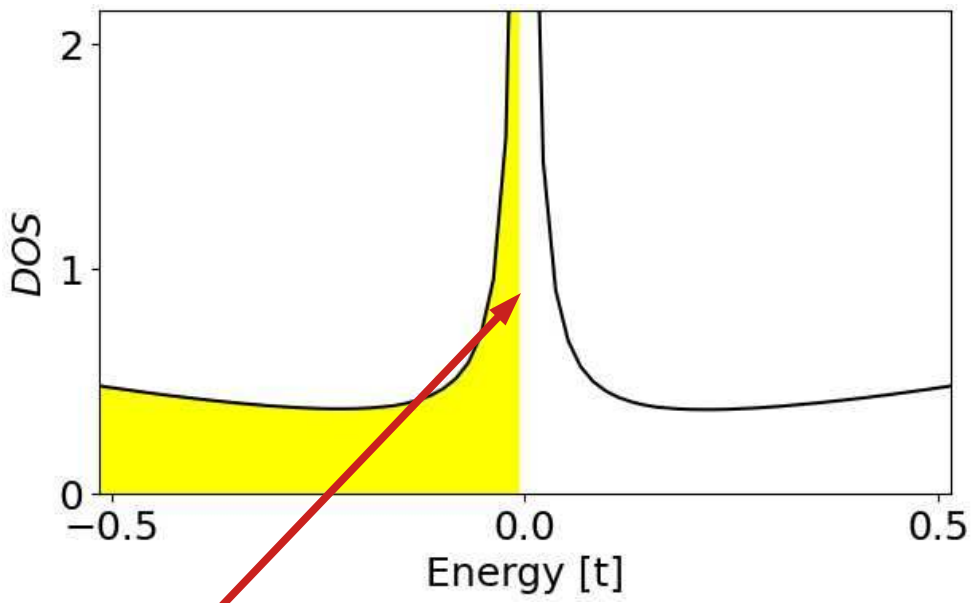
Hydrogen atoms create zero modes in graphene

Single impurity in monolayer graphene

Pristine graphene



Single impurity in infinite graphene

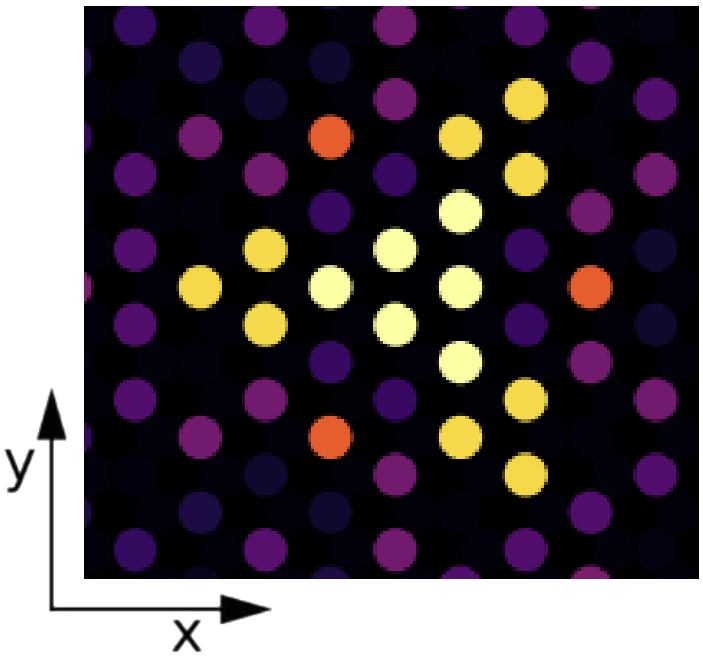


Phys. Rev. B 75, 125408 (2008)
Rep. Prog. Phys. 73 056501 (2010)

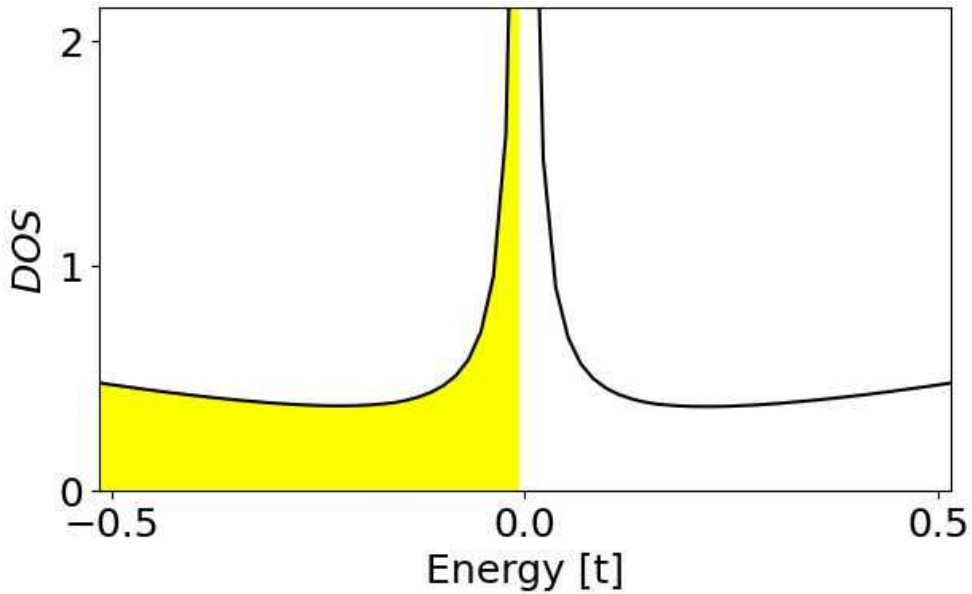
Hydrogen atoms create zero modes in graphene

Single impurity in monolayer graphene

Local density of states



Single impurity in infinite graphene

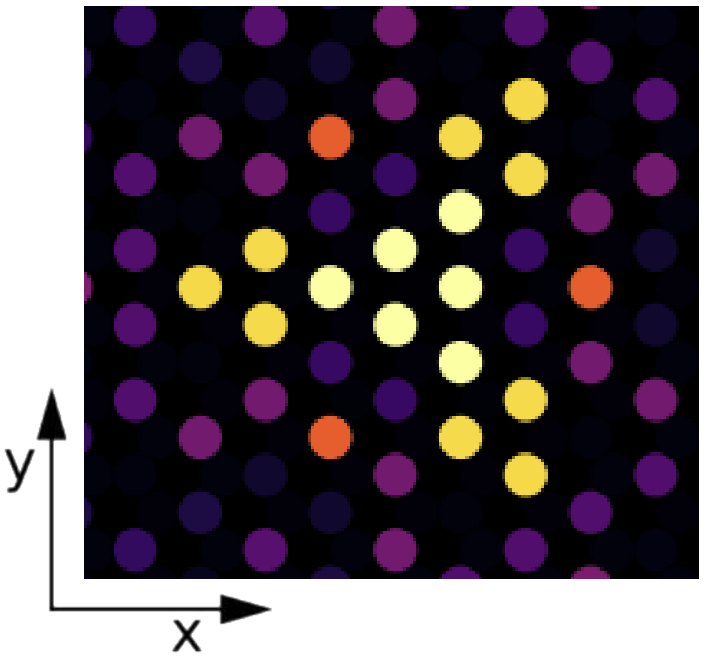


Phys. Rev. B 75, 125408 (2008)
Rep. Prog. Phys. 73 056501 (2010)

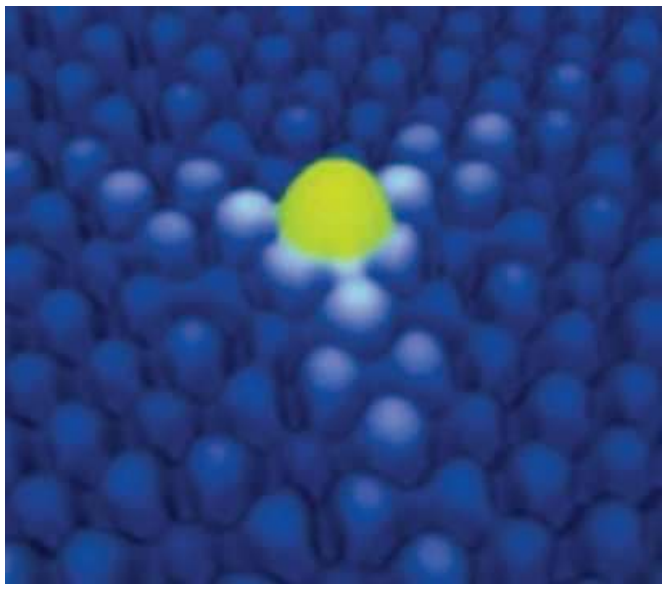
Hydrogen atoms create zero modes in graphene

Single impurity in monolayer graphene

Local density of states



Experimental local density of states

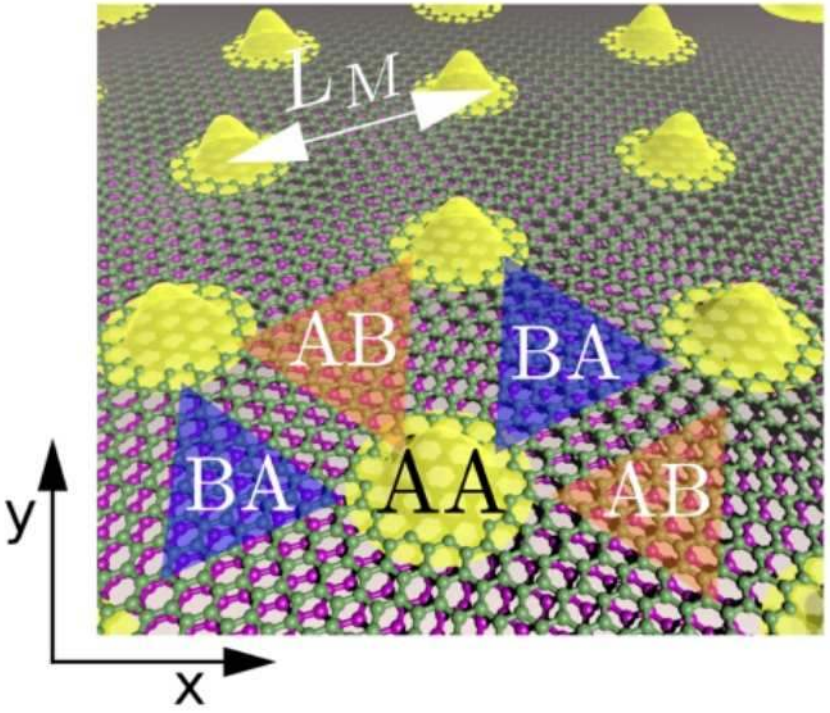


Science, 352(6284), 437-441 (2016)

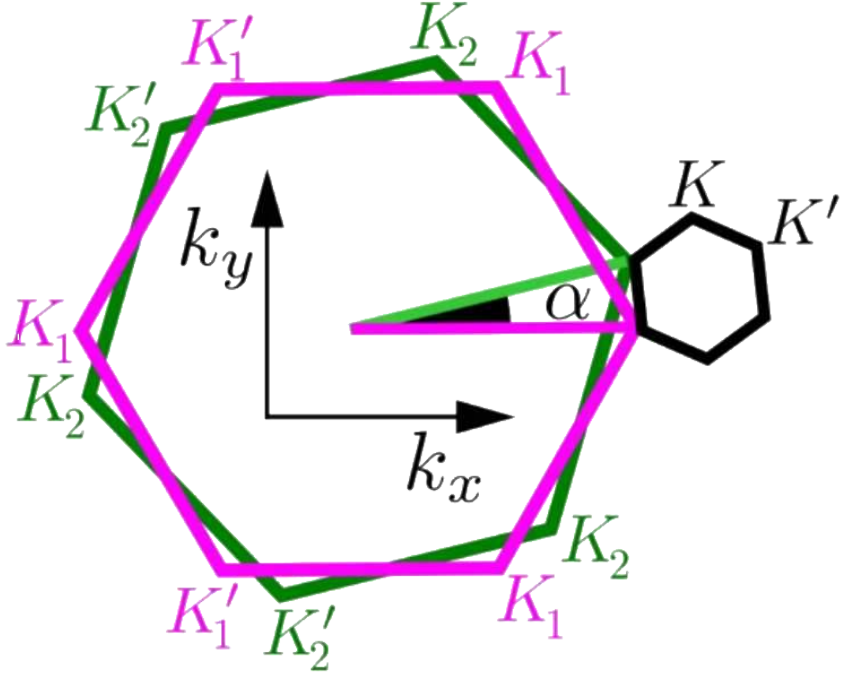
Hydrogen atoms create zero modes in graphene

Twisted graphene bilayers

Structure



Reciprocal space

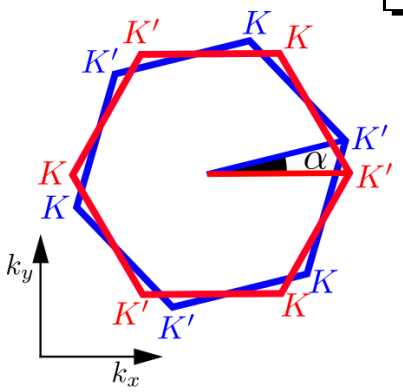


Uncoupled layers

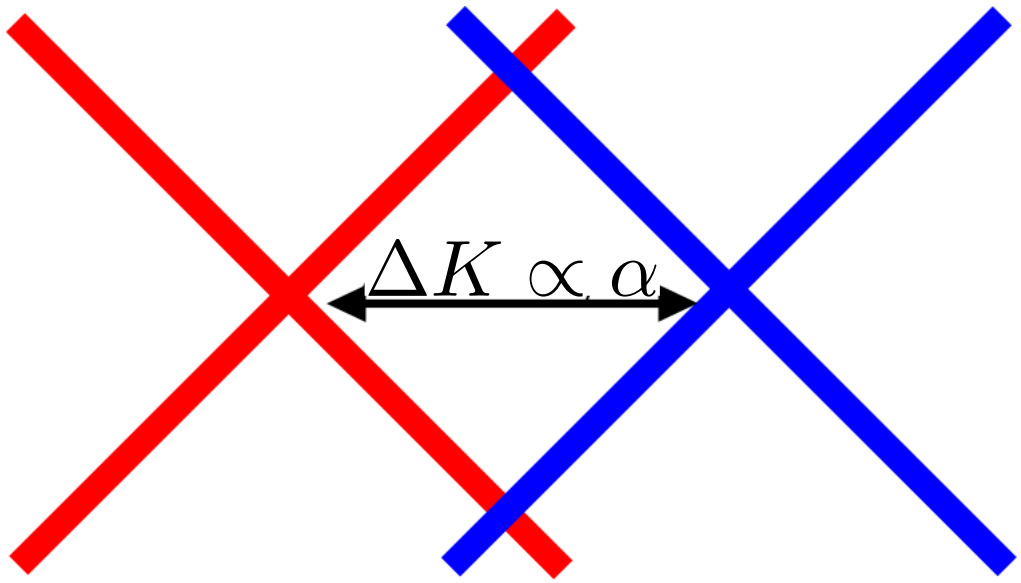
Dirac cone from layer #1

Momentum

Dirac cone from layer #2



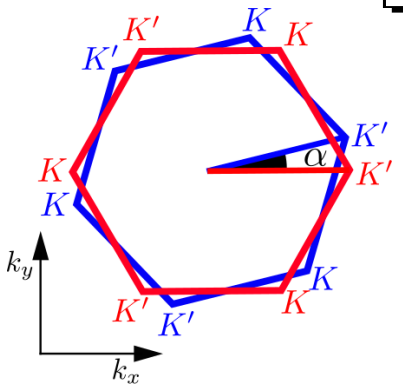
Energy

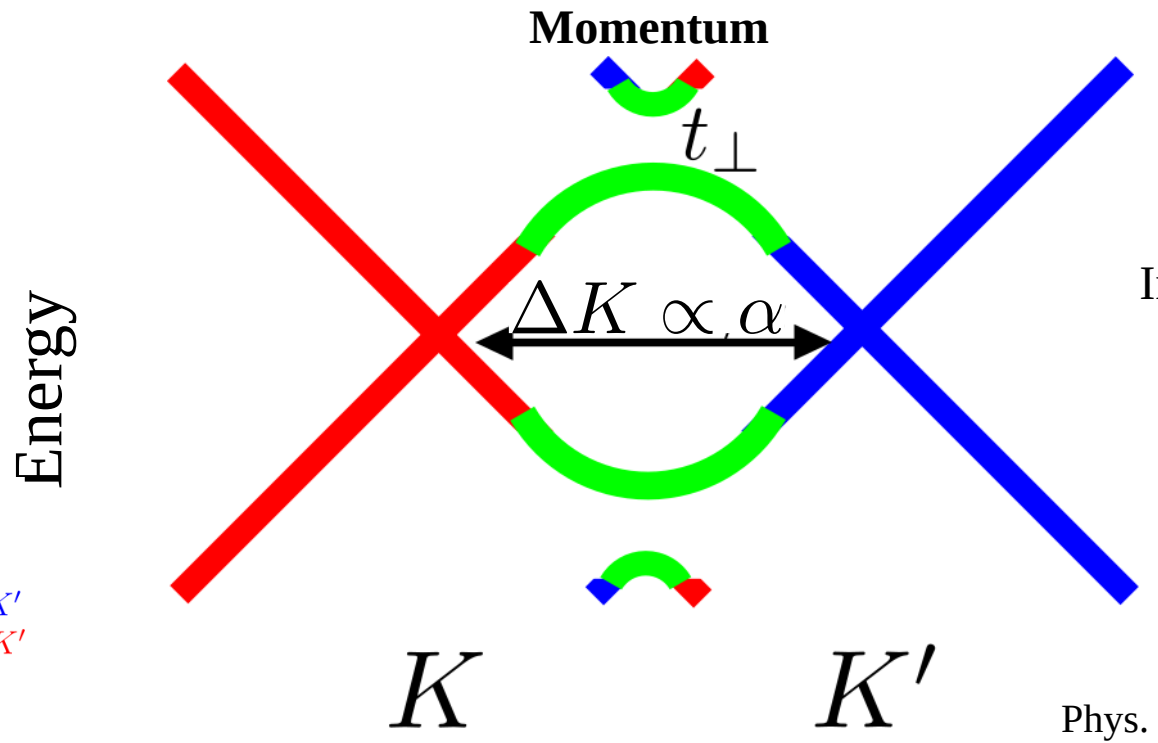


K K'

Phys. Rev. Lett. 99, 256802 (2007)
Phys. Rev. B 82, 121407(R) (2010)
PNAS 108 (30) 12233-12237 (2011)

Coupling the layers





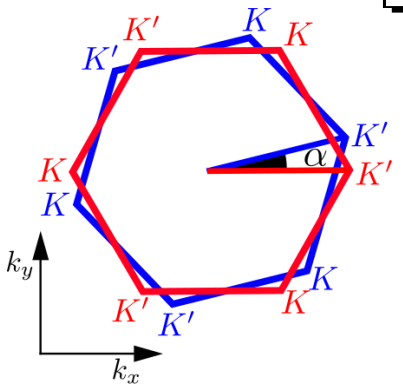
Relevant ratio

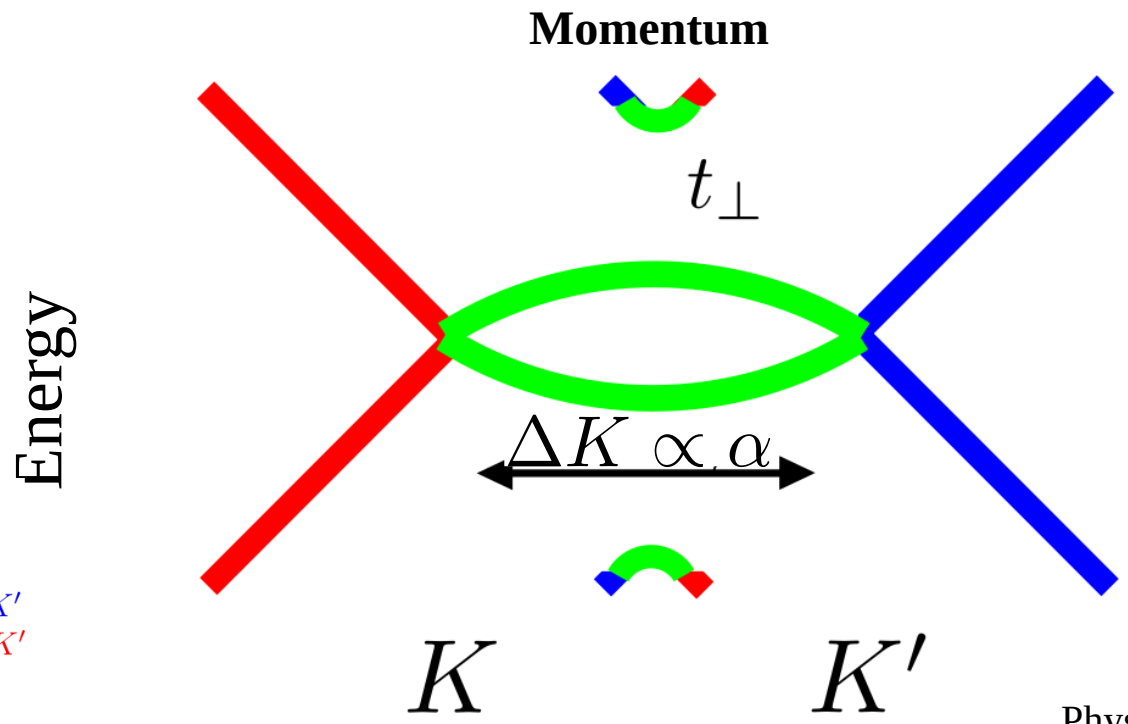
Inter-layer coupling $\dfrac{t_\perp}{\alpha}$

Twist angle

Phys. Rev. Lett. 99, 256802 (2007)
Phys. Rev. B 82, 121407(R) (2010)
PNAS 108 (30) 12233-12237 (2011)

Coupling the layers





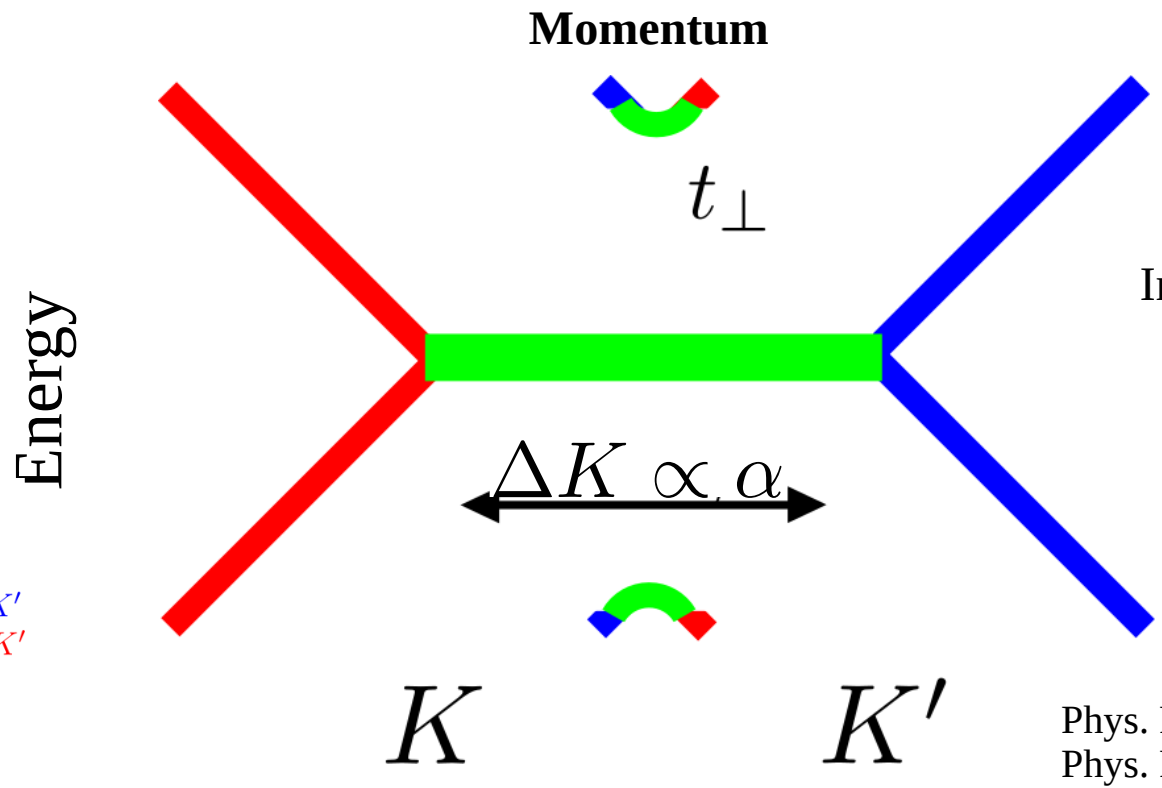
Relevant ratio

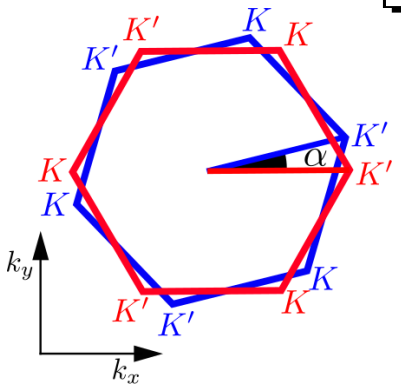
Inter-layer coupling $\dfrac{t_\perp}{\alpha}$

Twist angle

Phys. Rev. Lett. 99, 256802 (2007)
Phys. Rev. B 82, 121407(R) (2010)
PNAS 108 (30) 12233-12237 (2011)

Coupling the layers





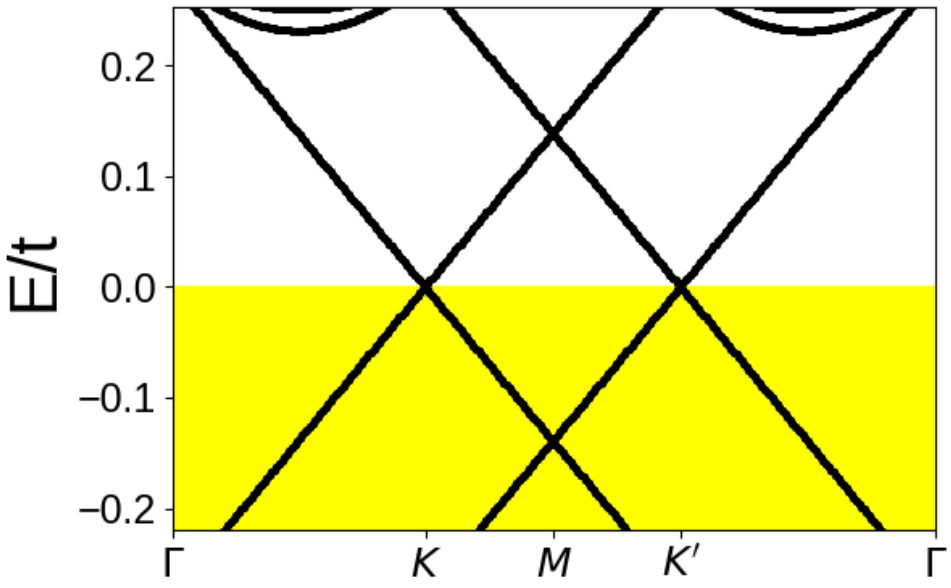
Relevant ratio

Inter-layer coupling $\dfrac{t_\perp}{\alpha}$ Twist angle

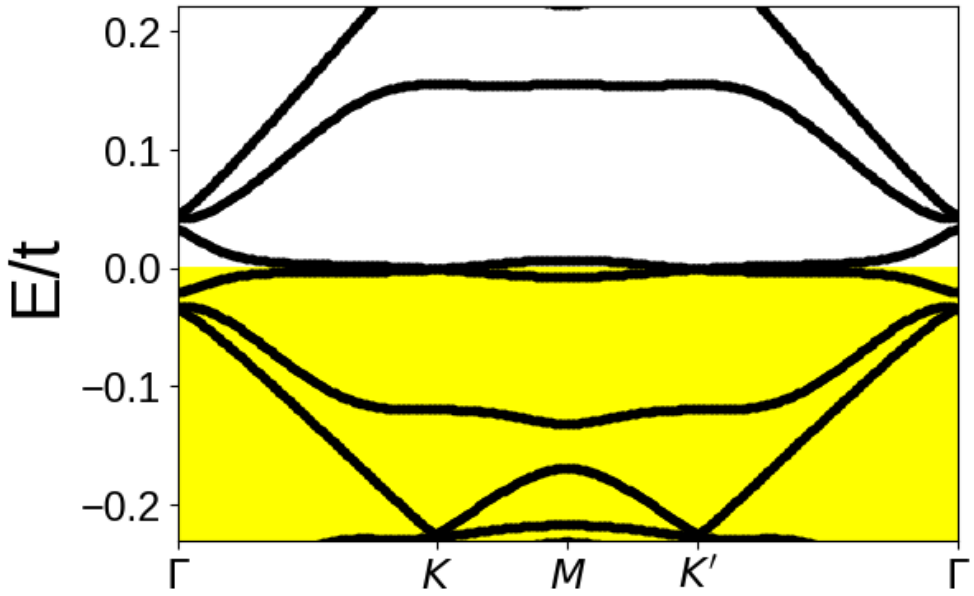
Phys. Rev. Lett. 99, 256802 (2007)
Phys. Rev. B 82, 121407(R) (2010)
PNAS 108 (30) 12233-12237 (2011)

Electronic structure of twisted bilayer

Uncoupled layers



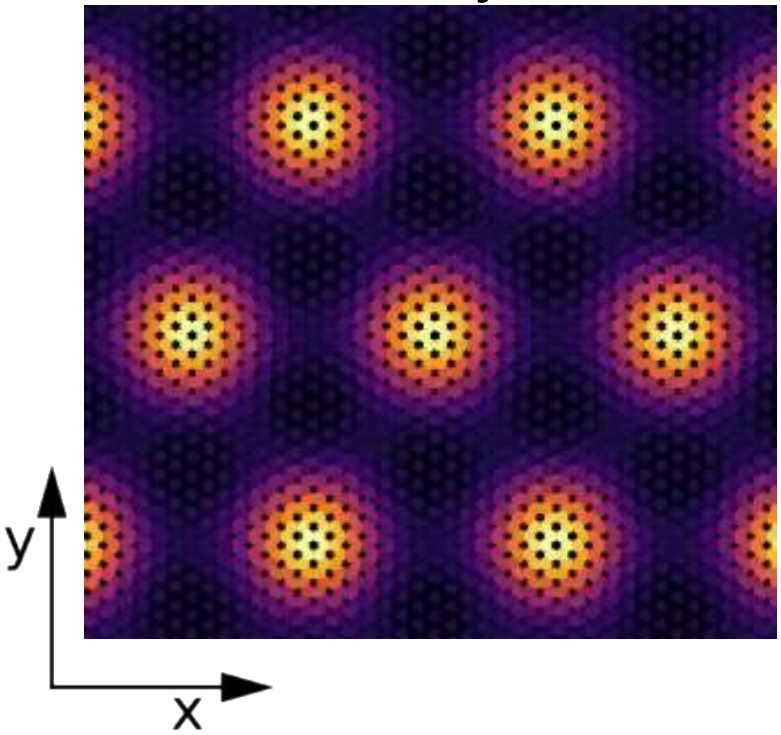
Coupled layers



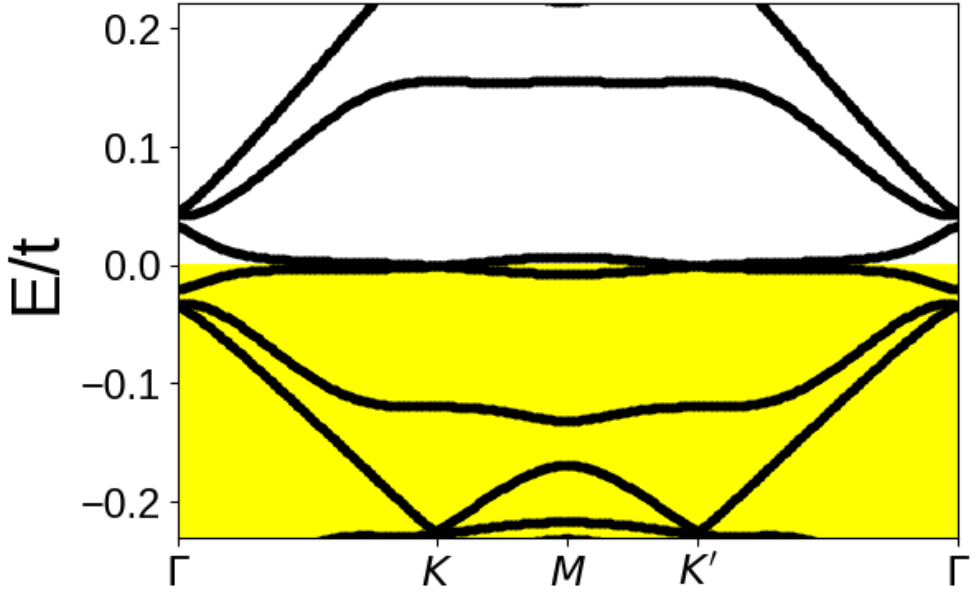
Twisted graphene bilayers feature flat bands driven by the twist

Electronic structure of twisted bilayer

Local density of states



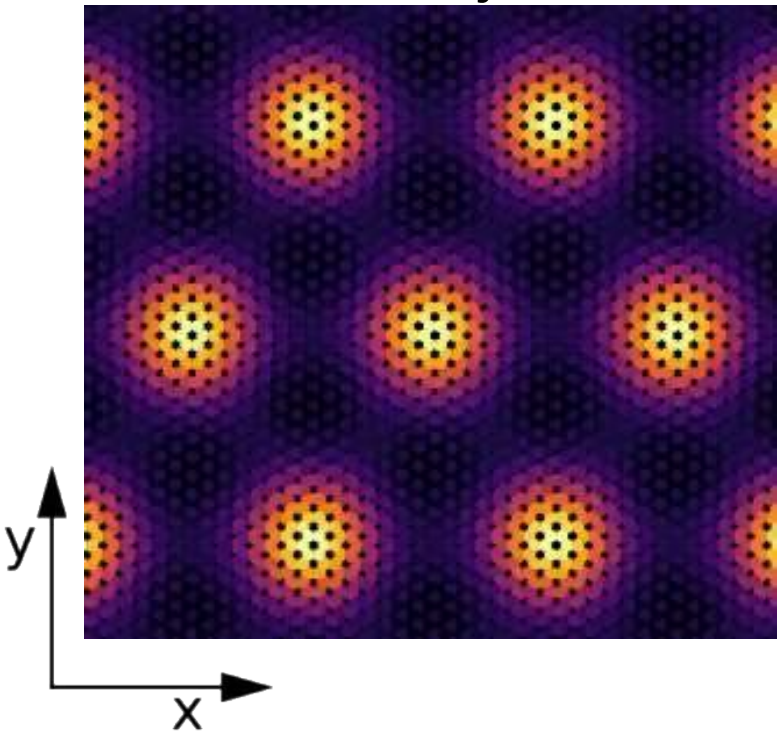
Coupled layers



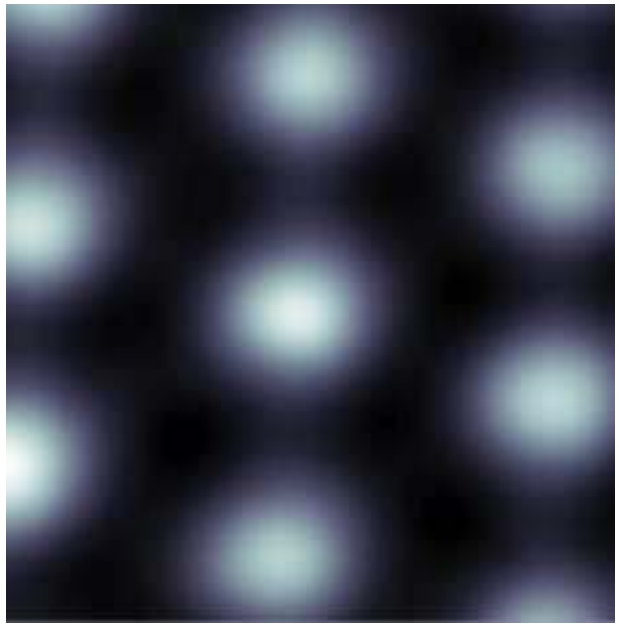
How do these moire localized modes interact with local impurities?

Electronic structure of twisted bilayer

Local density of states



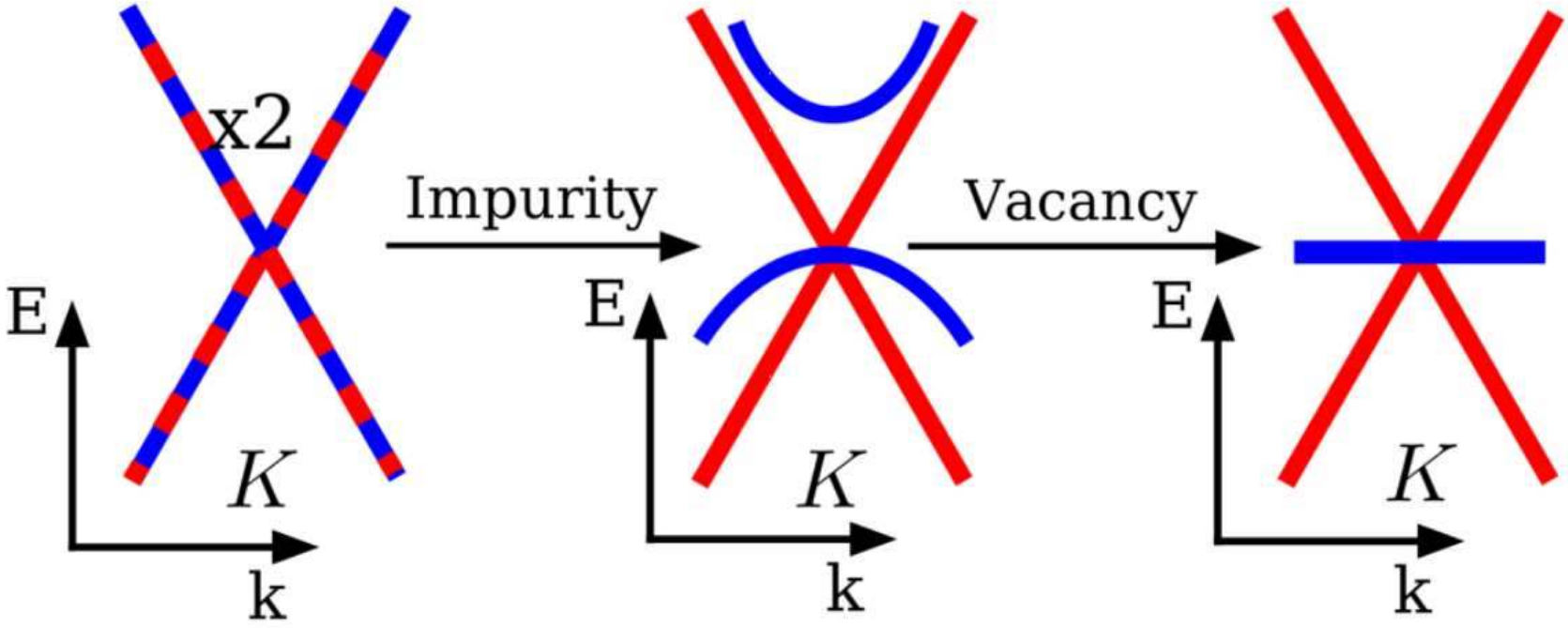
Experimental local density of states



Nature Physics 6, 109–113 (2010)
Nature 572, 101–105 (2019)
Nature 572, 95–100 (2019)

How do these moire localized modes interact with local impurities?

Impact of single impurities in the twisted bilayer electronic structure



Single impurities give rise to a triple point crossing in the electronic structure

Model for impurities in twisted multilayers

Full Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{W}$$

Pristine twisted bilayer graphene

$$\mathcal{H}_0 = t \sum_{\langle i,j \rangle} c_i^\dagger c_j + \sum_{i,j} \bar{t}_\perp(\mathbf{r}_i, \mathbf{r}_j) c_i^\dagger c_j$$

Intra-layer hopping

Inter-layer hopping

Local impurity

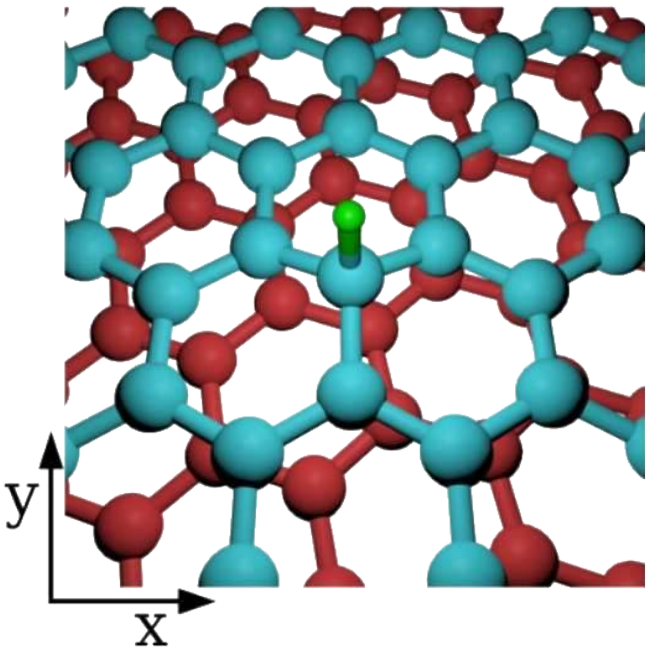
$$\mathcal{W} = w c_n^\dagger c_n$$

The limit of infinite potential corresponds to removing a site

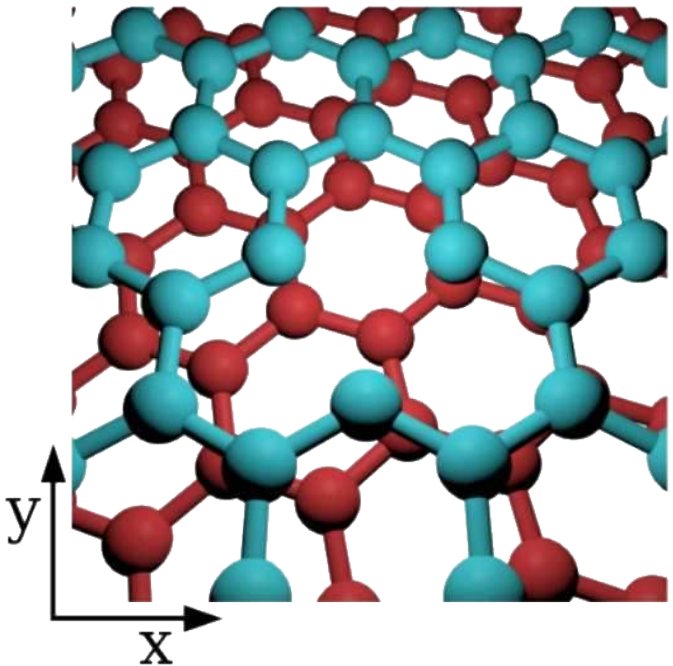
$$w \to \infty$$

The strong impurity limit

Adsorbed hydrogen in TBG



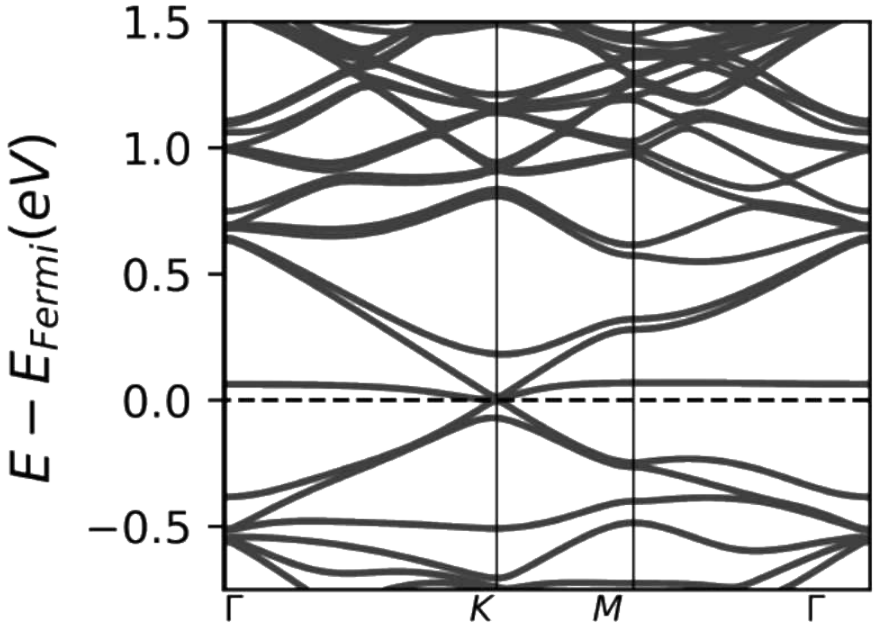
Effective tight binding model



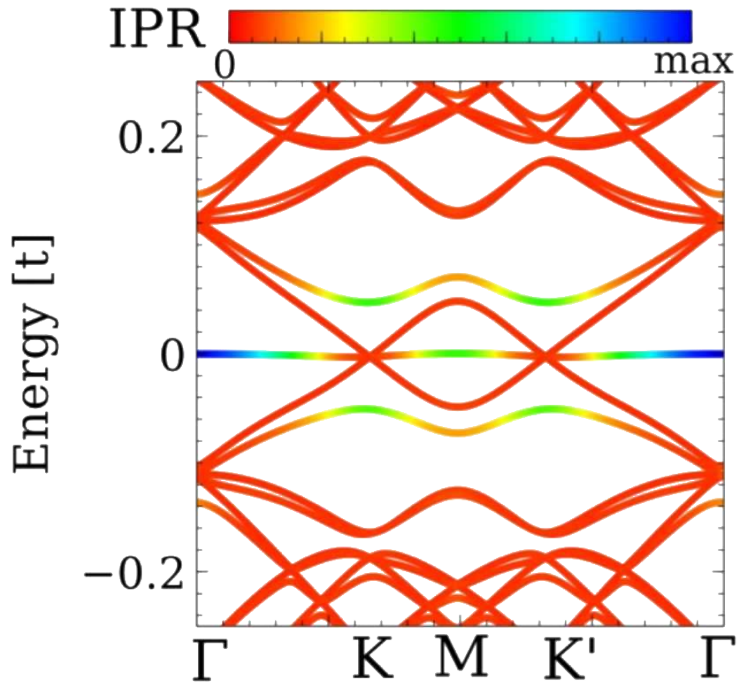
Hydrogen adsorption effectively gives rise to a vacancy in the low energy model

DFT versus tight binding

Density functional theory



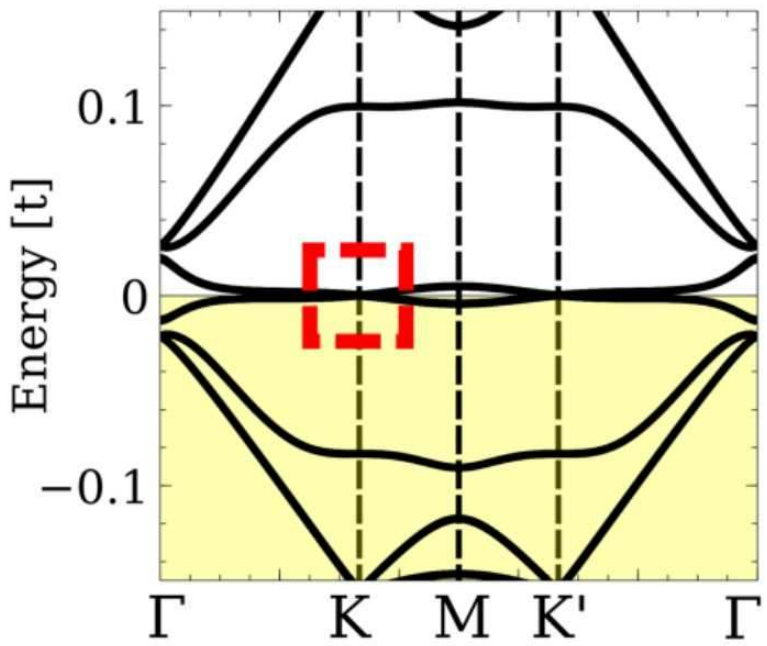
Tight binding



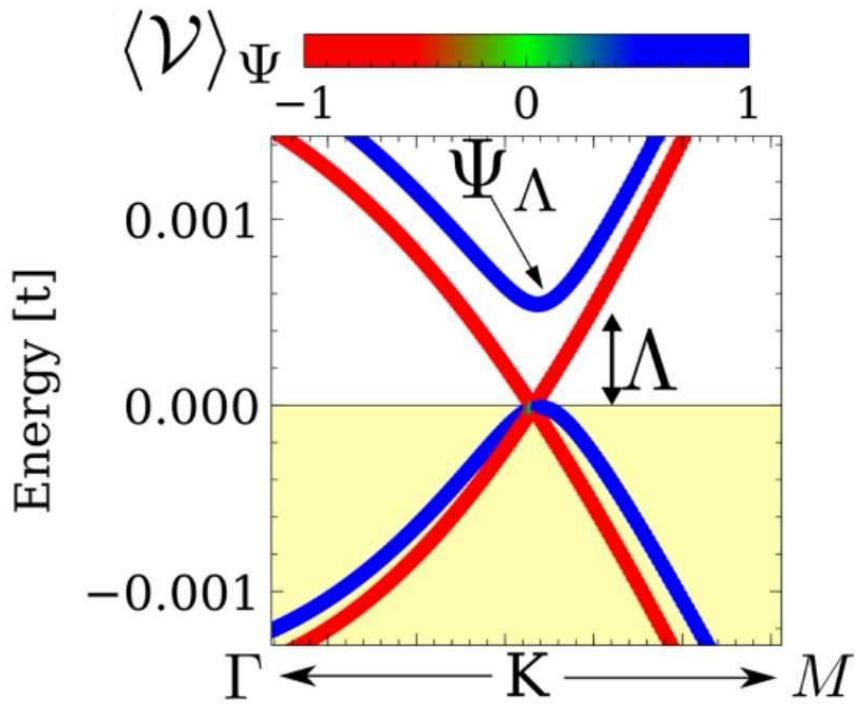
DFT and tight binding give rise to comparable electronic structures

Impact of a single weak defect

Band structure



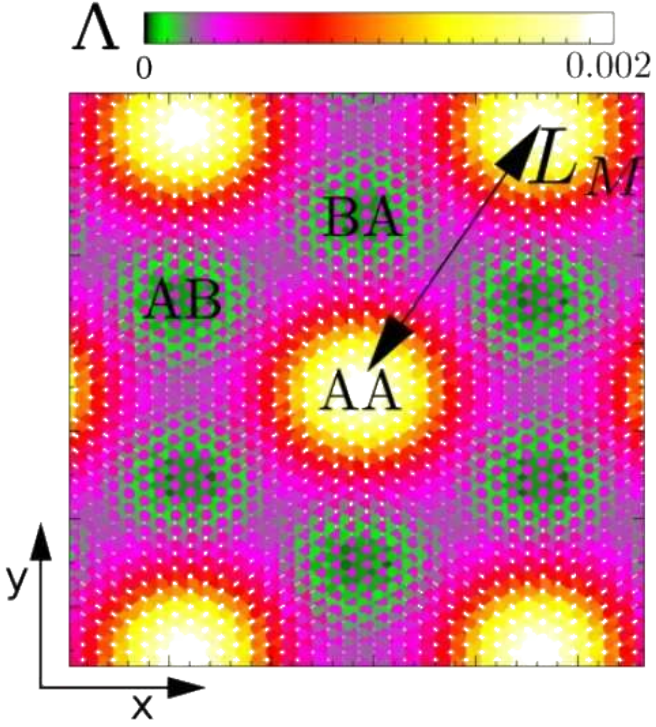
Valley expectation value



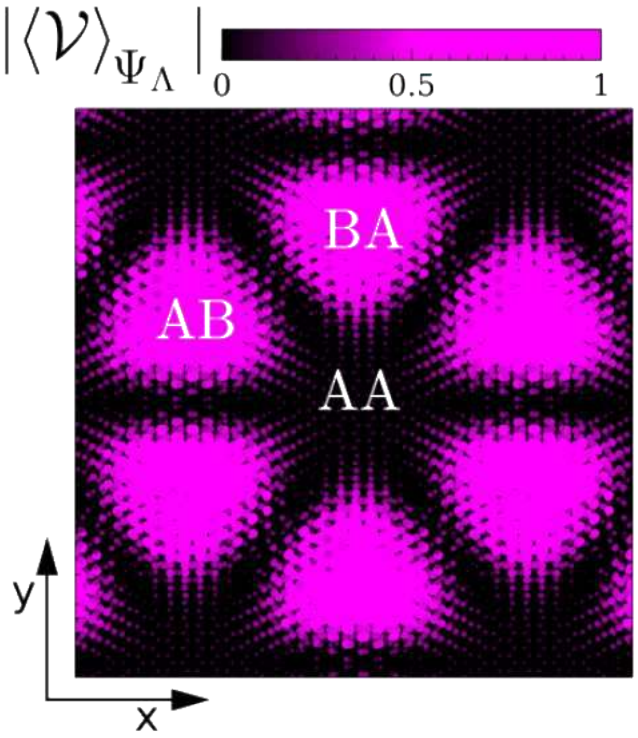
A weak impurity generates a triple band crossing at the Dirac point

Position-dependent valley mixing

Gap opening depending on the location



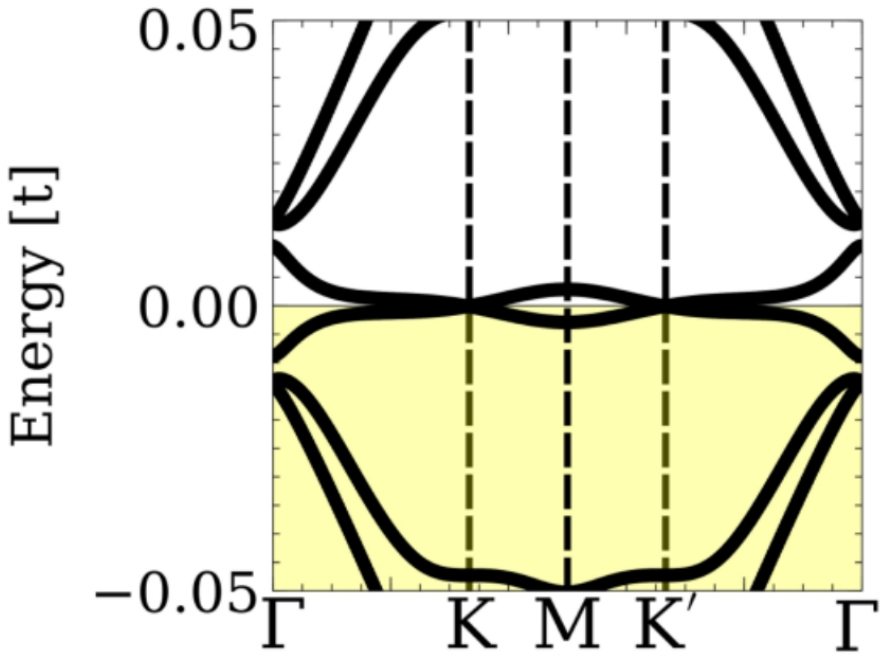
Valley polarization depending on the location



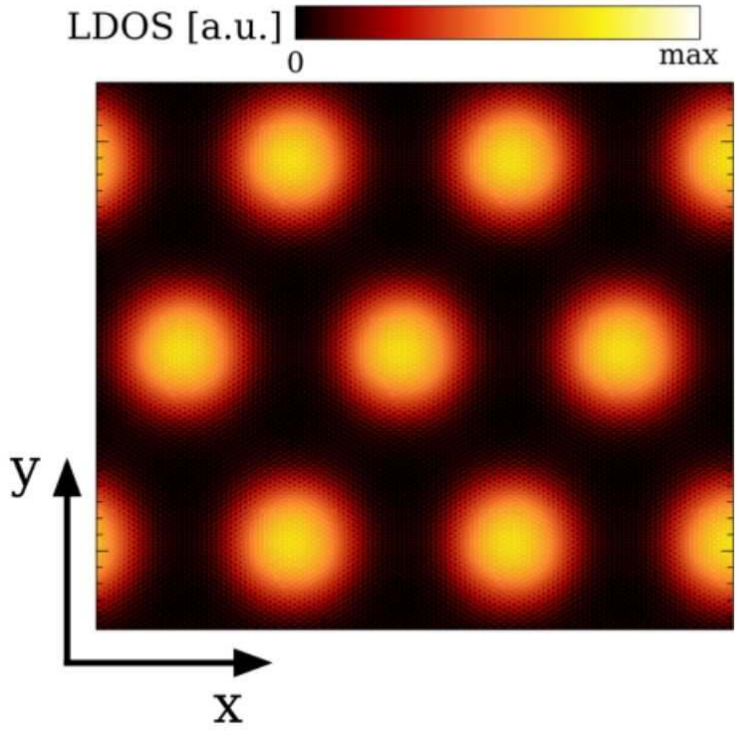
The location of the impurities determines the strength of the valley mixing

Low energy states, pristine bilayer

Band structure



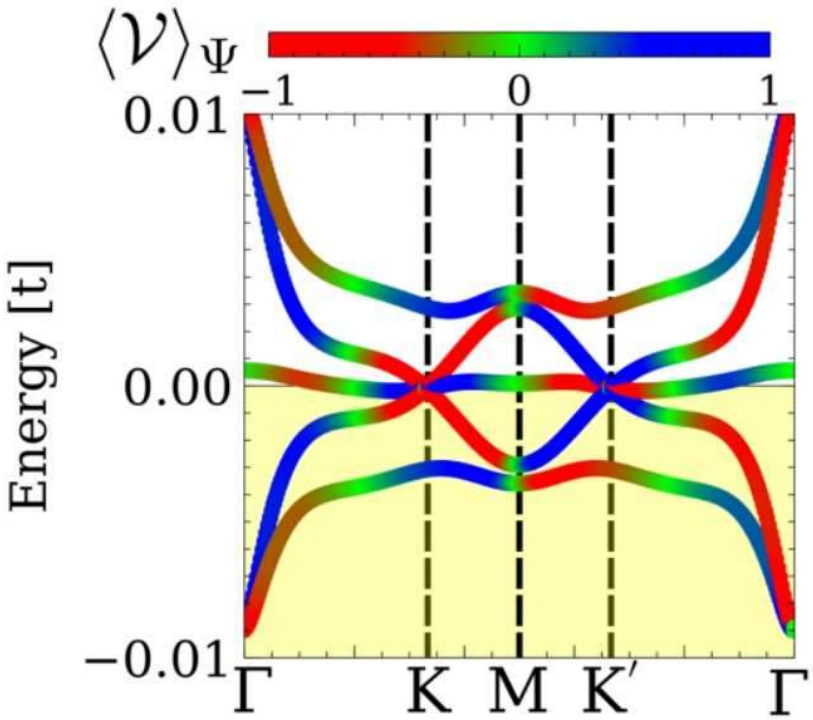
Spatial distribution of the low energy states



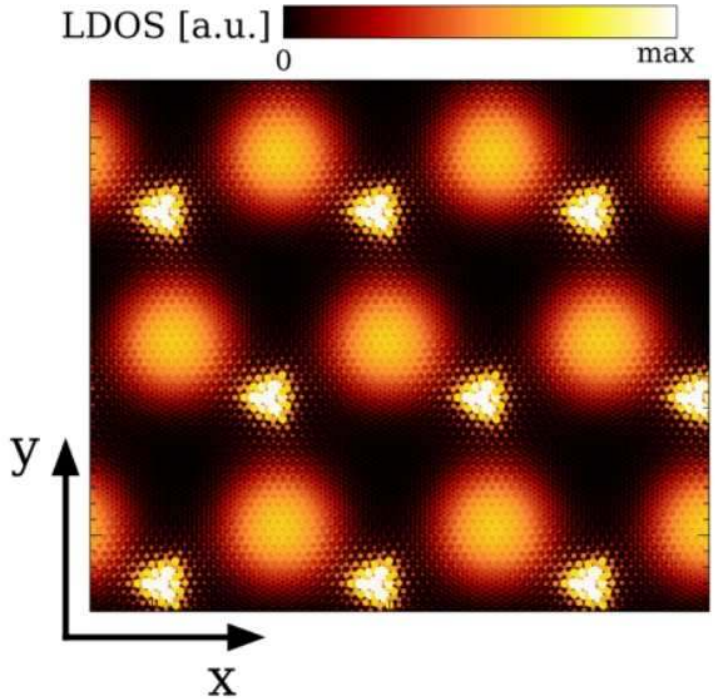
The nearly flat bands create an emergent triangular superlattice

Low energy states, strong single defect

Band structure



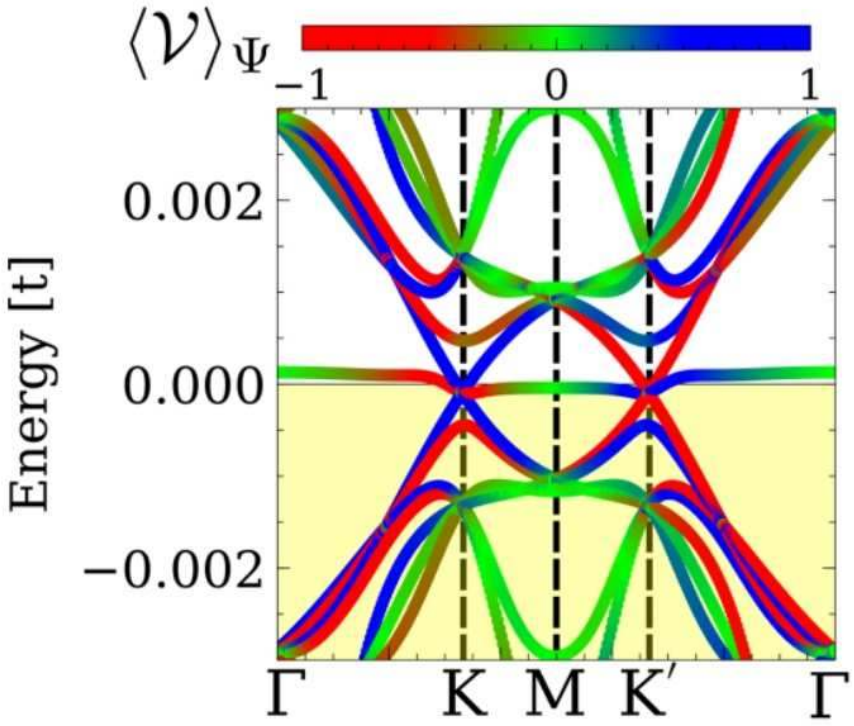
Spatial distribution of the low energy states



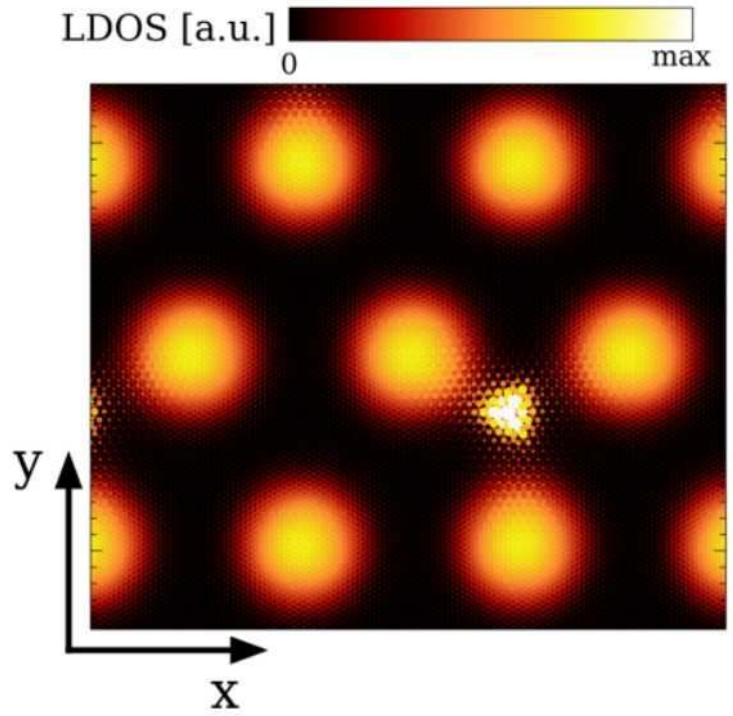
The nearly flat bands coexist with a localized vacancy state

Low energy states, strong single defect in a 4x4 supercell

Band structure



Spatial distribution of the low energy states



The nearly flat bands coexist with a localized vacancy state

Including interactions in a minimal way

We will include interactions with a minimal Hubbard repulsion

$$H_U = \sum_i U c^\dagger_{i,\uparrow} c_{i,\uparrow} c^\dagger_{i,\downarrow} c_{i,\downarrow} = \sum_i U n_{i,\uparrow} n_{i,\downarrow}$$

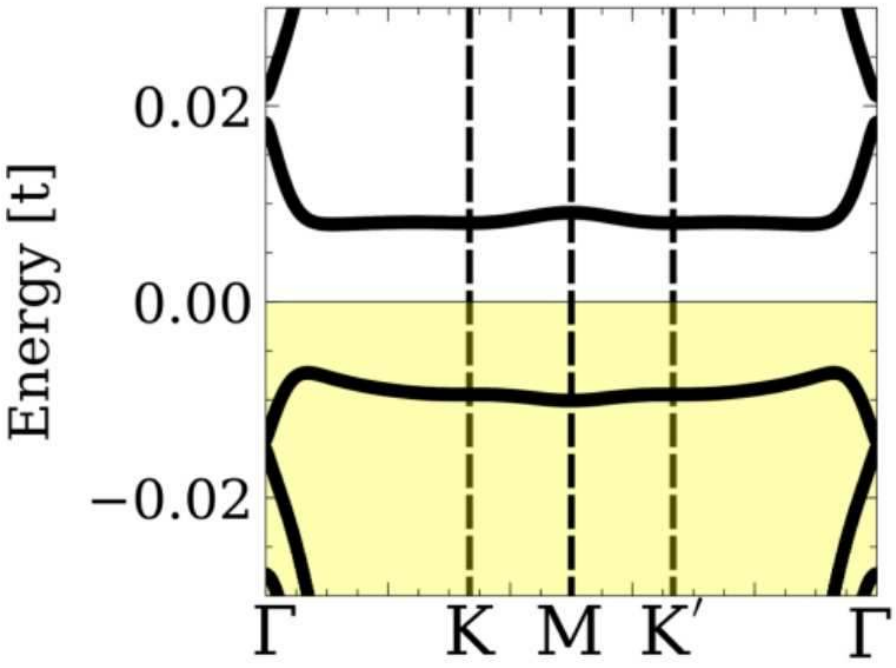
Solved at the mean-field level

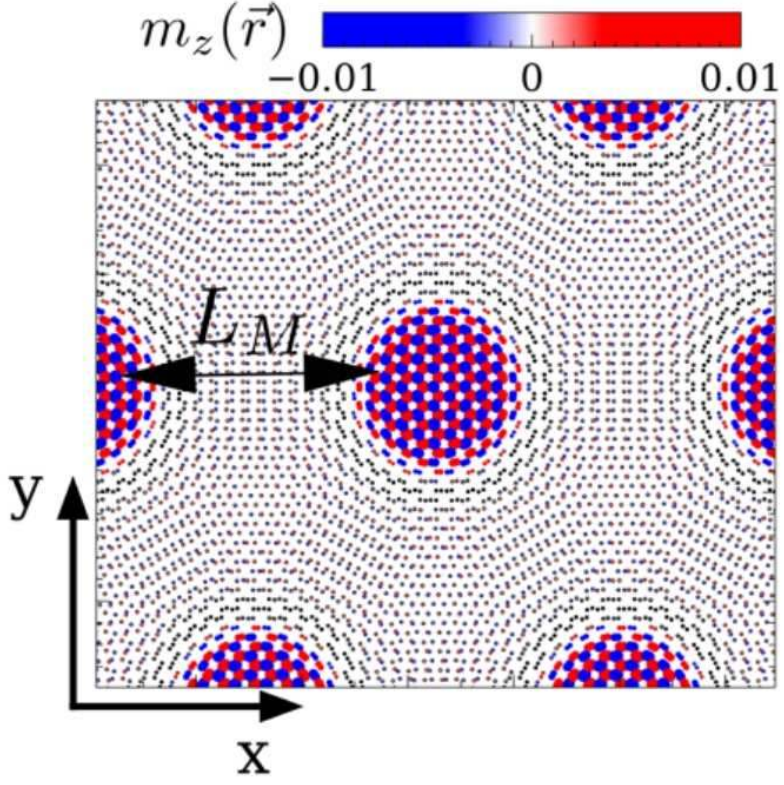
$$H_U^{\mathrm{MF}} = \sum_i U [\langle n_{i,\uparrow} \rangle n_{i,\downarrow} + \langle n_{i,\downarrow} \rangle n_{i,\uparrow} - \langle n_{i,\uparrow} \rangle \langle n_{i,\downarrow} \rangle]$$

Let us see the impact on the low energy states

Impact of interactions, pristine system

Band structure

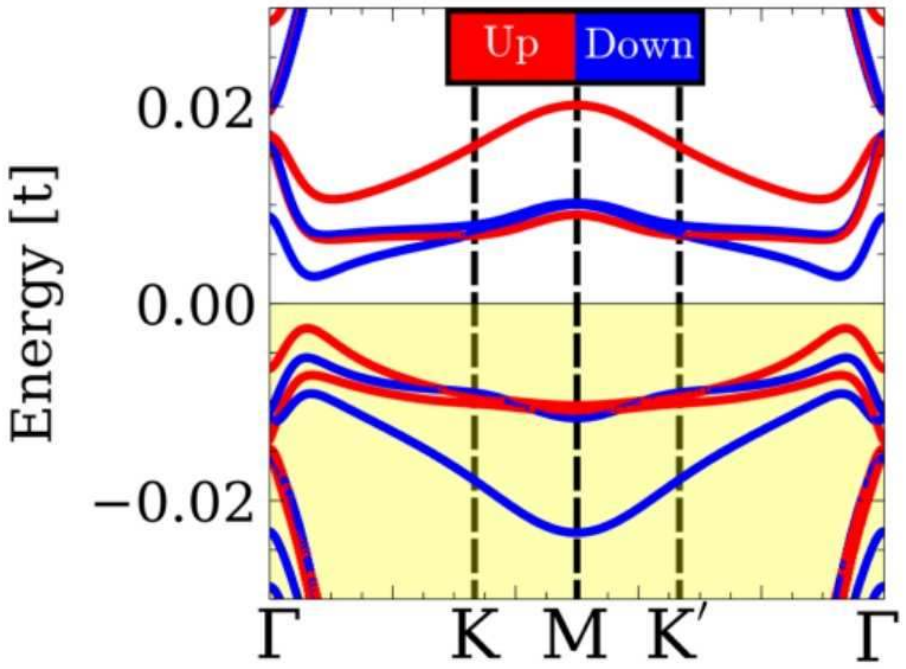


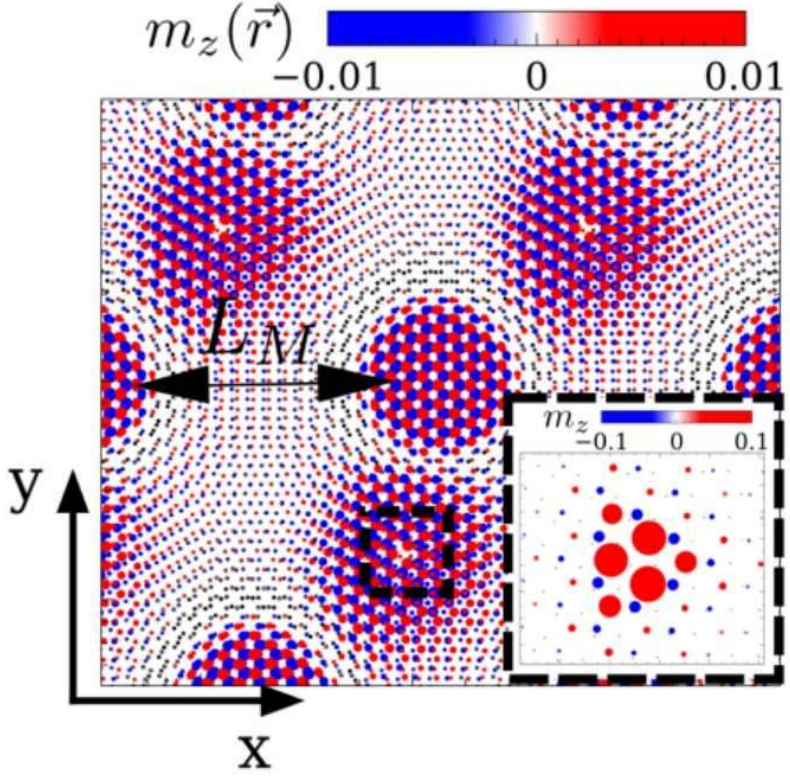


Gap opening at charge neutrality

Impact of interactions, single vacancy at charge neutrality

Band structure

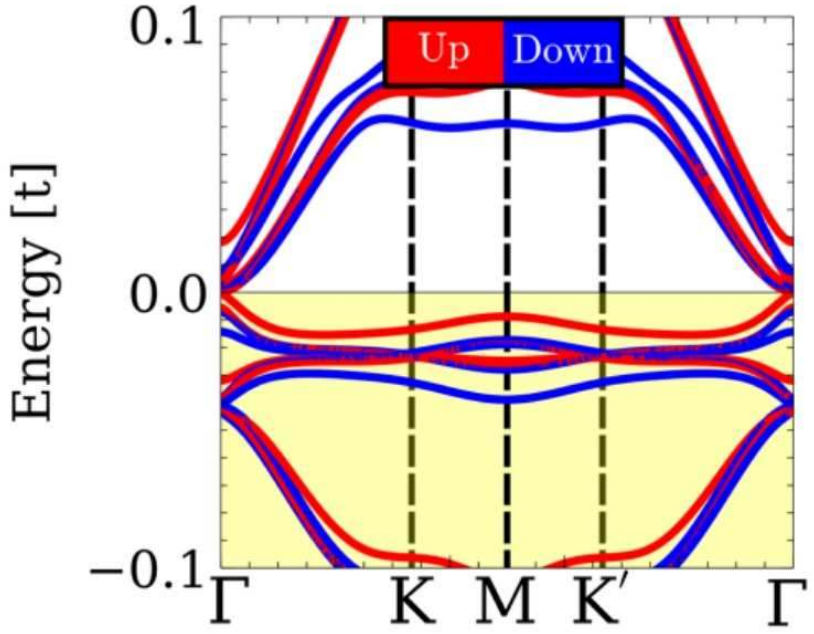


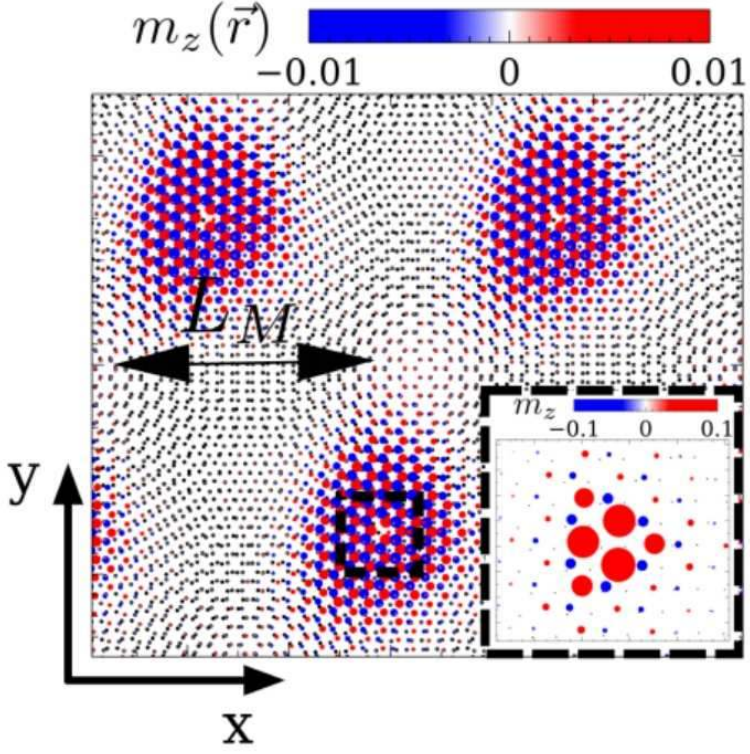


Interaction-driven symmetry breaking in the impurity and flat bands coexists

Impact of interactions, single vacancy at 4e per moire unit cell

Band structure

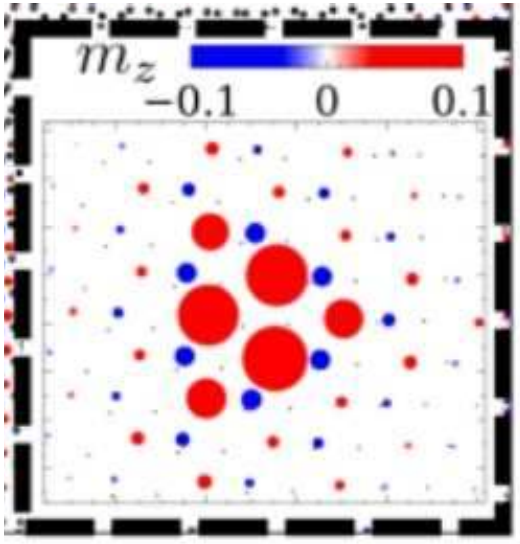




The vacancy magnetism survives full filling of the original twisted bilayer flat bands

Interplay between impurity magnetism and twisted bilayer superconductivity

The impurity induced moment survives doping of the flat bands



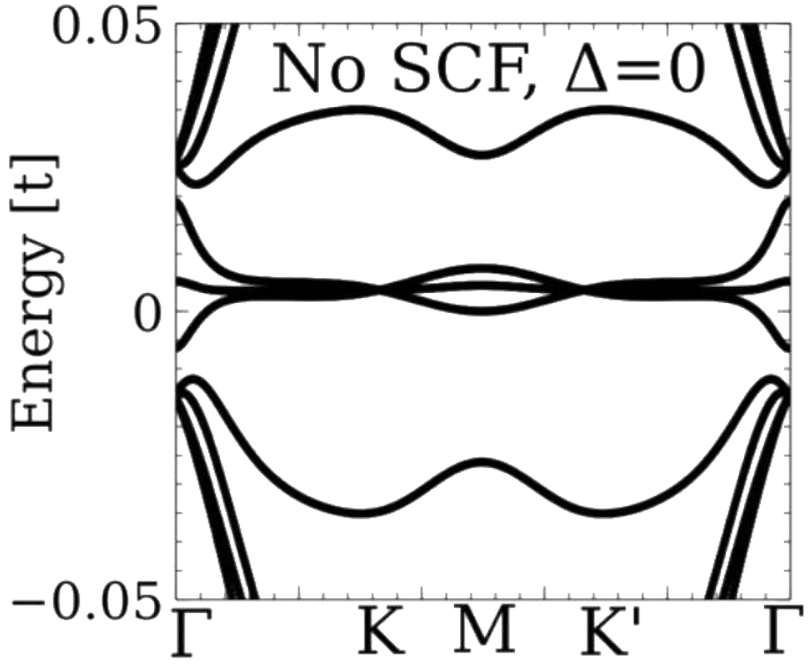
What happens when its magnetism coexists with the twisted bilayer superconducting state?

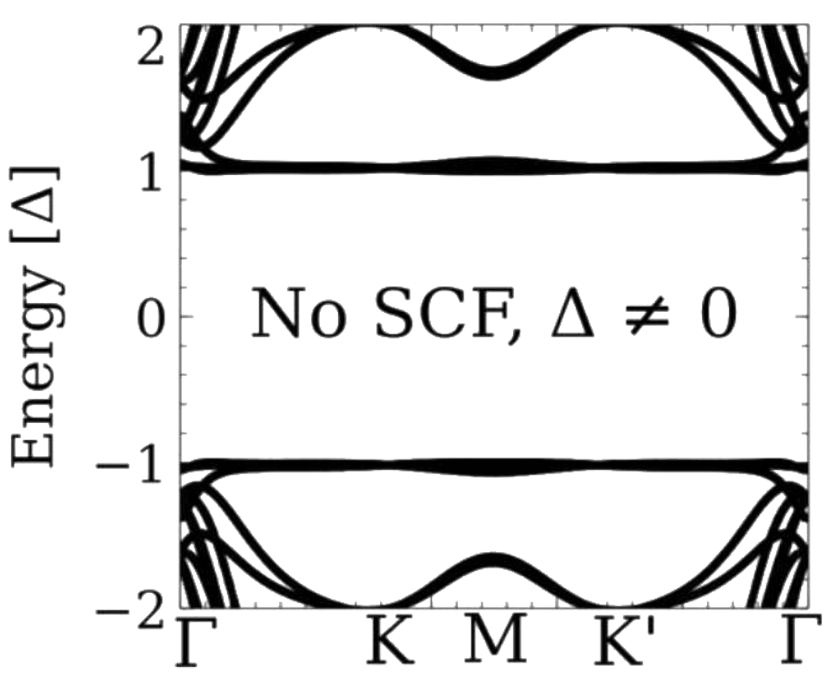
We will now include a superconducting state

$$H_{\mathrm{SC}} = \sum_i \Delta_i [c_{i,\uparrow} c_{i,\downarrow} + c^{\dagger}_{i,\downarrow} c^{\dagger}_{i,\uparrow}]$$

Working in the regime of (close to) 2 holes per moire unit cell

Electronic structure without including interactions

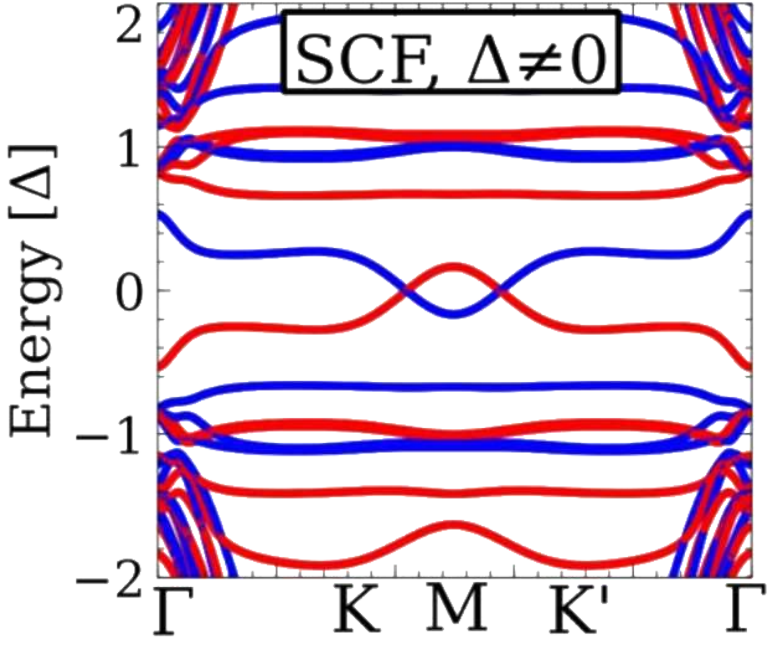




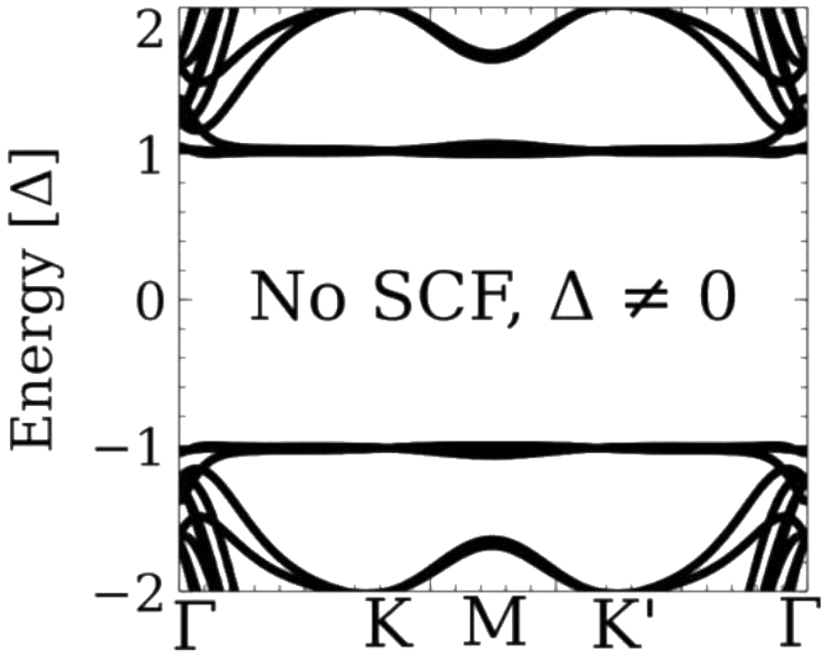
s-wave superconductivity opens up a gap in the electronic spectra

Electronic structure including repulsive interactions

With interaction-induced magnetism



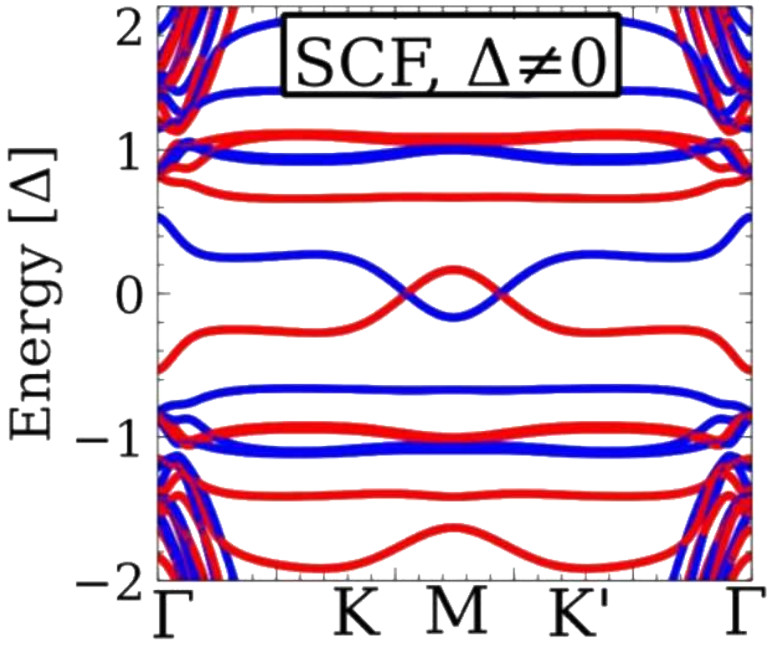
Without interaction-induced magnetism



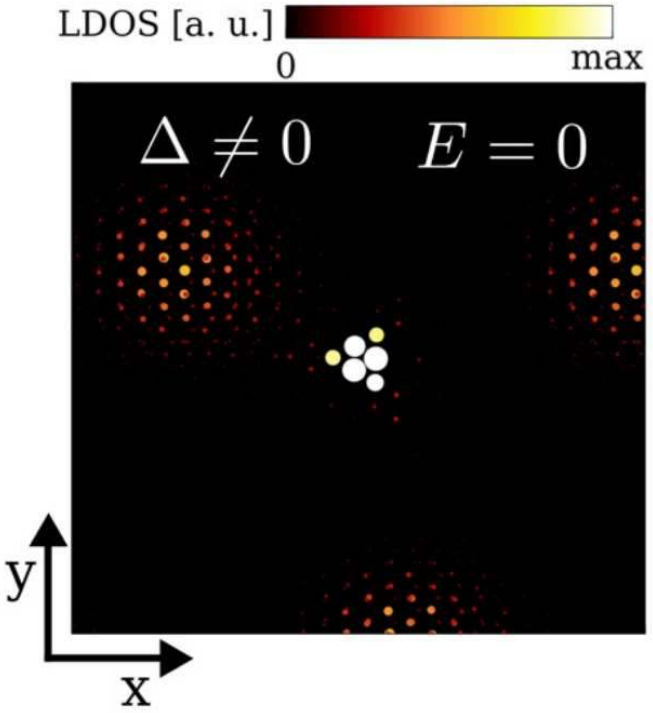
Impurity-induced magnetism and s-wave superconductivity gives rise to in-gap Yu-Shiba-Rusinov states

Electronic structure including repulsive interactions

With interaction-induced magnetism



Spatial distribution of the in-gap YSR states

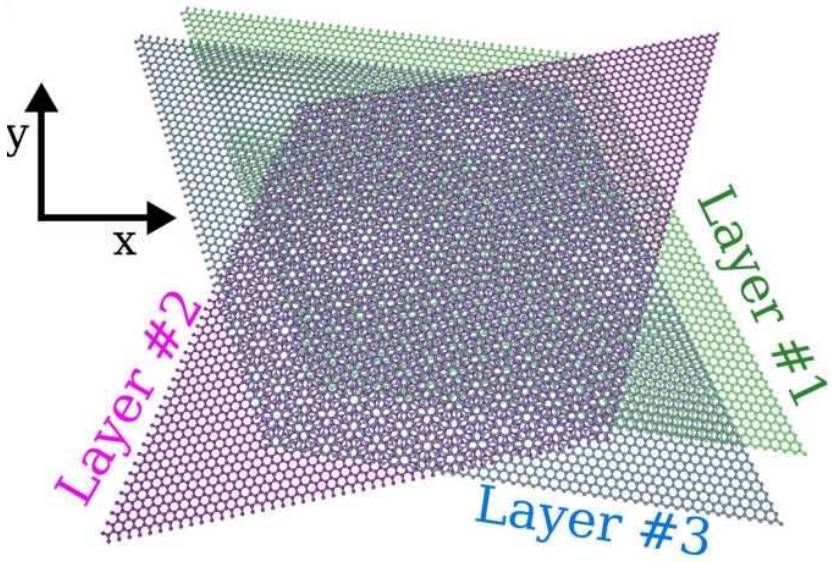


In-gap modes appear around the interaction-induced impurity magnetic moment

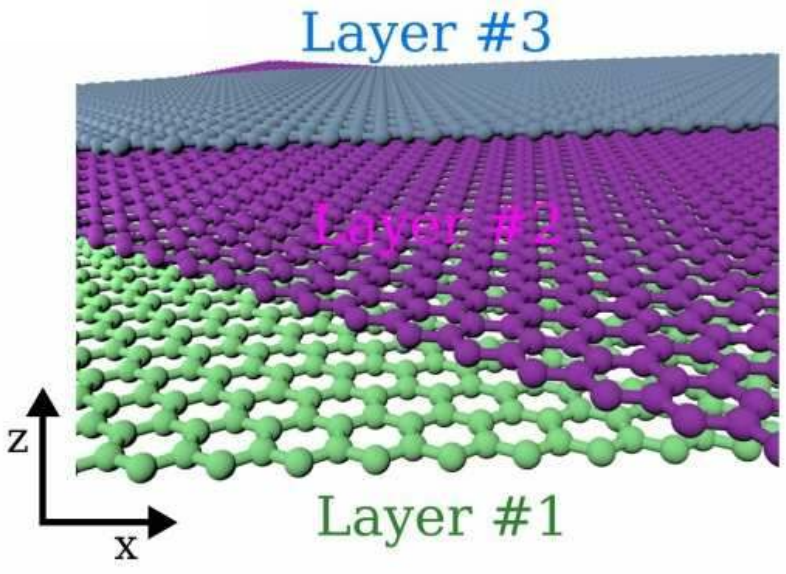
Impurities in twisted trilayer graphene

Structure of mirror-symmetric twisted graphene trilayer

Top view

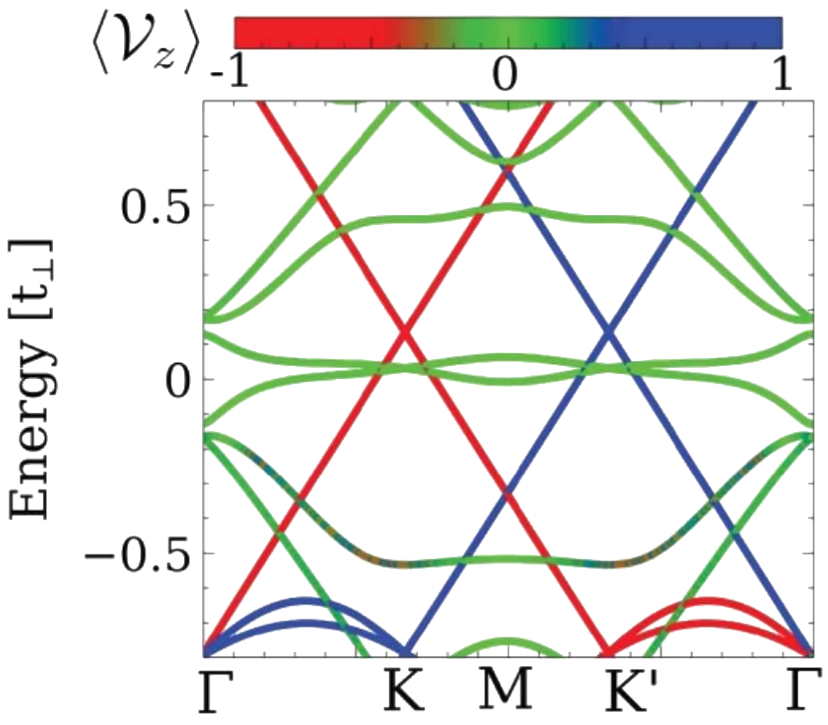


Lateral view

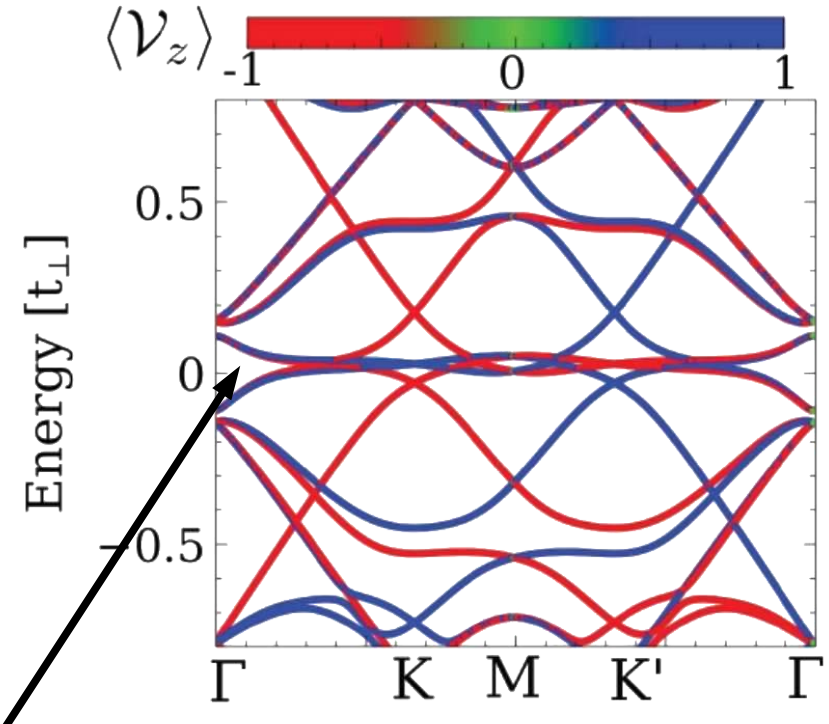


Electronic structure of twisted trilayer

Without an interlayer bias



With an interlayer bias



An interlayer bias creates internal topological valley currents

Internal topological valley currents in a twisted trilayer

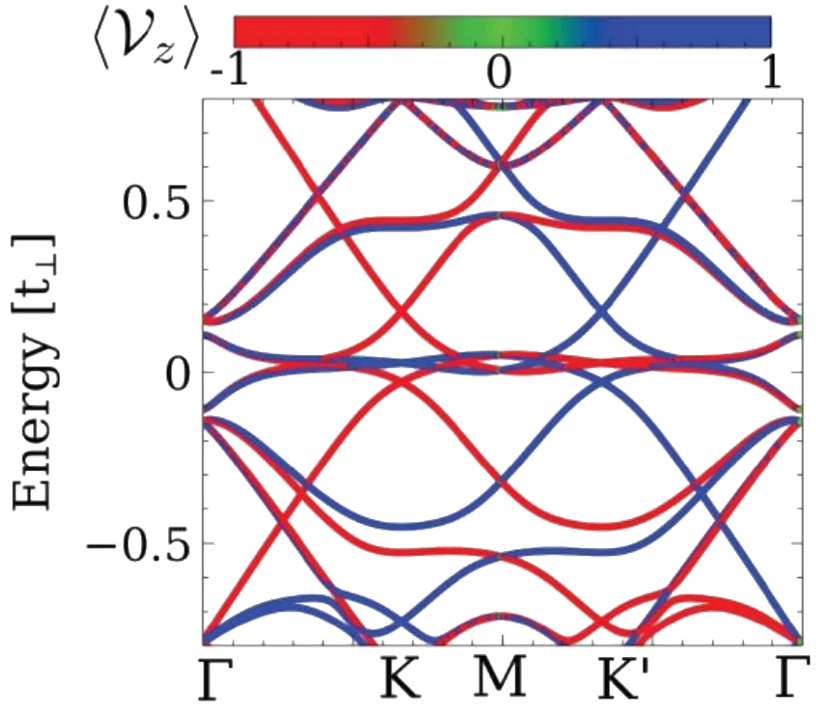
The spatially resolved valley current is defined from the valley Chern number

$$C_V = C_K - C_{K'} = \int \chi_V(\mathbf{r}, \omega) d^2\mathbf{r}\, d\omega$$

Spatially and frequency resolved valley Chern density

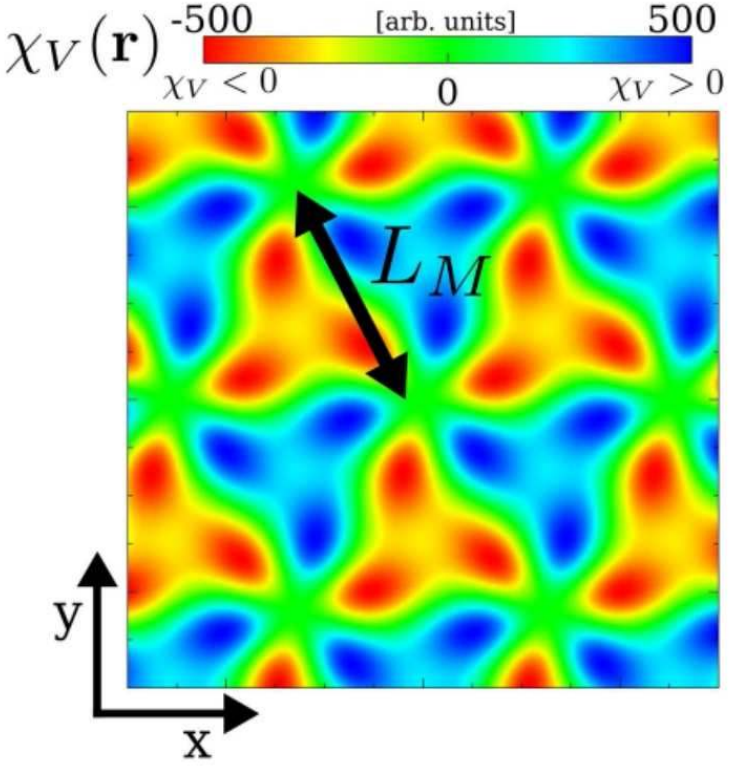
$$\chi_V(\mathbf{r}, \omega)$$

It represents bulk valley currents

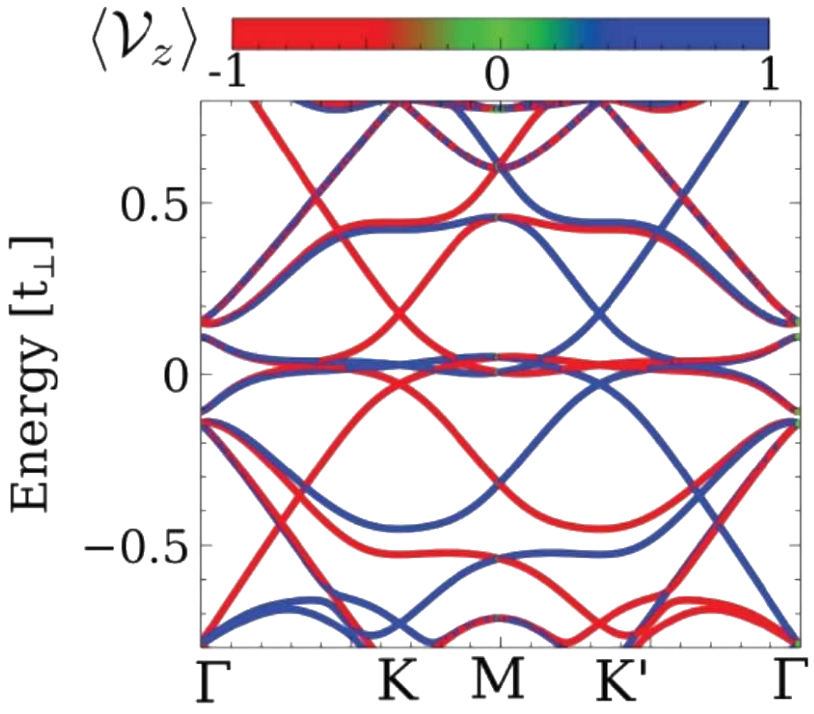


Internal topological valley currents in a twisted trilayer

Bulk valley current



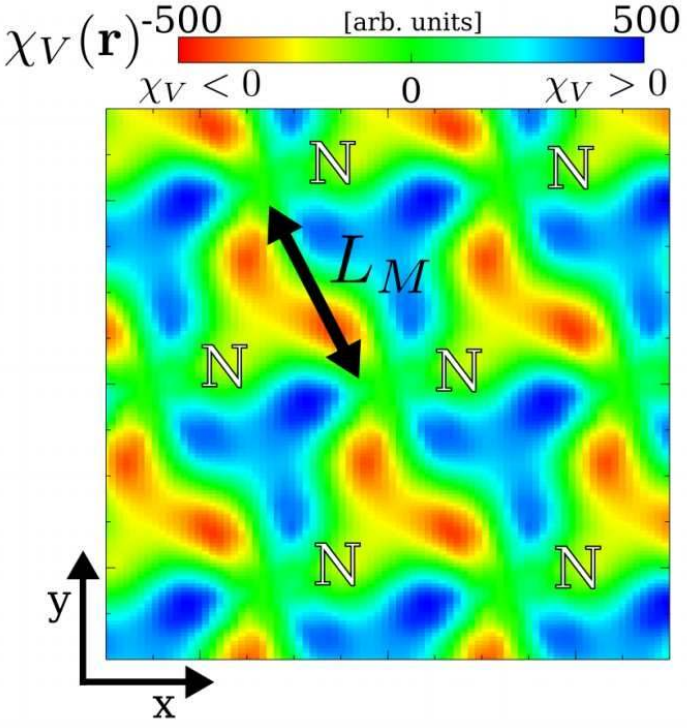
Band structure



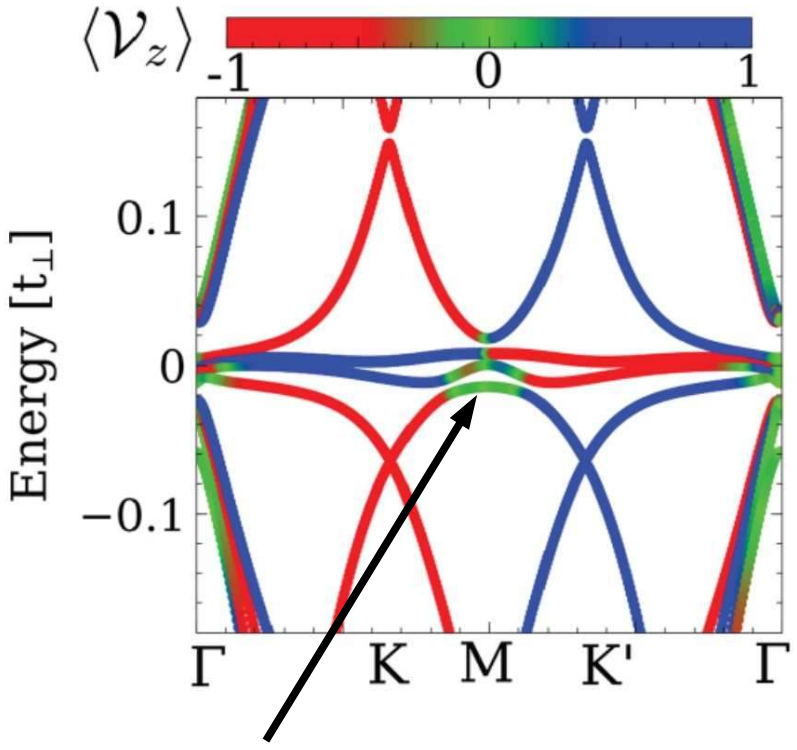
Internal topological valley currents emerge in the moire supercell

Weak impurity in a twisted graphene trilayer

Bulk valley current



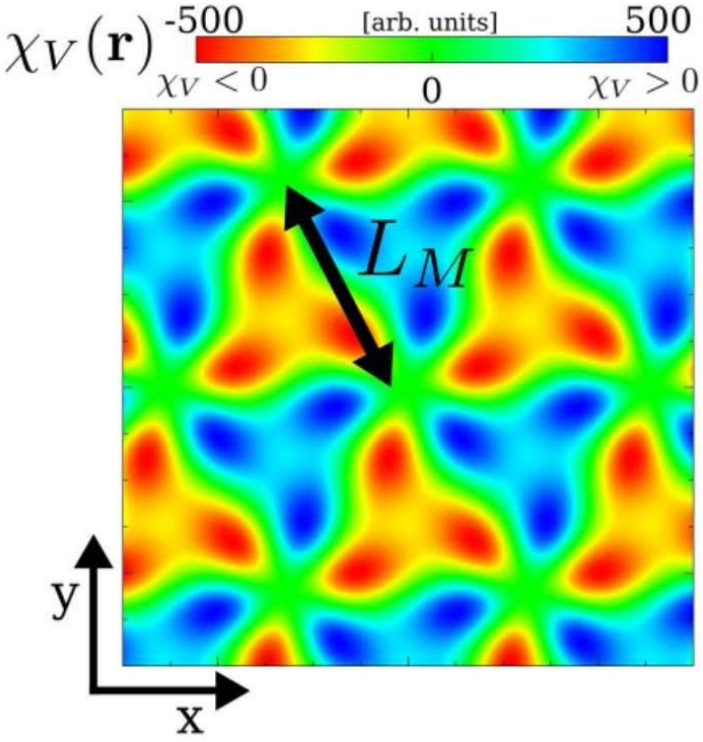
Band structure



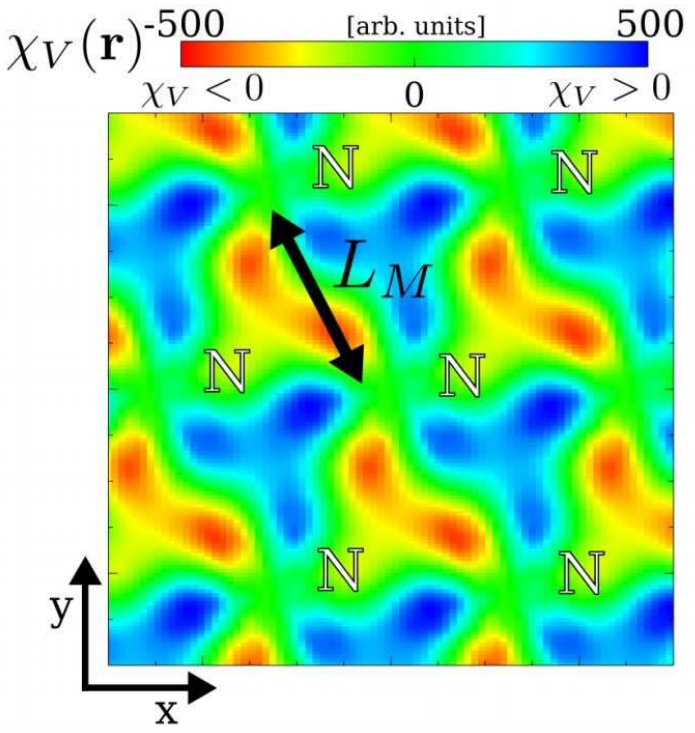
Weak impurities give rise to valley mixing at low energies

Disruption of valley currents by an impurity

Pristine



Weak impurity

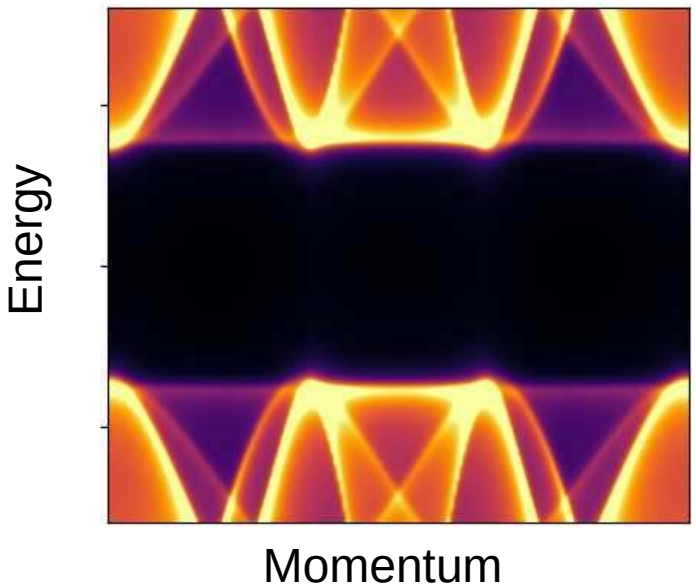


Weak impurities disrupt the valley currents in twisted graphene trilayers

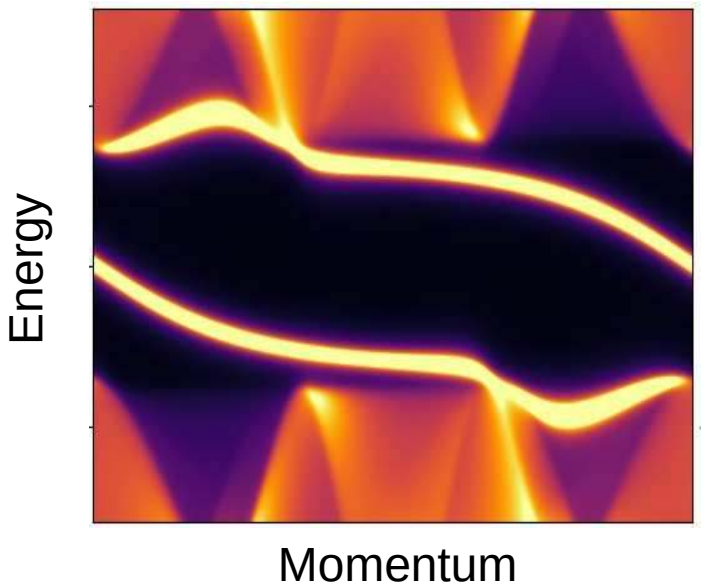
Impurities in a twisted topological superconductor

Topological superconductors

Full gap in the bulk



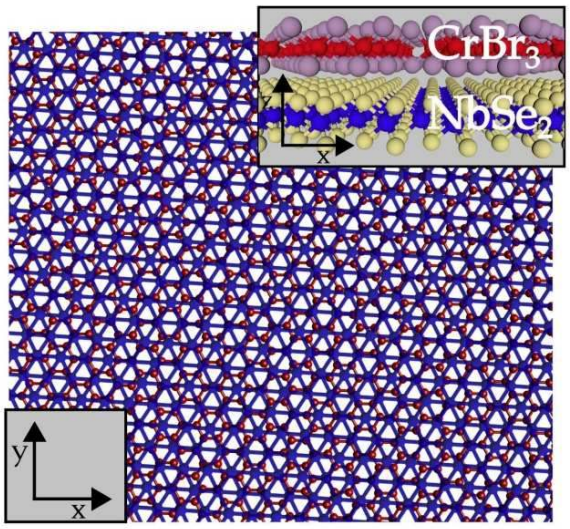
Gapless (Majorana) mode
at the edge



Topological superconductors are rare in nature, and thus have to be artificially engineered

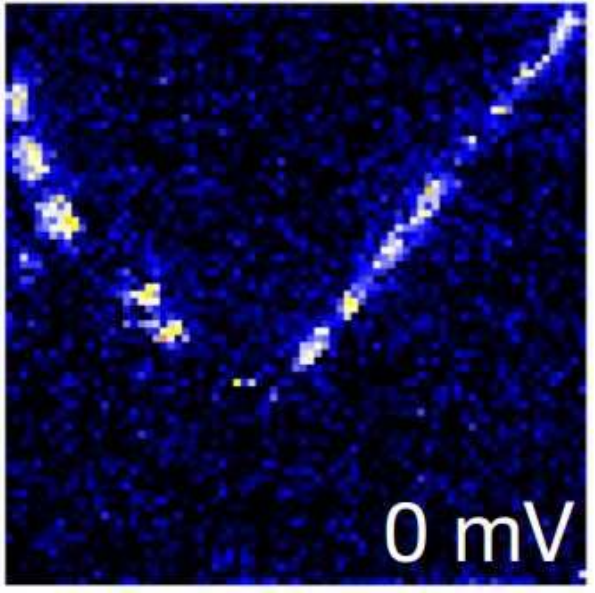
Topological superconductivity in $CrBr_3/NbSe_2$ heterostructure

$CrBr_3/NbSe_2$ heterostructure



Nature 588, 424–428 (2020)
Nano Lett. 2022, 22, 1, 328–333

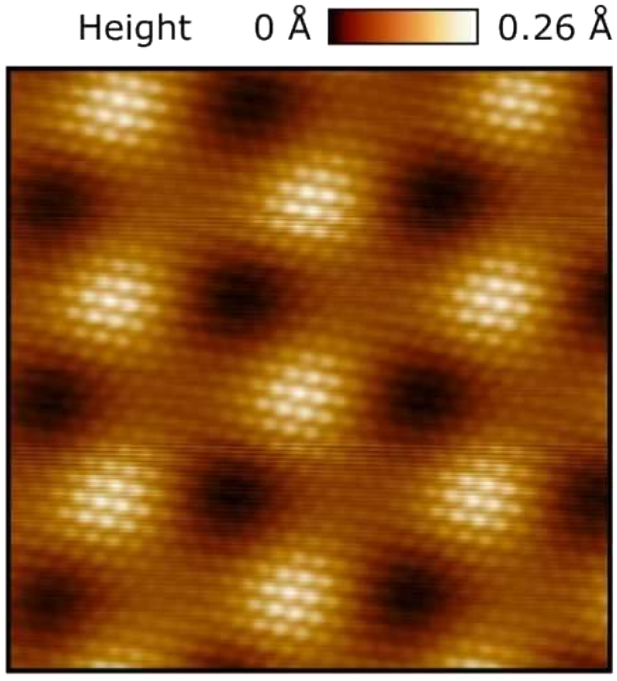
Zero bias dIdV



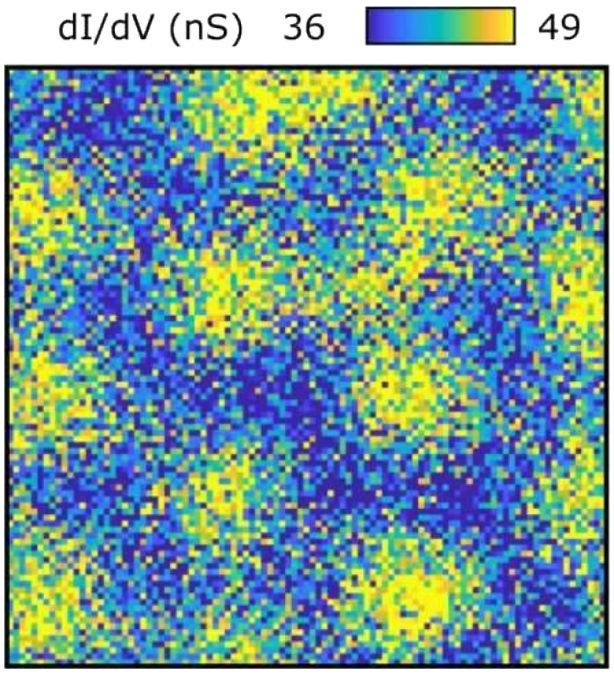
A topological superconducting state appears in $CrBr_3/NbSe_2$

$CrBr_3/NbSe_2$ heterostructure

Topography



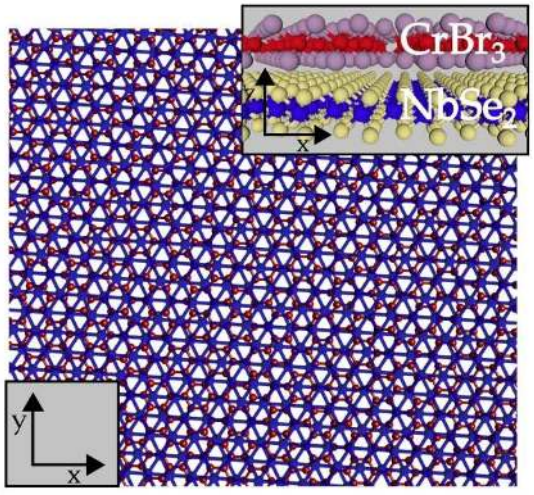
dIdV of the Yu-Shiba-Rusinov states



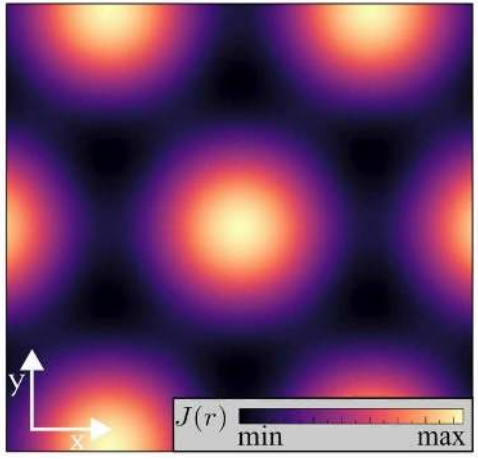
The low energy properties are affected by the moire pattern

Hamiltonian for the pristine moire topological superconductor

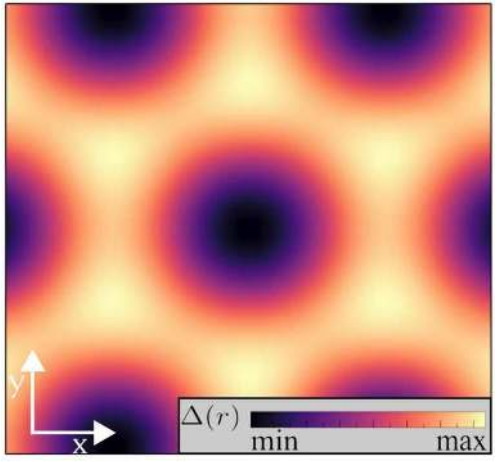
Structure



Exchange profile



Superconducting profile



$$\mathcal{H}_0 = \mathcal{H}_{\text{kin}} + \mathcal{H}_J + \mathcal{H}_R + \mathcal{H}_{\text{SC}}$$

Kinetic energy

Exchange coupling

Rashba SOC

s-wave superconductivity

Hamiltonian for the defective moire topological superconductor

Full Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{imp}}$$

Local impurity in the moire system

$$\mathcal{H}_{\text{imp}} = w \sum_s c^\dagger_{n,s} c_{n,s}$$

Weak impurity

$$w \to 0$$

Strong impurity

$$w \to \infty$$

$$\mathcal{H}_0 = \mathcal{H}_{\text{kin}} + \mathcal{H}_J + \mathcal{H}_R + \mathcal{H}_{\text{SC}}$$

Kinetic energy

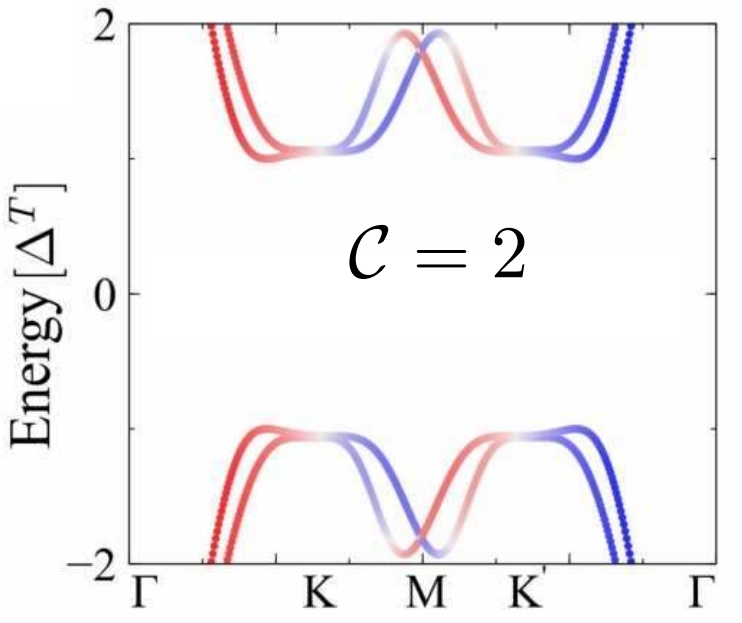
Exchange coupling

Rashba SOC

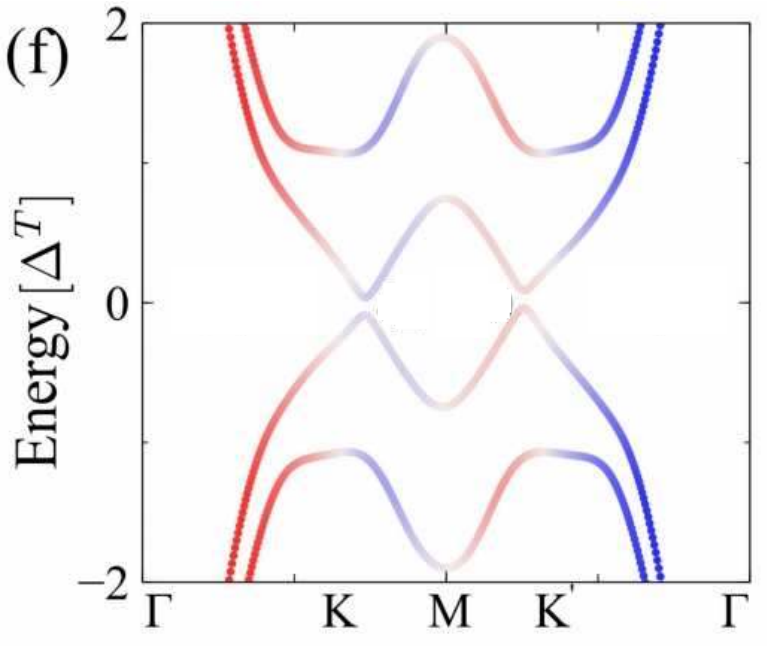
s-wave superconductivity

Impurities in the moire topological superconductor

Band structure in the absence of impurity



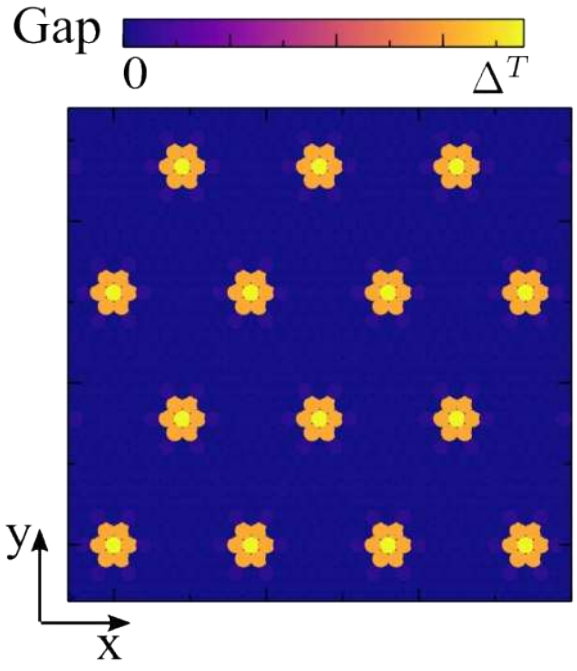
Band structure in the presence of an impurity



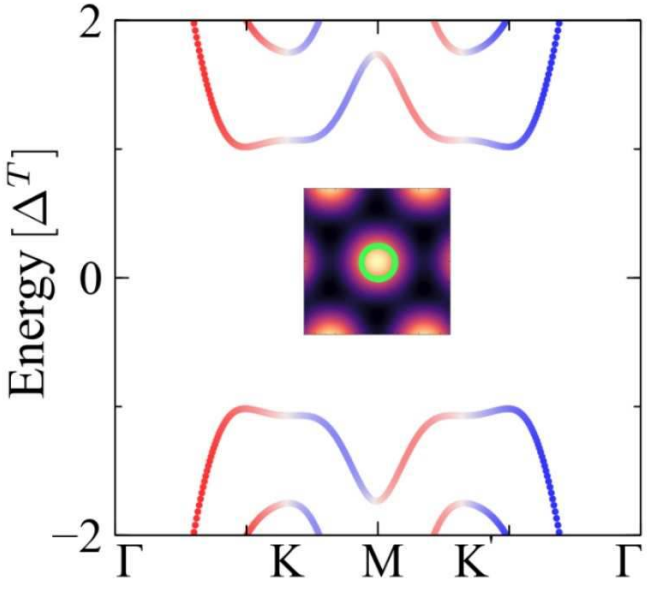
Strong local impurities decrease the size of a topological gap

Gap as a function of the location of a strong impurity

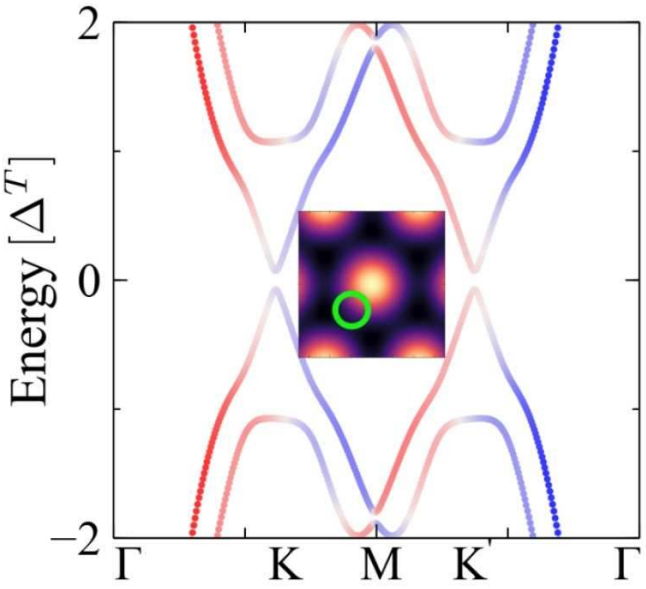
Gap as function of the location of the impurity



Band structure for impurity at the center



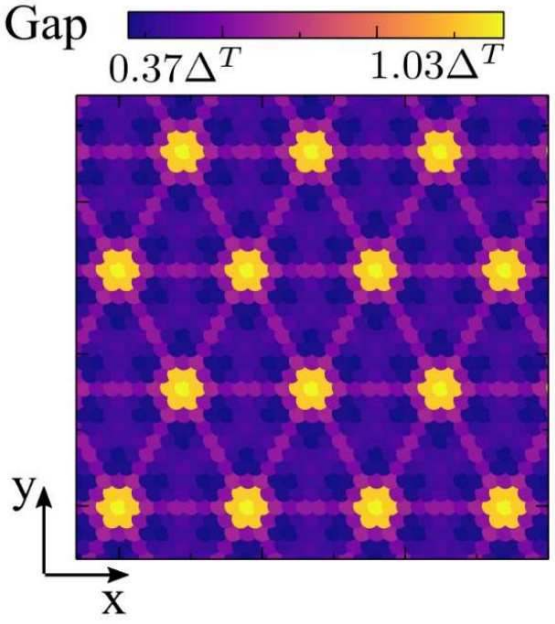
Band structure for impurity away from the center



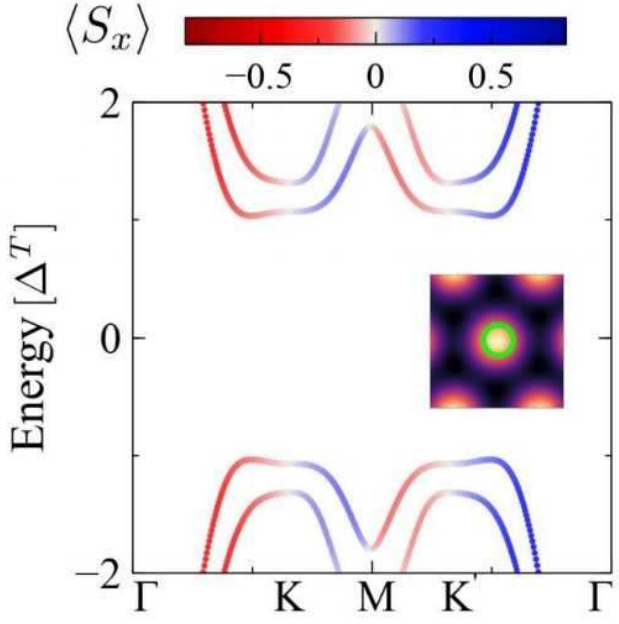
The location of the impurity strongly affects the resulting gap

Gap as a function of the location of a strong impurity

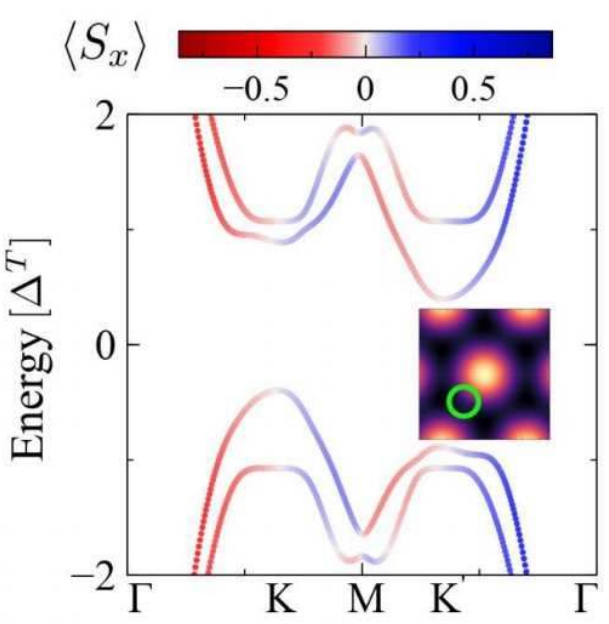
Gap as function of the location of the impurity



Band structure for impurity at the center



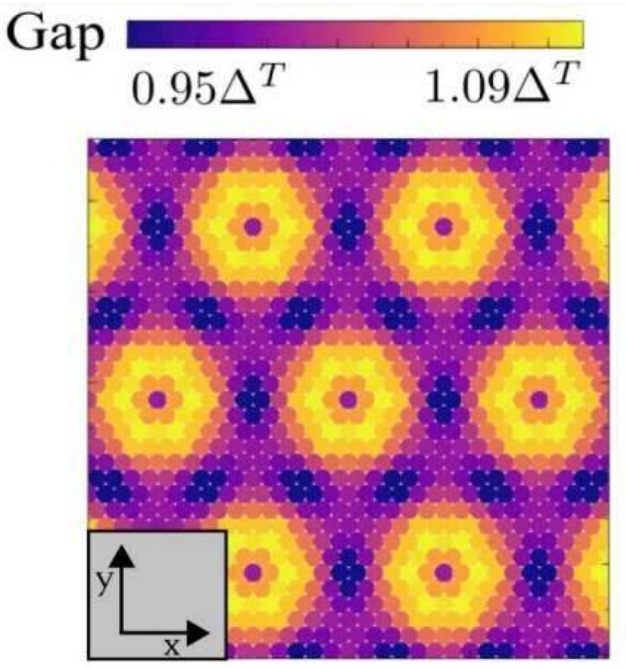
Band structure for impurity away from the center



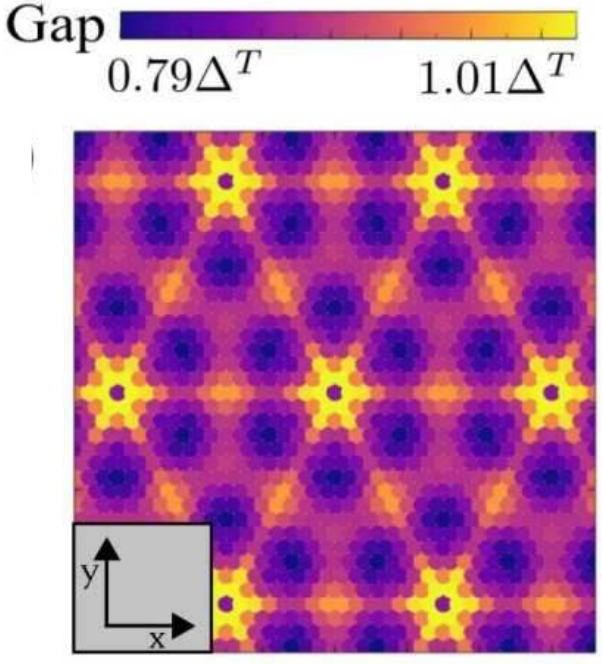
The location of the impurity strongly affects the resulting gap

Gap as function of location for different moire unit cells

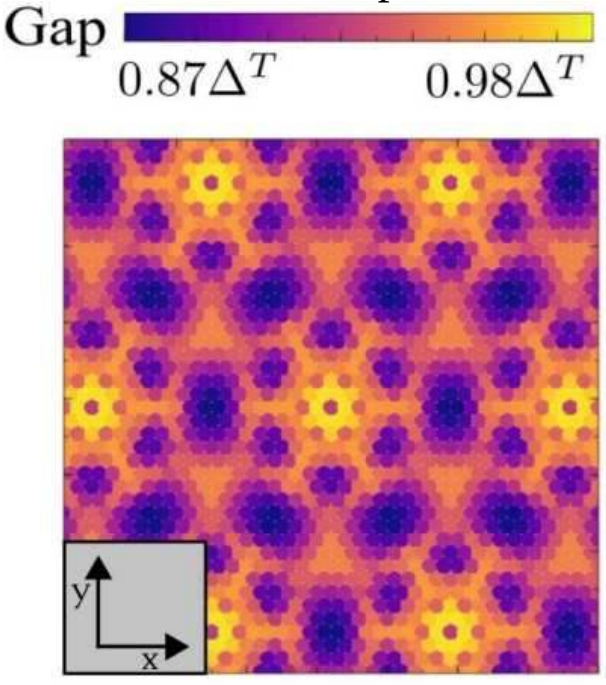
11x11 supercell



15x15 supercell



17x17 supercell



Detrimental locations for impurities depend on the specific moire supercell

The single impurity limit

We now look at the single impurity regime

From the Green's function, we can compute the spatially resolved density of states at the energy of the YSR state

$$\rho(\mathbf{x},\omega) = -\frac{1}{\pi}\sum_{s,\tau}\langle \mathbf{x},s,\tau|\mathrm{Im}(G_V(\omega))|\mathbf{x},s,\tau\rangle$$

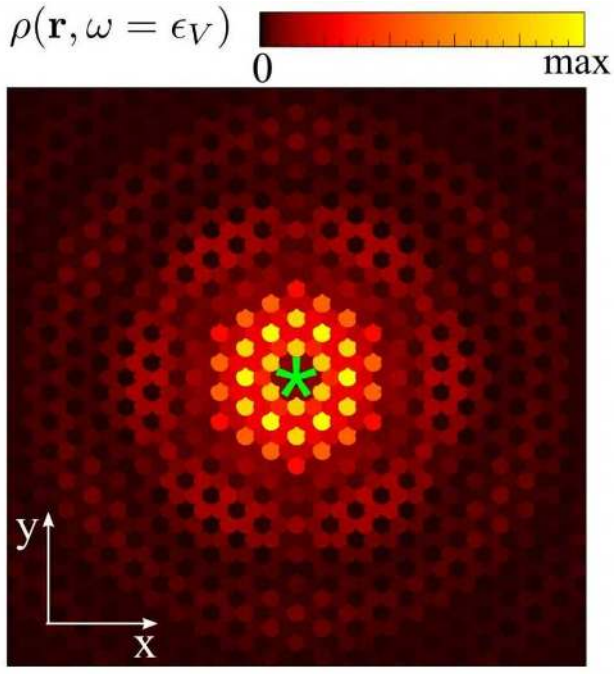
Dyson equation for a single impurity

$$G_V(\omega) = [\omega - H_V - \Sigma(\omega) + i0^+]^{-1}$$

Selfenergy from the pristine regime

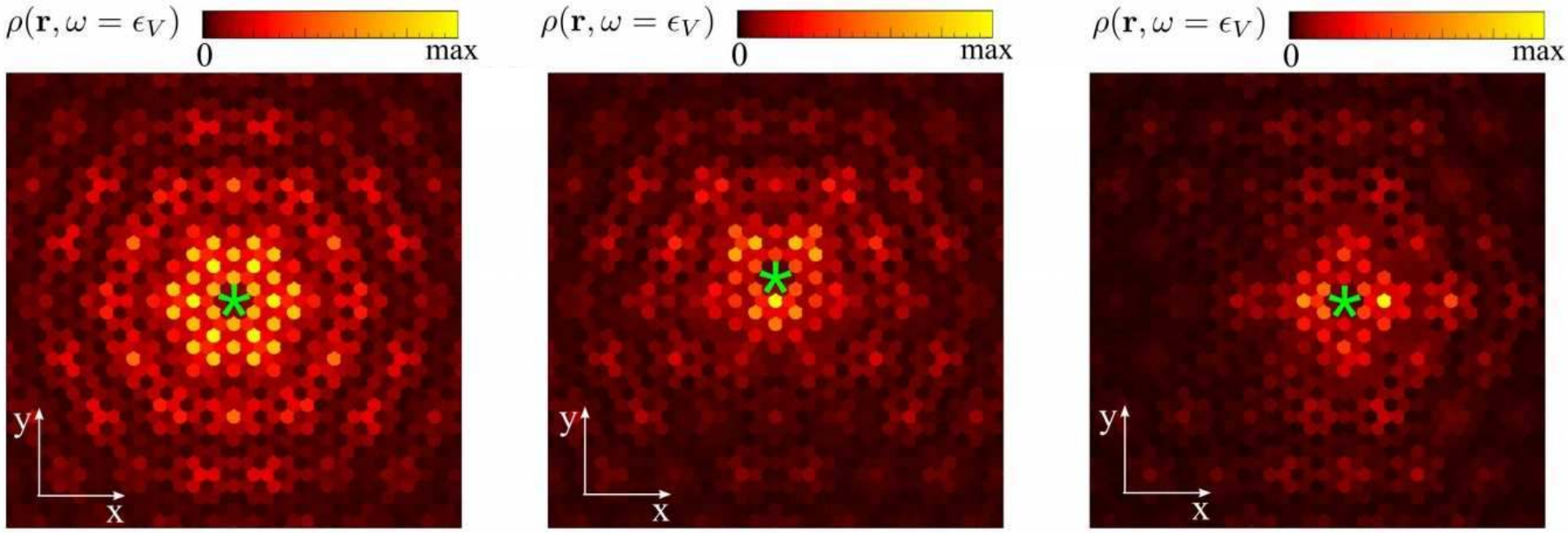
$$G_0(\omega) = [\omega - H_0 - \Sigma(\omega) + i0^+]^{-1}$$

YSR without moire



Single impurity in a topological moire superconductor

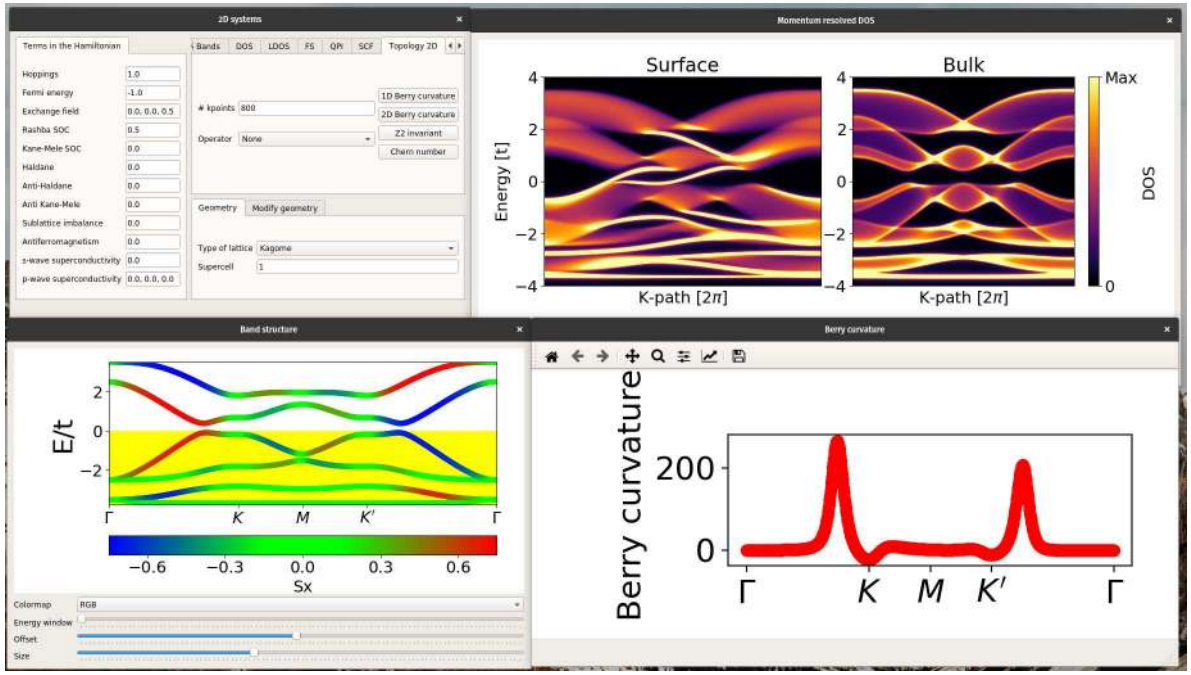
Wavefunction of the YSR state for different impurity locations



The YSR state wavefunction and the moire give rise to interference effects

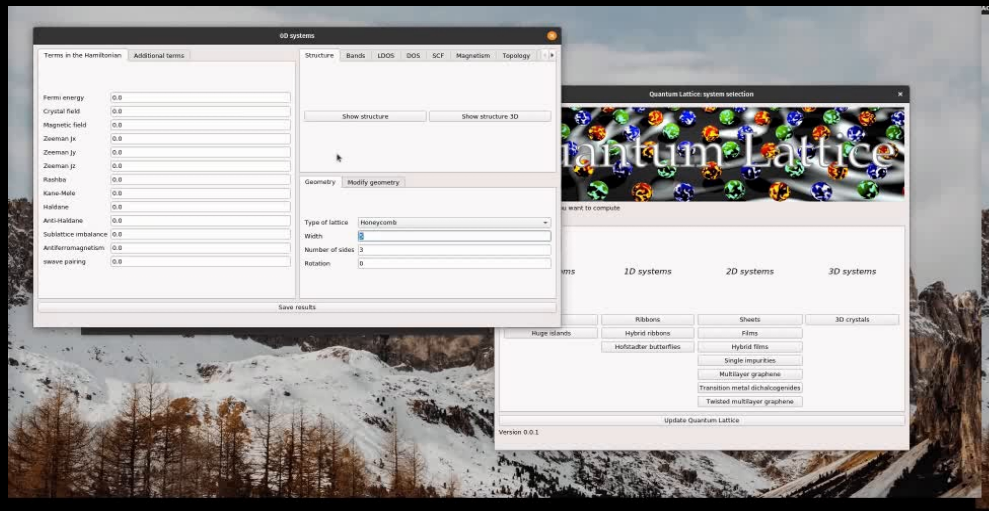
Computing electronic properties

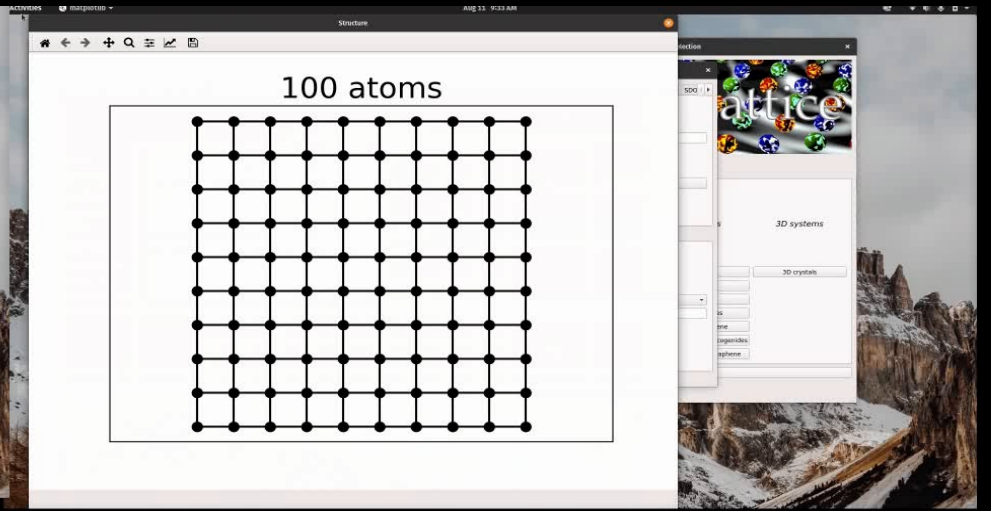
Quantum Lattice: A user interface to compute electronic properties

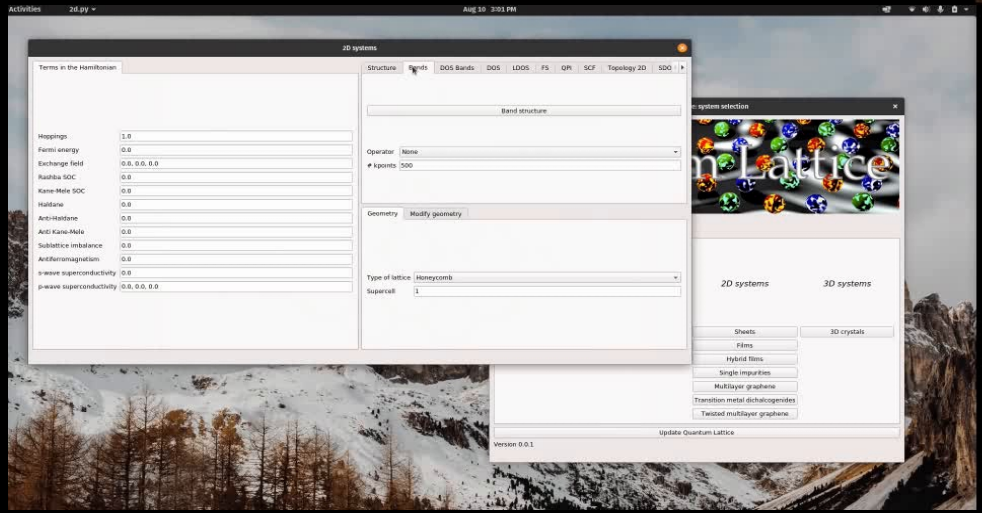


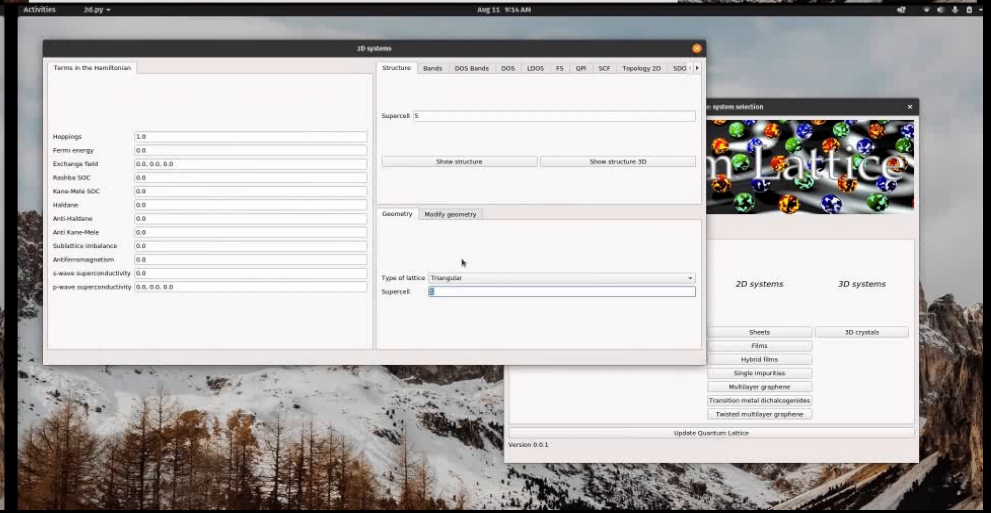
Quantum Lattice: open source interactive interface for tight binding modeling

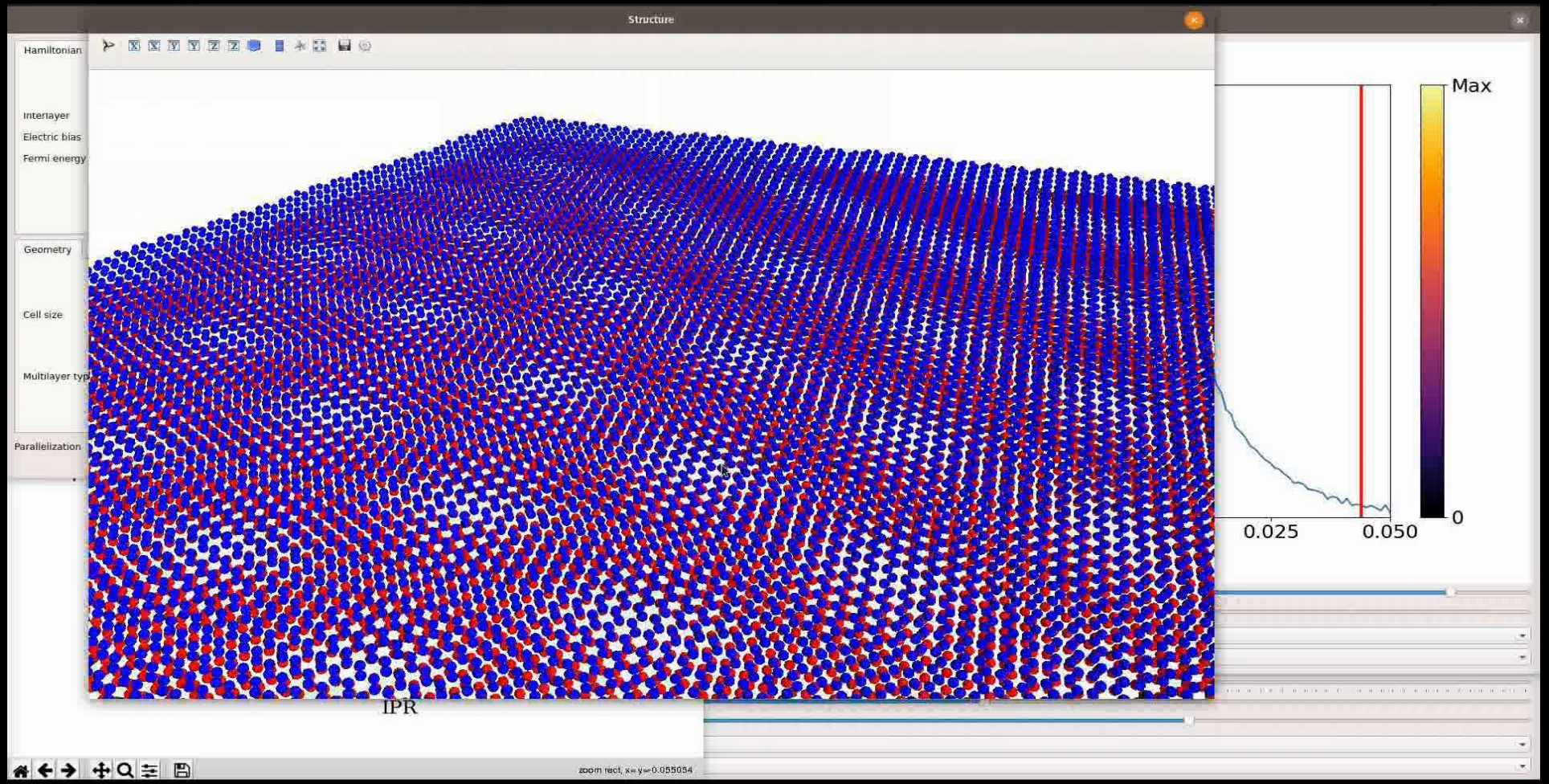
https://github.com/joselado/quantum-lattice


0D systems
Terms in the Hamiltonian
Additional terms
Structure
Bands
LDOS
DOS
SCF
Magnetism
Topology
Fermi energy
Crystal field
Magnetic field
Zeeman Jx
Zeeman Jy
Zeeman Jz
Rashba
Kane-Mele
Haldane
Anti-Haldane
Sublattice imbalance
Antiferromagnetism
swave pairing
Show structure
Show structure 3D
Geometry
Modify geometry
Type of lattice
Honeycomb
Width
Number of sides
Rotation
Save results
Quantum Lattice: system selection
1D systems
2D systems
3D systems
Huge islands
Ribbons
Hybrid ribbons
Hofstadter butterflies
Sheets
Films
Hybrid films
Single impurities
Multilayer graphene
Transition metal dichalcogenides
Twisted multilayer graphene
3D crystals
Update Quantum Lattice
Version 0.0.1


Structure
100 atoms


2D systems
Terms in the Hamiltonian
Structure
Bands
DOS Bands
DOS
LDOS
FS
QPI
SCF
Topology 2D
SDO
Band structure
Operator
None
kpoints
500
Hoppings
Fermi energy
Exchange field
Rashba SOC
Kane-Mele SOC
Haldane
Anti-Haldane
Anti Kane-Mele
Sublattice imbalance
Antiferromagnetism
s-wave superconductivity
p-wave superconductivity
Geometry
Modify geometry
Type of lattice
Honeycomb
Supercell
Quantum Lattice: system selection
2D systems
3D systems
Sheets
Films
Hybrid films
Single impurities
Multilayer graphene
Transition metal dichalcogenides
Twisted multilayer graphene
3D crystals
Update Quantum Lattice
Version 0.0.1

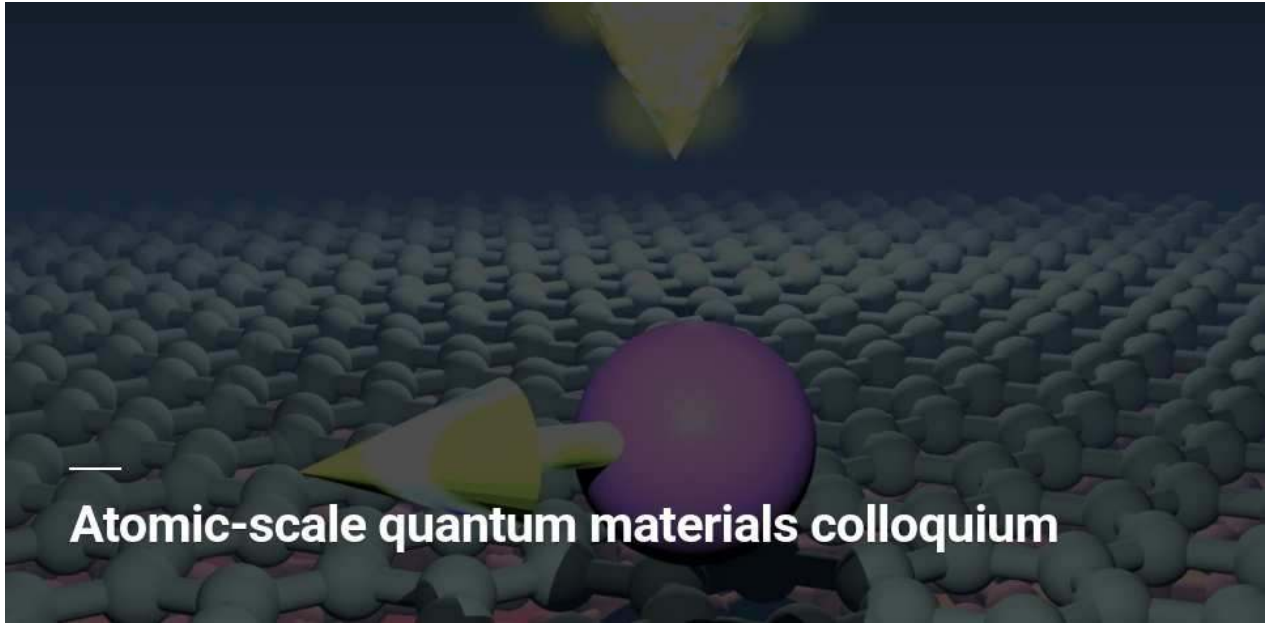

2D systems
Terms in the Hamiltonian
Structure
Bands
DOS Bands
DOS
LDOS
FS
QPI
SCF
Topology 2D
SDO
Supercell
Show structure
Show structure 3D
Geometry
Modify geometry
Type of lattice
Triangular
Supercell
Quantum Lattice: system selection
2D systems
3D systems
Sheets
Films
Hybrid films
Single impurities
Multilayer graphene
Transition metal dichalcogenides
Twisted multilayer graphene
3D crystals
Update Quantum Lattice
Version 0.0.1


Structure
Hamiltonian
Interlayer
Electric bias
Fermi energy
Geometry
Cell size
Multilayer typ
Parallelization
IPR
Max
0
0.025
0.050
zoom rect, x=-y=0.055054

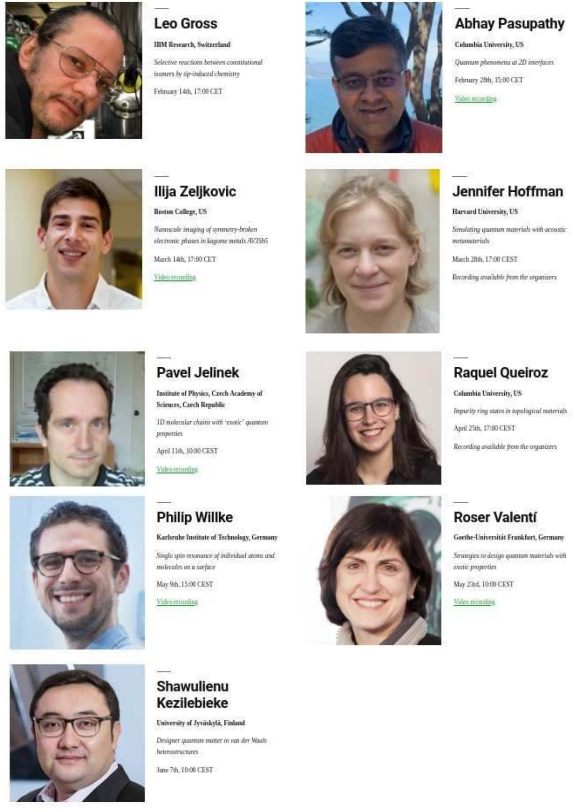
Atomic-scale quantum materials colloquium

Colloquium series on atomic-scale manipulation and engineering of quantum materials

https://asqm-colloquium.aalto.fi/

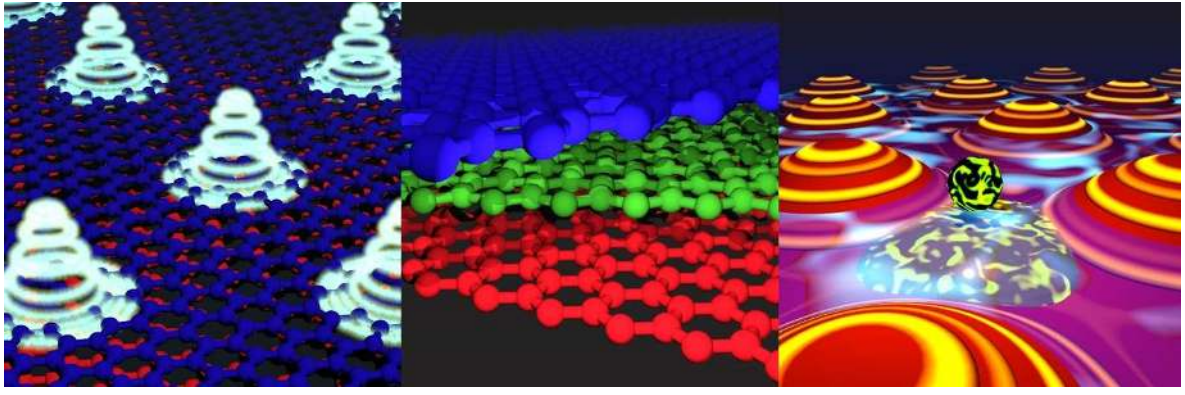


Recordings of past talks available in the colloquium webpage



Take home

Impurities in twisted moire systems can have strong impacts in the low energy dispersion, substantially modifying topological and correlated states



Phys. Rev. B 99, 245118 (2019)
Phys. Rev. Materials 3, 084003 (2019)
Phys. Rev. Research 2, 033357 (2020)
arXiv:2202.11003 (2022)

Thank you!

Funding from

J&AE JANE AND AATOS ERKKO FOUNDATION

SUOMEN AKATEMIA