Twisted bilayer graphene: from DFT to moiré-scale models

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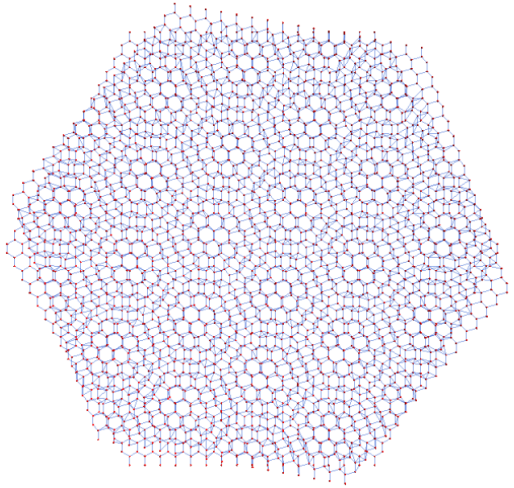
Twisted bilayer graphene (TBG)

- **bottom layer in the plane $z = -\frac{d}{2}$, top layer in the plane $z = \frac{d}{2}$ ($d \simeq 6.45$ bohr)**
- **twist angle θ around the z axis**



Moiré scale

$$a_{\mathrm{M}} = \frac{a_0}{2\sin\frac{\theta}{2}} \quad \sim \quad a_0\theta^{-1} \text{ (small twist angle)}$$

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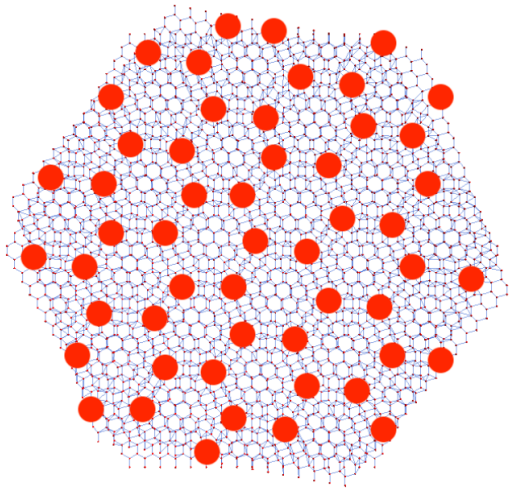

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TBG at "magic" angles (e.g. $\theta \simeq 1.1°$) ∼ honeycomb lattice of quantum dots

- **mean site occupation can be tuned by gating**
- **magnetic flux per moiré cell $\Phi = B|\Omega_{\mathrm{M}}| \sim \Phi_0 = \frac{h}{2e}$ for $\theta \sim 1°$ and $B \sim 20\,\mathrm{T}$**

$\longrightarrow$ **tunable quantum material**

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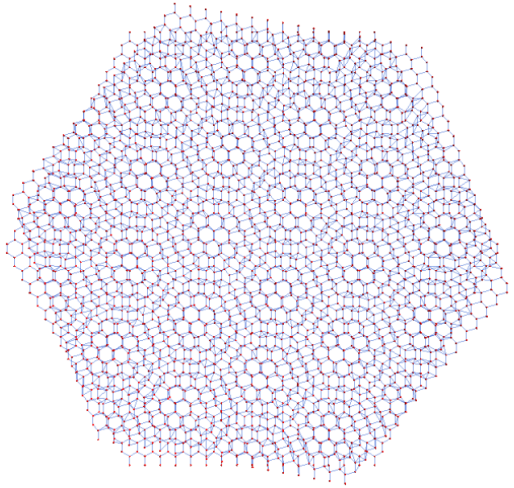
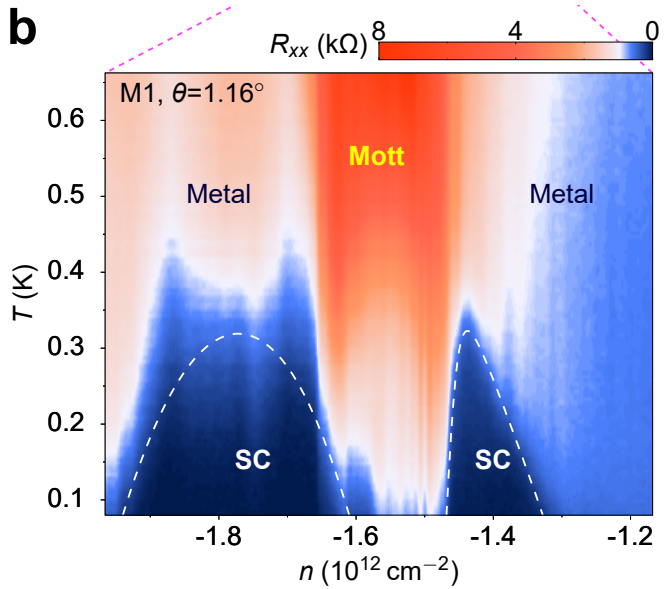


Unconventional(?) superconducting phases for some "magic" angles θ
(Cao et al., Nature '18, 4000+ citations)



TBG phase diagram at twist angle $\theta = 1.16°$

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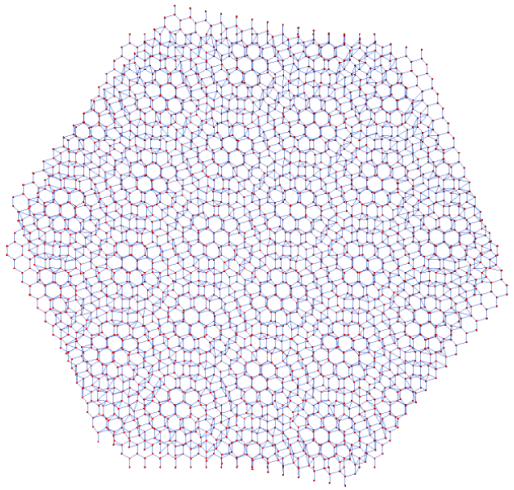

Small twist angles

- **moiré pattern of size $\sim \theta^{-1}a$:** $\theta = 1.16^\circ \Rightarrow$ **$\sim$11,000 carbon atoms/moiré cell**

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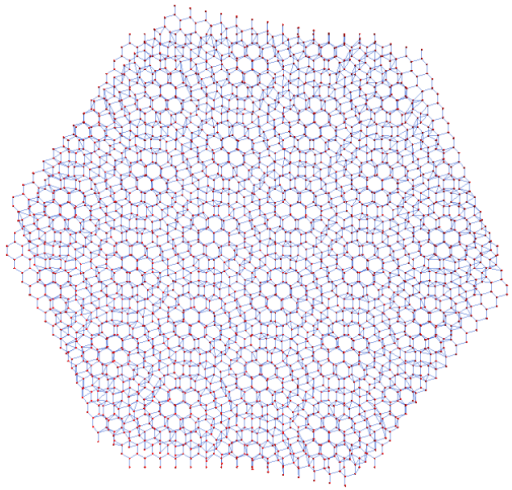

Small twist angles

- **moiré pattern of size $\sim \theta^{-1}a$:** $\theta = 1.16^\circ \Rightarrow$ **~11,000 carbon atoms/moiré cell**
- **most theoretical/numerical studies based on Bistritzer-MacDonald model ('11)**
  Effective model at the moiré scale; two empirical parameters w_{AA}, w_{AB}
  Some math papers: Becker, Embree, Wittsten & Zworski '20, Luskin & Watson '21, Bal, Cazeaux, Massatt, Quinn '22, ...

Derivation of a BM-like model from first principles

A possible approach (Carr-Fang-Zhu-Kaxiras '19, Luskin-Watson '22):

DFT calculations

↓

Tight-binding model (parameterized using Wannier functions)

↓

Continuous model at the moiré scale

Alternative approach followed here

DFT calculations

↓

Continuous model at the moiré scale

1. A simple (formal) derivation of moiré scale models from DFT

C, Garrigue, Gontier, Phys. Rev. B '23

2. Mathematical analysis of Kohn-Sham Hamiltonians for moiré materials

Homogenization methods:

C, Garrigue, Gontier, SIAM J. Math. Anal. '23

Semiclassical analysis (better suited for TBG at 1st magic angle?):

C, Meng, in preparation

1 - A simple (formal) derivation of moiré scale models from DFT

Density Functional Theory: finite systems (spin-unpolarized, smeared nuclei)

Hohenberg & Kohn '64, Kohn & Sham '65, Lieb '84 (W. Kohn 1998 Nobel Laureate in Chemistry)

DFT aims at computing electronic ground-state energies and densities

System with $2N$ electrons, M nuclei with charge density

$$\rho^{\mathrm{nuc}}(\mathbf{r}) = \sum_{m=1}^{M} z_m \phi(\mathbf{r}-\mathbf{R}_m), \quad \phi \in C_{\mathrm{c}}^{\infty}(\mathbb{R}^3), \textbf{ radial, nonnegative, s.t. } \int_{\mathbb{R}^3} \phi = 1$$

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$$E_{\mathrm{GS}} = \inf_{\gamma \in \mathcal{K}_N} E^{\mathrm{KS}}(\gamma)$$

$$\mathcal{K}_N := \left\{ \gamma \in \mathcal{S}(L^2(\mathbb{R}^3)) \cap \mathfrak{S}_1(L^2(\mathbb{R}^3)),\ 0 \le \gamma \le 1,\ \mathrm{Tr}(\gamma) = N,\ \mathrm{Tr}(-\Delta\gamma) < \infty \right\}$$

$$\gamma \in \mathcal{K}_N \ \Leftrightarrow\ \gamma = \sum_{i=1}^{+\infty} n_i |\phi_i\rangle\langle\phi_i| \quad \textbf{with} \quad \begin{cases} \phi_i \in L^2(\mathbb{R}^3),\ (\phi_i,\phi_j)_{L^2} = \delta_{ij},\ n_i \in \mathbb{R} \\ 0 \le n_i \le 1,\ \displaystyle\sum_{i=1}^{+\infty} n_i = N, \\ \phi_i \in H^1(\mathbb{R}^3),\ \displaystyle\sum^{+\infty} n_i \|\nabla\phi_i\|_{L^2}^2 < \infty \end{cases}$$

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$$E^{\mathrm{H}}(\rho) = \frac{1}{2} \int_{\mathbb{R}^3} \int_{\mathbb{R}^3} \frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\, d\mathbf{r}\, d\mathbf{r}'$$

Density Functional Theory: **finite systems (spin-unpolarized, smeared nuclei)**

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Fortunately, E^{xc} only represents about 10% of the total energy and can be successfully approximated ($E^{\mathrm{xc}} \equiv 0 \Rightarrow$ reduced Hartree-Fock - rHF)

Main approximations actually used in pratice

(Burke et al. ’14)

# of citations the B3LYP paper (Web of Science): 72,237

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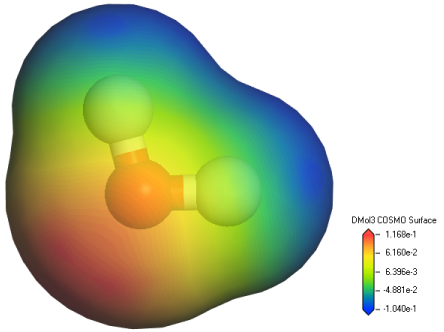
Mathematical justification: **LSDA** **(Lewin-Lieb-Seiringer ’19)** $E^{\mathrm{x}}_{\mathrm{LSDA}}(\rho) = -C_{\mathrm{D}} \int_{\mathbb{R}^3} \rho^{4/3}$

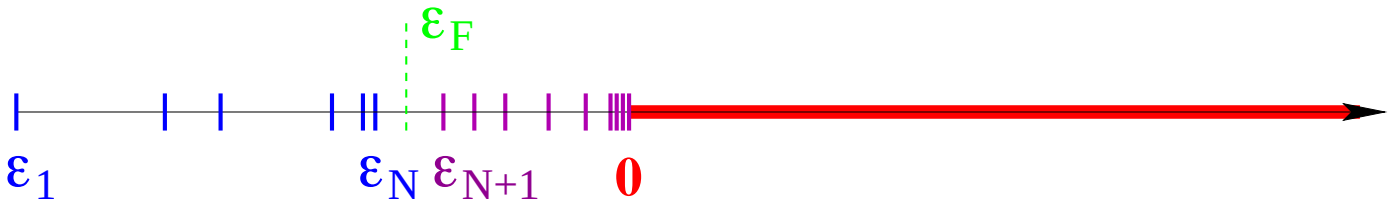
Existence of a ground-state for neutral molecules:

Lieb-Simon ’77, Lions ’83 (Hartree-Fock), **Solovej ’91** (rHF), **Anantharaman-C ’09** (LSDA)

Kohn-Sham equations for finite systems (rHF and LSDA)

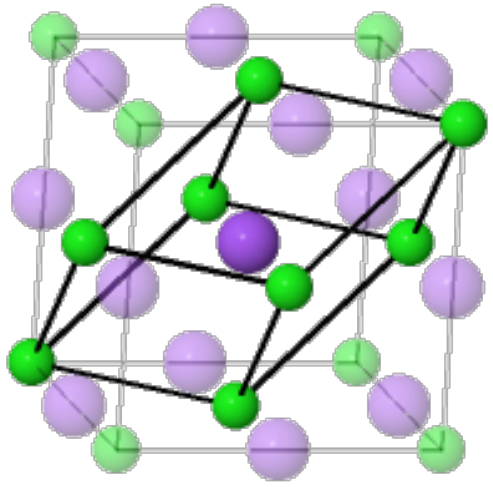
$$\begin{cases} H_{\rho^0} = -\frac{1}{2}\Delta + V^{\mathrm{H}}_{\rho^0} + V^{\mathrm{xc}}_{\rho^0}, \quad V^{\mathrm{H}}_{\rho^0} = (\rho^0 - \rho^{\mathrm{nuc}}) \star |\cdot|^{-1}, \quad V^{\mathrm{xc}}_{\rho^0}(\mathbf{r}) = v^{\mathrm{xc}}(\rho^0(\mathbf{r})) \\ \gamma^0 = \mathbb{1}_{(-\infty,\varepsilon_{\mathrm{F}})}(H_{\rho^0}) + \delta \quad \textbf{with} \quad \mathrm{Ran}(\delta) \subset \mathrm{Ker}(H_{\rho^0} - \varepsilon_{\mathrm{F}}), 0 \leq \delta = \delta^* \leq 1 \\ \text{“}\rho^0(\mathbf{r}) = 2\gamma^0(\mathbf{r},\mathbf{r})\text{”}, \quad \int_{\mathbb{R}^3} \rho^0 = 2\mathrm{Tr}(\gamma^0) = 2N \end{cases}$$

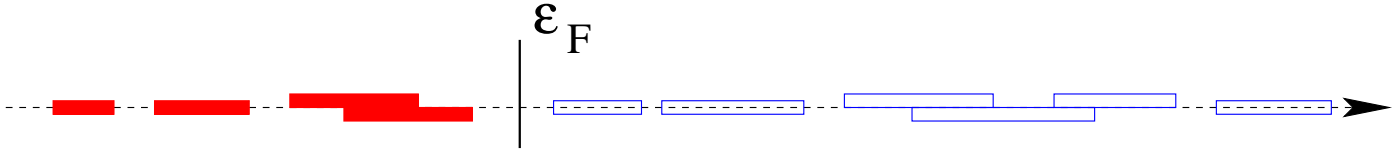




Kohn-Sham equations for periodic crystals (rHF and LSDA)

$$\begin{cases} H_{\rm per} = -\frac{1}{2}\Delta + V^{\rm H}_{\rm per} + V^{\rm xc}_{\rm per}, \quad -\Delta V^{\rm H}_{\rm per} = 4\pi(\rho^0_{\rm per} - \rho^{\rm nuc}_{\rm per}), \quad V^{\rm xc}_{\rm per}(\mathbf{r}) = v^{\rm xc}(\rho^0_{\rm per}(\mathbf{r})) \\ \gamma^0_{\rm per} = \mathbb{1}_{(-\infty,\varepsilon_{\rm F})}(H_{\rm per}) \\ \text{“}\rho^0_{\rm per}(\mathbf{r}) = 2\gamma^0_{\rm per}(\mathbf{r},\mathbf{r})\text{”}, \quad \int_{\rm UC}\rho^0_{\rm per} = 2\underline{\rm Tr}(\gamma^0) = 2N \end{cases}$$





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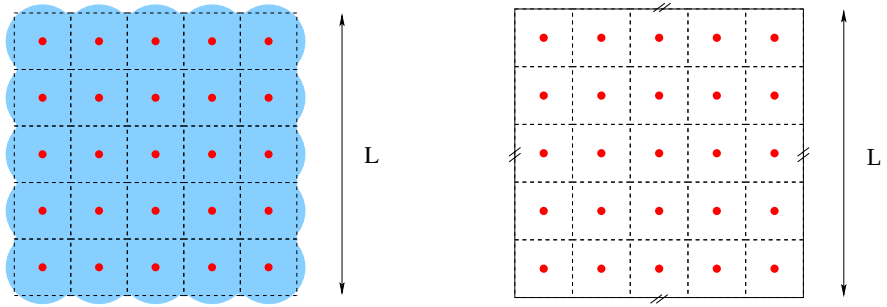
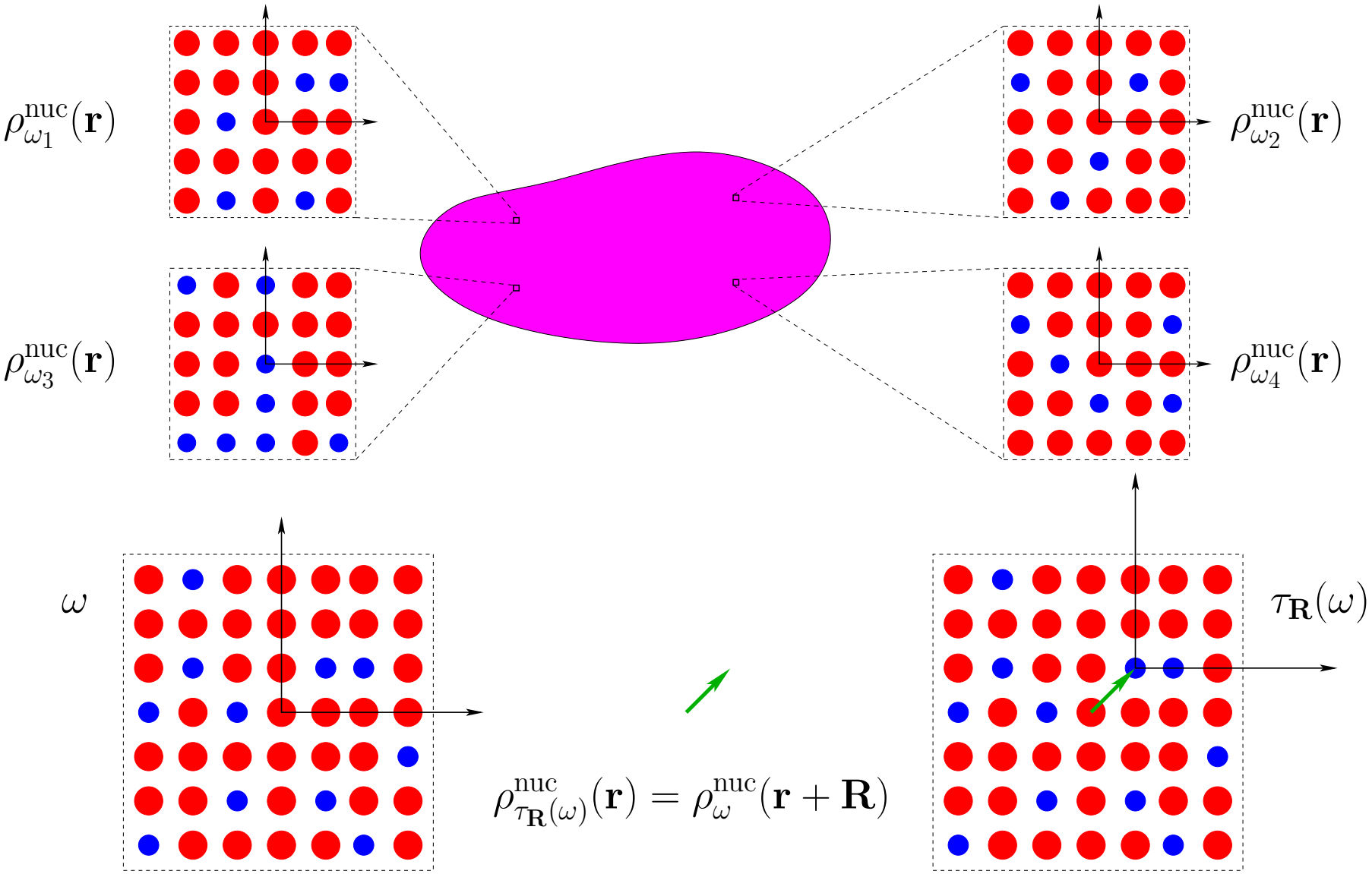
Mathematical justification by thermodynamic limit (for rHF)

- **Catto-Le Bris-Lions ’01: cluster to crystal for** $\rho^{\rm nuc}_{\rm per} = \sum_{\mathbf{R}\in\mathbb{Z}^3}\delta_{\rm R}$
- **E-Deleurence-Lewin ’08: periodic supercell to crystal for generic** $\rho^{\rm nuc}_{\rm per}$ **(closer to numerical practice, also works for crystals with local defects)**



Kohn-Sham equations for disordered crystals (stationary random nuclear charge)



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C, Lahbabi, Lewin '13

$$\begin{cases} H_\omega = -\frac{1}{2}\Delta + V_\omega^{\rm H} + V_\omega^{\rm xc}, \quad (-\Delta + \kappa^2) V_\omega^{\rm H} = 4\pi(\rho_\omega^0 - \rho_\omega^{\rm nuc}), \quad V_\omega^{\rm xc}(\mathbf{r}) = v^{\rm xc}(\rho_\omega^0(\mathbf{r})) \\ \gamma_\omega^0 = \mathbb{1}_{(-\infty,\varepsilon_{\rm F})}(H_\omega) \\ \text{“}\rho_\omega^0(\mathbf{r}) = 2\gamma_\omega^0(\mathbf{r},\mathbf{r})\text{”}, \quad \mathbb{E}\left(\int_{\rm UC} \rho_\bullet^0\right) = 2N \end{cases}$$

$\rho_\omega^0, V_\omega^{\rm H}, V_\omega^{\rm xc}$**: stationary functions,** H_ω, γ_ω**: self-adjoint ergodic operators on** $L^2(\mathbb{R}^3)$

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Roadmap (rHF model):

1. **derive formally a variational Kohn-Sham model for this system**
2. **show existence and uniqueness of the ground-state density**
3. **show existence and uniqueness of the stationary Hartree potential**
4. **show that the ergodic Schrödinger operator has suitable properties**
5. **justify the formal model by thermodynamic limit (periodic supercell)**

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for Coulomb interaction ($\kappa = 0$), we were not able to complete Step 3

Reason: for a given stationary ρ_ω with zero mean, the Poisson equation $-\Delta V_\omega = 4\pi\rho_\omega$ has no stationary solution in general (see e.g. Papanicolaou-Varadhan '82)

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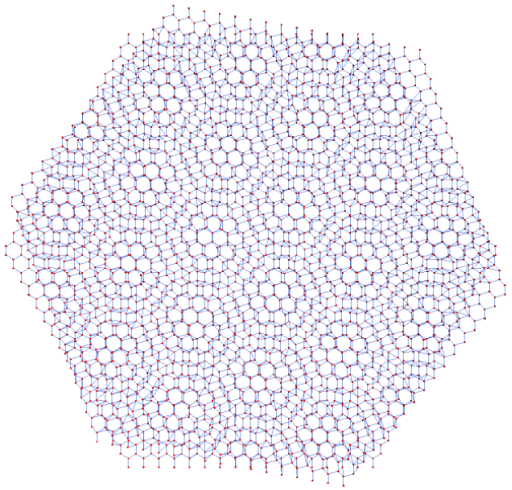

DFT model for TBG?

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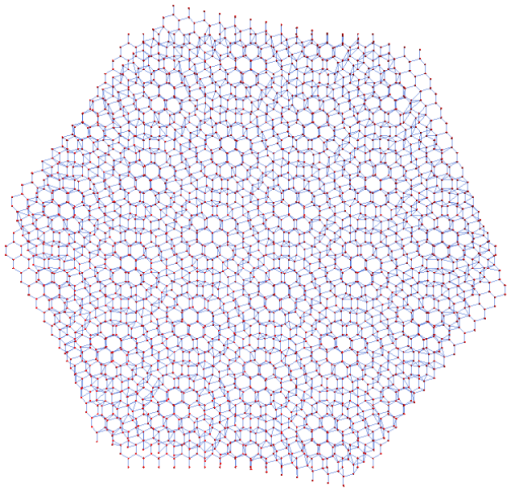

DFT model for TBG?

If $\exists! n, r \in \mathbb{N}^*$, **coprime, s.t.** $\cos\theta = \dfrac{3n^2 + 3nr + r^2/2}{3n^2 + 3nr + r^2}$**, then the TBG is a periodic 2D crystal**

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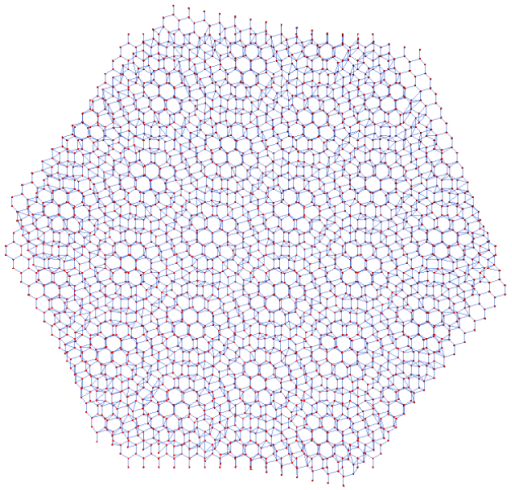

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Otherwise, it is aperiodic (incommensurate)

$\longrightarrow$ | **interesting mathematical questions (PhD thesis of Solal Perrin-Roussel)**
extremely challenging computational / numerical analysis problem

Twisted bilayer graphene (TBG)

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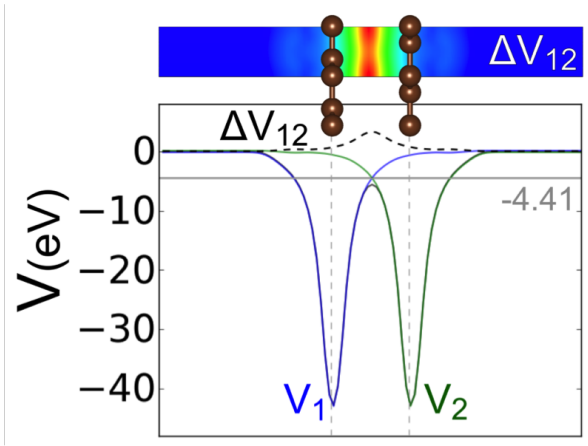
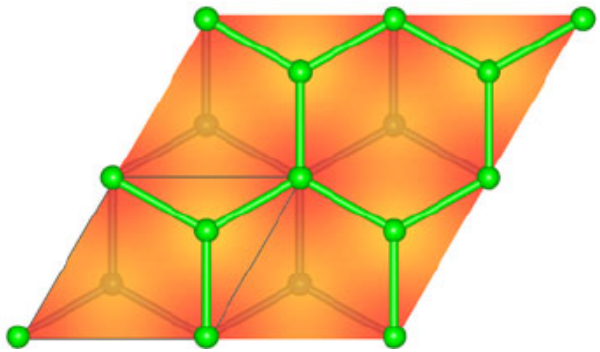
- **twist angle θ around the z axis**
- **approximate Kohn-Sham potential proposed in Tritsaris et al. '16**

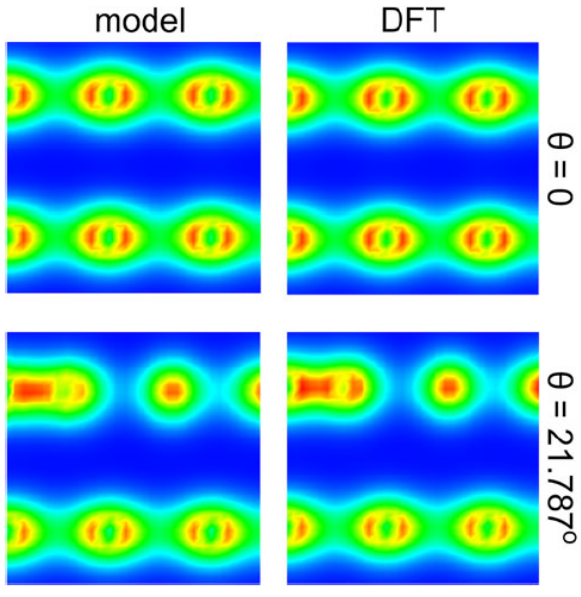
$$\forall \mathbf{r} = (\mathbf{x}, z) \in \mathbb{R}^3, \quad V_{d,\theta}(\mathbf{r}) := (U_{d,\theta} V)(\mathbf{x}, z) + (U_{-d,-\theta} V)(\mathbf{x}, z) + V_{\mathrm{ind},d}(z)$$

$$(U_{d,\theta} f)(\mathbf{x}, z) := f\left(R_{\theta/2}\mathbf{x}, z - d/2\right), \qquad R_{\theta/2} := \begin{pmatrix} \cos(\theta/2) & -\sin(\theta/2) \\ \sin(\theta/2) & \cos(\theta/2) \end{pmatrix}$$

V Kohn-Sham potential of monolayer graphene (untwisted, in the $z = 0$ plane)







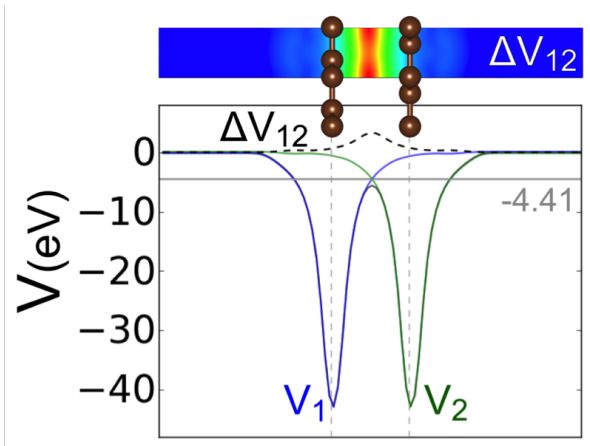
Twisted bilayer graphene (TBG)

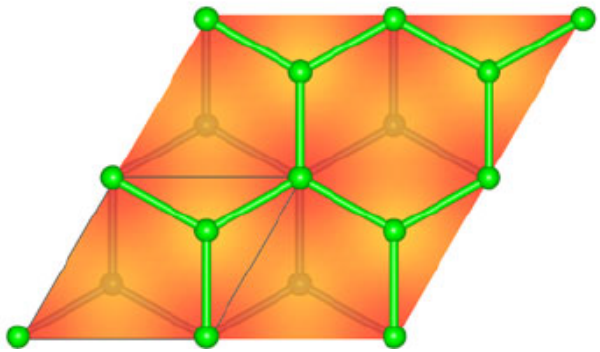
- **bottom layer in the plane $z = -\frac{d}{2}$, top layer in the plane $z = \frac{d}{2}$**
- **twist angle θ around the z axis**
- **approximate Kohn-Sham potential proposed in Tritsaris et al. '16**

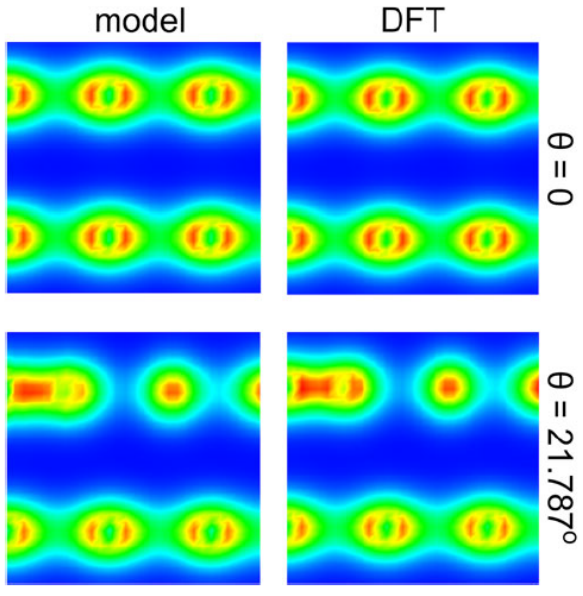
$$\forall \mathbf{r} = (\mathbf{x}, z) \in \mathbb{R}^3, \quad V_{d,\theta}(\mathbf{r}) := (U_{d,\theta} V)(\mathbf{x}, z) + (U_{-d,-\theta} V)(\mathbf{x}, z) + V_{\mathrm{ind},d}(z)$$

$$(U_{d,\theta} f)(\mathbf{x}, z) := f\left(R_{\theta/2}\mathbf{x}, z - d/2\right), \qquad R_{\theta/2} := \underbrace{\cos(\theta/2)}_{c_\theta} \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} - \underbrace{\sin(\theta/2)}_{\frac{1}{2}\varepsilon_\theta} \underbrace{\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}}_{J}$$

V Kohn-Sham potential of monolayer graphene (untwisted, in the $z = 0$ plane)







Twisted bilayer graphene (TBG)

- **bottom layer in the plane $z = -\frac{d}{2}$, top layer in the plane $z = \frac{d}{2}$**
- **twist angle θ around the z axis**
- **approximate Kohn-Sham potential proposed in Tritsaris et al. '16**

$$\forall \mathbf{r} = (\mathbf{x}, z) \in \mathbb{R}^3, \quad V_{d,\theta}(\mathbf{r}) := (U_{d,\theta}V)(\mathbf{x}, z) + (U_{-d,-\theta}V)(\mathbf{x}, z) + V_{\mathrm{ind},d}(z)$$

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V Kohn-Sham potential of monolayer graphene

$$V_{d,\theta}(\mathbf{r}) = v_d\left(c_\theta\mathbf{x}, \varepsilon_\theta\mathbf{x}, z\right) \qquad \textbf{(two-scale potential)}$$

where $v_d : \mathbb{R}^2 \times \mathbb{R}^2 \times \mathbb{R} \to \mathbb{R}$ is defined by

$$v_d\left(\mathbf{x}, \mathbf{X}, z\right) = V(\mathbf{x} - \tfrac{1}{2}J\mathbf{X}, z - d/2) + V(\mathbf{x} + \tfrac{1}{2}J\mathbf{X}, z + d/2) + V_{\mathrm{ind},d}(z)$$

and is

- **$\mathbb{L}$-periodic in the atomic-scale variable $\mathbf{x}$ ($\mathbb{L}$: untwisted MLG Bravais lattice)**
- **$2J\mathbb{L}$-periodic in the moiré-scale variable $\mathbf{X}$**

TBG Hamiltonian

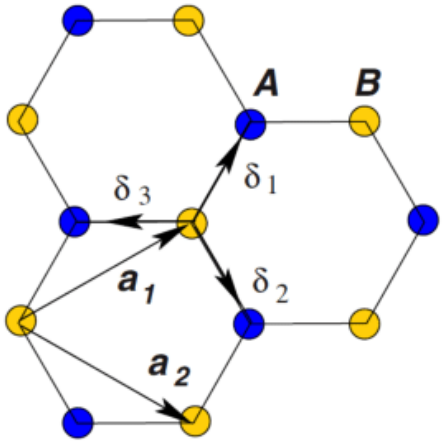
$$H_{d,\theta} = -\frac{1}{2}\Delta + v_d(c_\theta \mathbf{x}, \varepsilon_\theta \mathbf{x}, z)$$

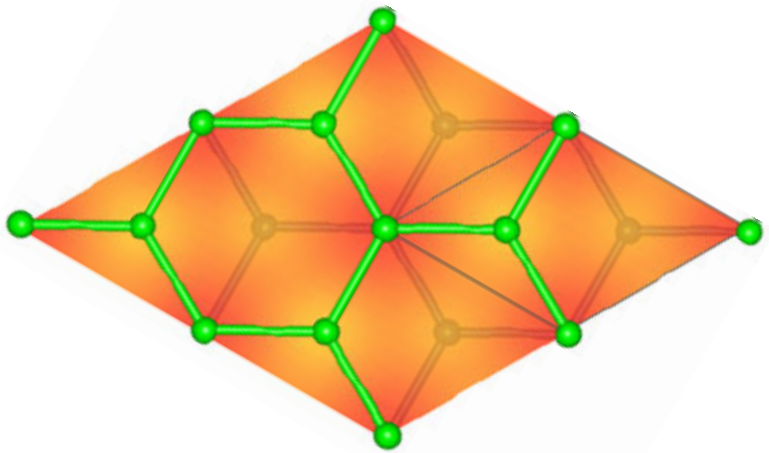
Remark 1. **The Hamiltonian**

$$h_{d,\mathbf{X}} = -\frac{1}{2}\Delta_{\mathbf{x},z} + v_d(\mathbf{x}, \mathbf{X}, z) \quad \textbf{acting on } L^2(\mathbb{R}^2_{\mathbf{x}} \times \mathbb{R}_z; \mathbb{C})$$

describes an untwisted bilayer graphene with disregistry $\mathbf{y} = J\mathbf{X}$

$\mathbf{y} = 0$**:** **A-A stacking** $\mathbf{y} = \frac{1}{3}(\mathbf{a}_1 + \mathbf{a}_2)$**:** **A-B stacking**





TBG Hamiltonian

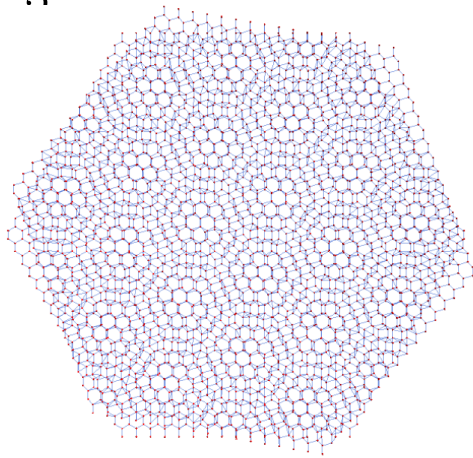
$$H_{d,\theta} = -\frac{1}{2}\Delta + v_d(c_\theta \mathbf{x}, \varepsilon_\theta \mathbf{x}, z)$$

Remark 2. $H_{d,\theta}$ **can also be written as a 3-scale operator**

$$H_{d,\theta} = -\frac{1}{2}\Delta + \widetilde{v}_d(\mathbf{x}, \varepsilon_\theta \mathbf{x}, \widetilde{\varepsilon}_\theta{}^2 \mathbf{x}), \qquad \varepsilon_\theta = 2\sin\frac{\theta}{2}, \quad \widetilde{\varepsilon}_\theta = \sqrt{1-\cos\frac{\theta}{2}} \sim 2^{-3/2}\varepsilon_\theta$$

where $\widetilde{v}_d(\mathbf{x}, \mathbf{X}, \mathfrak{X})$ **is**

- $\mathbb{L}$**-periodic w.r.t. the atomic scale variable** $\mathbf{x}$
- $2J\mathbb{L}$**-periodic w.r.t. the moiré scale variable** $\mathbf{X}$
- $\mathbb{L}$**-periodic w.r.t. the (micron scale - for** $\theta \sim 1°$**) variable** $\mathfrak{X}$



Rescaling the length variable as $\mathbf{x} \to \varepsilon_\theta^{-1}\mathbf{X}$ **and letting** $\theta \to 0$**, we can expect to obtain a 'moiré-periodic' model in the limit**

TBG Hamiltonian

$$H_{d,\theta} = -\frac{1}{2}\Delta + v_d(c_\theta \mathbf{x}, \varepsilon_\theta \mathbf{x}, z)$$

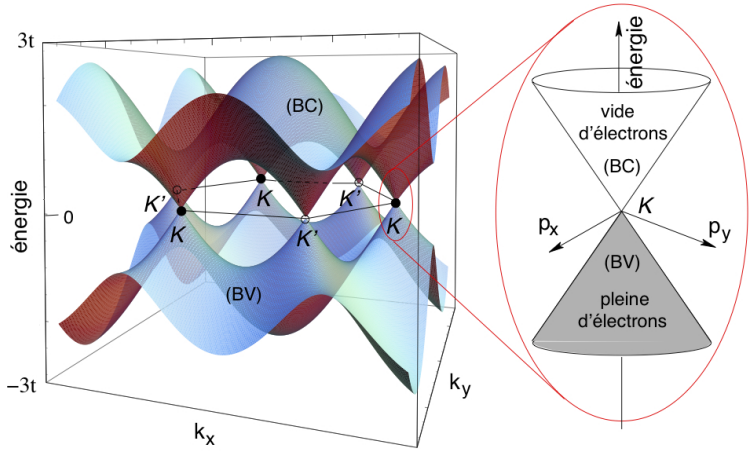
Remark 3. **There are actually two small parameters**

- **the angle θ**
- **the interlayer coupling energies (related to d)**

Electronic structure of monolayer graphene

$H = -\frac{1}{2}\Delta + V$ **(space group Dg80**$:= \underline{\mathrm{D}}_{6h} \ltimes \mathbb{L}$ **- honeycomb p6/mmm symmetry)**

Bloch transform: $H_{\mathbf{k}} = \frac{1}{2}(-i\nabla + \mathbf{k})^2 + V$ **on** $L^2_{\mathrm{per}}(\Omega \times \mathbb{R})$



At $\mathbf{k} = \mathbf{K}$ **(Dirac point): two-fold degenerate eigenvalue right at the Fermi level**

$$H_{\mathbf{K}} u_j = \mu_{\mathrm{F}} u_j, \qquad \Phi_j(\mathbf{x}, z) = u_j(\mathbf{x}, z) e^{i\mathbf{K}\cdot\mathbf{x}}, \qquad R_{\frac{2\pi}{3}} \Phi_j = \omega^j \Phi_j, \qquad \omega = e^{i\frac{2\pi}{3}}$$

$$\langle \Phi_j, (-\mathrm{i}\nabla_{\mathbf{x}})\Phi_{\mathrm{j}} \rangle = \begin{pmatrix} 0 \\ 0 \end{pmatrix}, \quad \langle \Phi_1, (-\mathrm{i}\nabla_{\mathbf{x}})\Phi_2 \rangle = \mathrm{v_F} \begin{pmatrix} 1 \\ -\mathrm{i} \end{pmatrix}, \qquad \mathrm{v_F} > 0$$

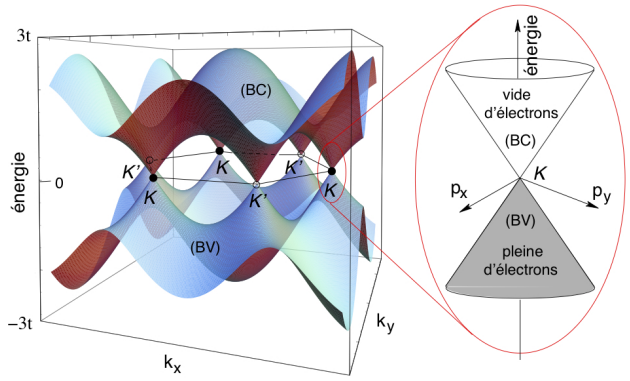
Wavepacket propagation in monolayer graphene (K-valley)

Fefferman-Weinstein '14 (2D, but can be easily extended to 3D graphene)

$$\begin{cases} \psi_\varepsilon(t=0,\mathbf{x},z)=\displaystyle\sum_{j=1}^{2}\varepsilon\alpha_j^0(\varepsilon\mathbf{x})\Phi_j(\mathbf{x},z) \\ i\partial_t\psi_\varepsilon=(H-\mu_{\rm F})\psi_\varepsilon \end{cases} \underset{t<\varepsilon^{-2+\delta}}{\Rightarrow} \begin{cases} \psi_\varepsilon(t,\mathbf{x},z)=\displaystyle\sum_{j=1}^{2}\varepsilon\alpha_j(\varepsilon t,\varepsilon\mathbf{x})\Phi_j(\mathbf{x},z)+\text{small} \\ i\partial_\tau\alpha=v_{\rm F}\boldsymbol{\sigma}\cdot(-i\nabla)\alpha, \quad \alpha(\tau=0)=\alpha^0 \end{cases}$$

$$\boldsymbol{\sigma}\cdot(-i\nabla)=\sigma_1\partial_{x_1}+\sigma_2\partial_{x_2}=\begin{pmatrix} 0 & \partial_{x_1}-i\partial_{x_2} \\ \partial_{x_1}+i\partial_{x_2} & 0 \end{pmatrix} \quad \textbf{self-adjoint op } L^2(\mathbb{R}^2;\mathbb{C}^2)$$

$$v_{\rm F}\sim 10^6\ \mathrm{m\ s^{-1}} \quad \textbf{(slope of the Dirac cone)}$$



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Alternative (formal) derivation of the massless Dirac eq. for monolayer graphene

Project the Schrödinger equation

$$i\varepsilon\partial_\tau\Psi_\varepsilon = (H-\mu_{\rm F})\Psi_\varepsilon$$

on the ε-dependent variational approximation space

$$\mathcal{X}_\varepsilon := \left\{ \sum_{j\in\{1,2\}} \alpha_j(\varepsilon\mathbf{x})\,\Phi_j(\mathbf{x},z),\ \alpha\in H^1(\mathbb{R}^2;\mathbb{C}^2) \right\} \subset H^1(\mathbb{R}^3;\mathbb{C})$$

and let ε go to zero

Derivation of an effective model for TBG (K-valley, small twist angle)

Project the Schrödinger equation

$$i\varepsilon_\theta \partial_\tau \Psi_{d,\theta}(\tau) = (H_{d,\theta} - \mu_{\mathrm{F}})\Psi_{d,\theta}(\tau), \qquad H_{d,\theta} = -\frac{1}{2}\Delta + v_d(c_\theta \mathbf{x}, \varepsilon_\theta \mathbf{x}, z), \qquad \begin{array}{l} c_\theta = \cos(\theta/2) \\ \varepsilon_\theta = 2\sin(\theta/2) \end{array}$$

on the (d,θ)-dependent variational approximation space

$$\mathcal{X}_{d,\theta} := \left\{ (\alpha : \Phi)_{d,\theta},\ \alpha \in H^1(\mathbb{R}^2;\mathbb{C}^4) \right\} \subset H^1(\mathbb{R}^3;\mathbb{C})$$

where

$$(\alpha : \Phi)_{d,\theta}(\mathbf{x}, z) := \sum_{\substack{\eta \in \{\pm 1\} \\ j \in \{1,2\}}} \alpha_{\eta,j}(\varepsilon_\theta \mathbf{x})\, (U_{\eta d, \eta\theta} \Phi_j)(\mathbf{x}, z), \qquad (U_{d,\theta} f)(\mathbf{x}, z) := f\left(R_{\theta/2}\mathbf{x}, z - d/2\right)$$

and let θ go to zero

Physical justification: **weakly interacting layers $\Rightarrow$ perturbation regime**

(Attempts of) mathematical justification: **see Part 2**

Effective Schrödinger equation in the small twist angle limit

$$i\mathcal{S}_d\partial_\tau\alpha(\tau) = \mathcal{H}_{d,\theta}\alpha(\tau), \qquad \mathcal{H}_{d,\theta} \textbf{ and } \mathcal{S}_d \textbf{ self-adjoint op. on } L^2(\mathbb{R}^2;\mathbb{C}^4)$$

Hamiltonian and overlap operators

$$\mathcal{S}_d = \begin{pmatrix} \mathbb{I}_2 & \Sigma_d(\mathbf{X}) \\ \Sigma_d^*(\mathbf{X}) & \mathbb{I}_2 \end{pmatrix} \qquad \textbf{and} \qquad \mathcal{H}_{d,\theta} = \varepsilon_\theta^{-1}\mathcal{V}_d + c_\theta T_d + \varepsilon_\theta T_d^{(1)}$$

Effective Schrödinger equation in the small twist angle limit

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$$[\Sigma_d(\mathbf{X})]_{jj'} := \int_{\Omega\times\mathbb{R}} \overline{\Phi_j\left(\mathbf{x} - \tfrac{1}{2}J\mathbf{X}, z - d/2\right)}\, \Phi_{j'}\left(\mathbf{x} + \tfrac{1}{2}J\mathbf{X}, z + d/2\right)\, d\mathbf{x}\, dz$$

Effective Schrödinger equation in the small twist angle limit

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$$\begin{aligned}[\Sigma_d(\mathbf{X})]_{jj'} :&= \int_{\Omega\times\mathbb{R}} \overline{\Phi_j\left(\mathbf{x}-\tfrac{1}{2}J\mathbf{X}, z-d/2\right)}\,\Phi_{j'}\left(\mathbf{x}+\tfrac{1}{2}J\mathbf{X}, z+d/2\right)\,d\mathbf{x}\,dz \\ &= e^{-i\mathbf{q}_1\cdot\mathbf{X}} \underbrace{\int_{\Omega\times\mathbb{R}} \overline{u_j\left(\mathbf{x}-\tfrac{1}{2}J\mathbf{X}, z-d/2\right)}\,u_{j'}\left(\mathbf{x}+\tfrac{1}{2}J\mathbf{X}, z+d/2\right)\,d\mathbf{x}\,dz}_{\mathbb{L}_M\textbf{-periodic function}}\end{aligned}$$

where

$$\mathbb{L}_M := J\mathbb{L} \quad \textbf{(moiré lattice)} \qquad \textbf{and} \qquad \mathbf{q}_1 := -J\mathbf{K}$$

Effective Schrödinger equation in the small twist angle limit

$$i\mathcal{S}_d\partial_\tau\alpha(\tau) = \mathcal{H}_{d,\theta}\alpha(\tau), \qquad \mathcal{H}_{d,\theta} \textbf{ and } \mathcal{S}_d \textbf{ self-adjoint op. on } L^2(\mathbb{R}^2;\mathbb{C}^4)$$

Hamiltonian and overlap operators

$$\mathcal{S}_d = \begin{pmatrix} \mathbb{I}_2 & \Sigma_d(\mathbf{X}) \\ \Sigma_d^*(\mathbf{X}) & \mathbb{I}_2 \end{pmatrix} \quad \textbf{and} \quad \mathcal{H}_{d,\theta} = \varepsilon_\theta^{-1}\mathcal{V}_d + c_\theta T_d + \varepsilon_\theta T_d^{(1)}$$

$$\mathcal{V}_d = \begin{pmatrix} \mathbb{W}_d^+(\mathbf{X}) & \mathbb{V}_d(\mathbf{X}) \\ \mathbb{V}_d(\mathbf{X})^* & \mathbb{W}_d^-(\mathbf{X}) \end{pmatrix} \quad \textbf{(multiplication operator / scalar potential)}$$

$[\mathbb{W}_d^\pm(\mathbf{X})]_{jj'}$ $\mathbb{L}_\mathrm{M}$**-periodic function**

$[\mathbb{V}_d(\mathbf{X})]_{jj'}$ $e^{-i\mathbf{q}_1\cdot\mathbf{X}} \times$ $\mathbb{L}_\mathrm{M}$**-periodic function**

Effective Schrödinger equation in the small twist angle limit

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$$T_d = \begin{pmatrix} v_F\boldsymbol{\sigma}\cdot(-\mathrm{i}\nabla) & \frac{1}{2}J(-\mathrm{i}\nabla\Sigma_\mathrm{d})(\mathbf{X})\cdot(-\mathrm{i}\nabla) \\ -\frac{1}{2}J(-\mathrm{i}\nabla\Sigma_\mathrm{d}^*)(\mathbf{X})\cdot(-\mathrm{i}\nabla) & v_F\boldsymbol{\sigma}\cdot(-\mathrm{i}\nabla) \end{pmatrix}$$

Effective Schrödinger equation in the small twist angle limit

$$i\mathcal{S}_d\partial_\tau\alpha(\tau) = \mathcal{H}_{d,\theta}\alpha(\tau), \qquad \mathcal{H}_{d,\theta} \textbf{ and } \mathcal{S}_d \textbf{ self-adjoint op. on } L^2(\mathbb{R}^2;\mathbb{C}^4)$$

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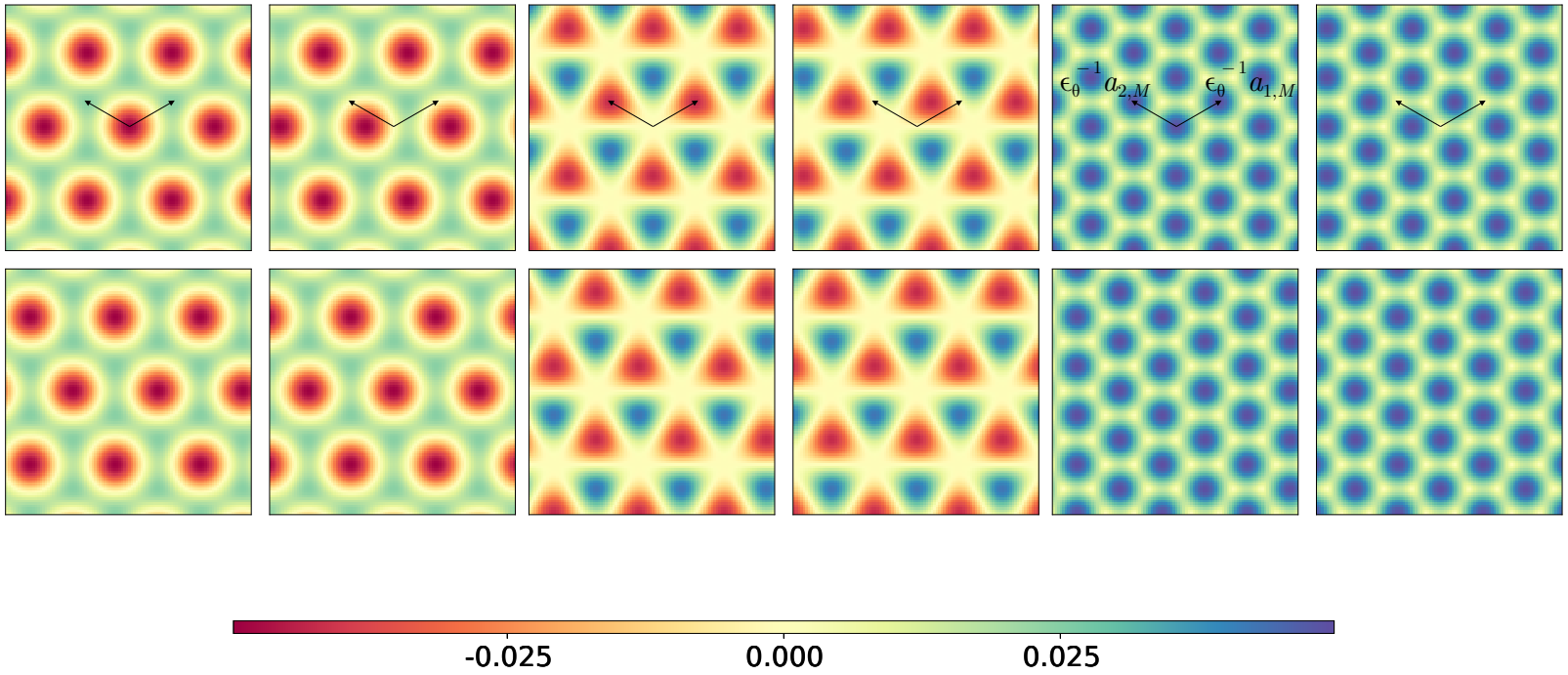
$$T_d^{(1)} = -\frac{1}{2}\mathrm{div}\,(\mathcal{S}_d(\mathbf{X})\nabla\bullet) + \begin{pmatrix} -v_F\boldsymbol{\sigma}\cdot J(-\mathrm{i}\nabla) & 0 \\ 0 & v_F\boldsymbol{\sigma}\cdot J(-\mathrm{i}\nabla) \end{pmatrix}.$$

Comparison with the Bistritzer-MacDonald model

The BM model is obtained by

1. neglecting the interlayer overlap matrix $\Sigma_d(\mathbf{X})$



Plots of $\Sigma(\mathbf{X})$ **for** $d = 6.45$ **a.u. (PBE xc functional)**

Comparison with the Bistritzer-MacDonald model

The BM model is obtained by

1. **neglecting the interlayer overlap matrix** $\Sigma_d(\mathbf{X})$
2. **assuming that the matrix** $\mathbb{V}_d(\mathbf{X})$ **is equal to**

$$\boldsymbol{V}(\mathbf{X}) = \begin{pmatrix} w_{\mathrm{AA}}\, G(\mathbf{X}) & w_{\mathrm{AB}}\, \overline{F(-\mathbf{X})} \\ w_{\mathrm{AB}}\, F(\mathbf{X}) & w_{\mathrm{AA}}\; G(\mathbf{X}) \end{pmatrix}, \qquad \begin{aligned} F(\mathbf{X}) &:= e^{-i\mathbf{q}_1\cdot\mathbf{X}} + e^{i\frac{2\pi}{3}} e^{-i\mathbf{q}_2\cdot\mathbf{X}} + e^{i\frac{4\pi}{3}} e^{-i\mathbf{q}_3\cdot\mathbf{X}} \\ G(\mathbf{X}) &:= e^{-i\mathbf{q}_1\cdot\mathbf{X}} + e^{-i\mathbf{q}_2\cdot\mathbf{X}} + e^{-i\mathbf{q}_3\cdot\mathbf{X}} \end{aligned}$$

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Taking $w_{\mathrm{AA}} = w_{\mathrm{AB}} = 126\ \mathrm{meV}$**, we get** $\dfrac{\|\mathbb{V}_d - \boldsymbol{V}\|_{L^2}}{\|\mathbb{V}_d\|_{L^2}} \sim 10^{-3}$

... to be compared with the values $w_{\mathrm{AA}} = w_{\mathrm{AB}} = 110\ \mathrm{meV}$ **in the BM paper**

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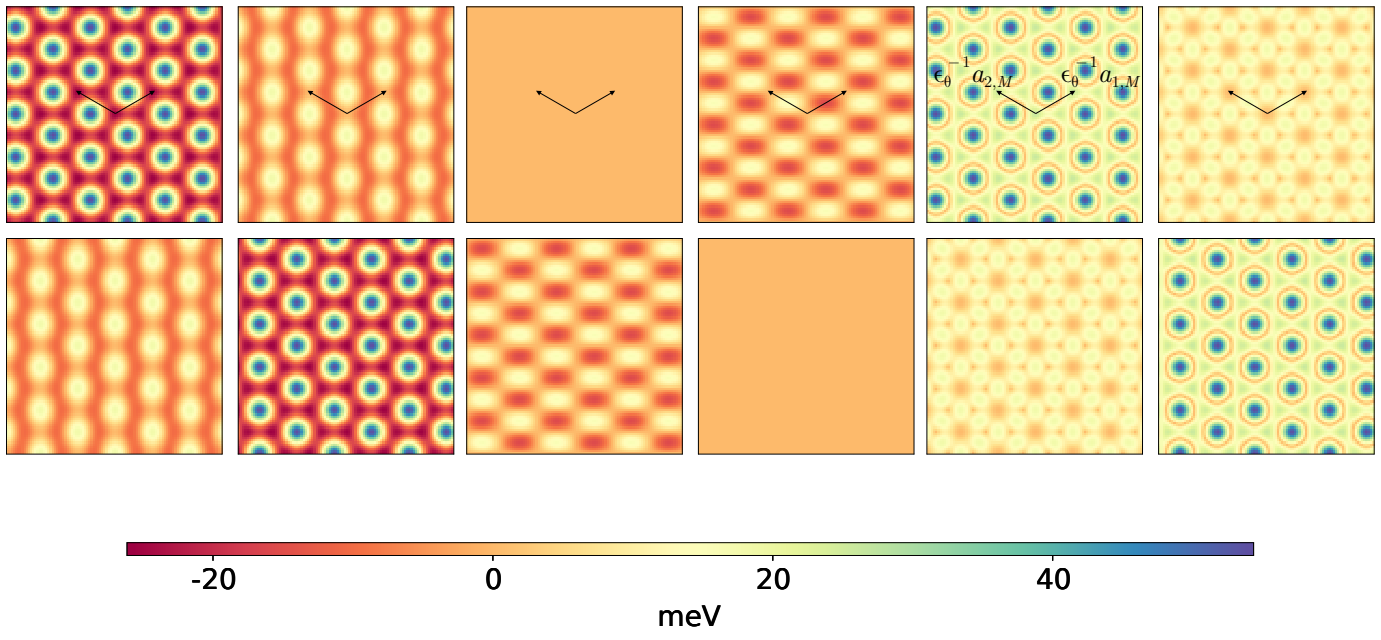
3. assuming that the functions $\mathbb{W}_d^{\pm}(\mathbf{X}) - \delta\mu_{\mathrm{F}} I_2$ **are very small**



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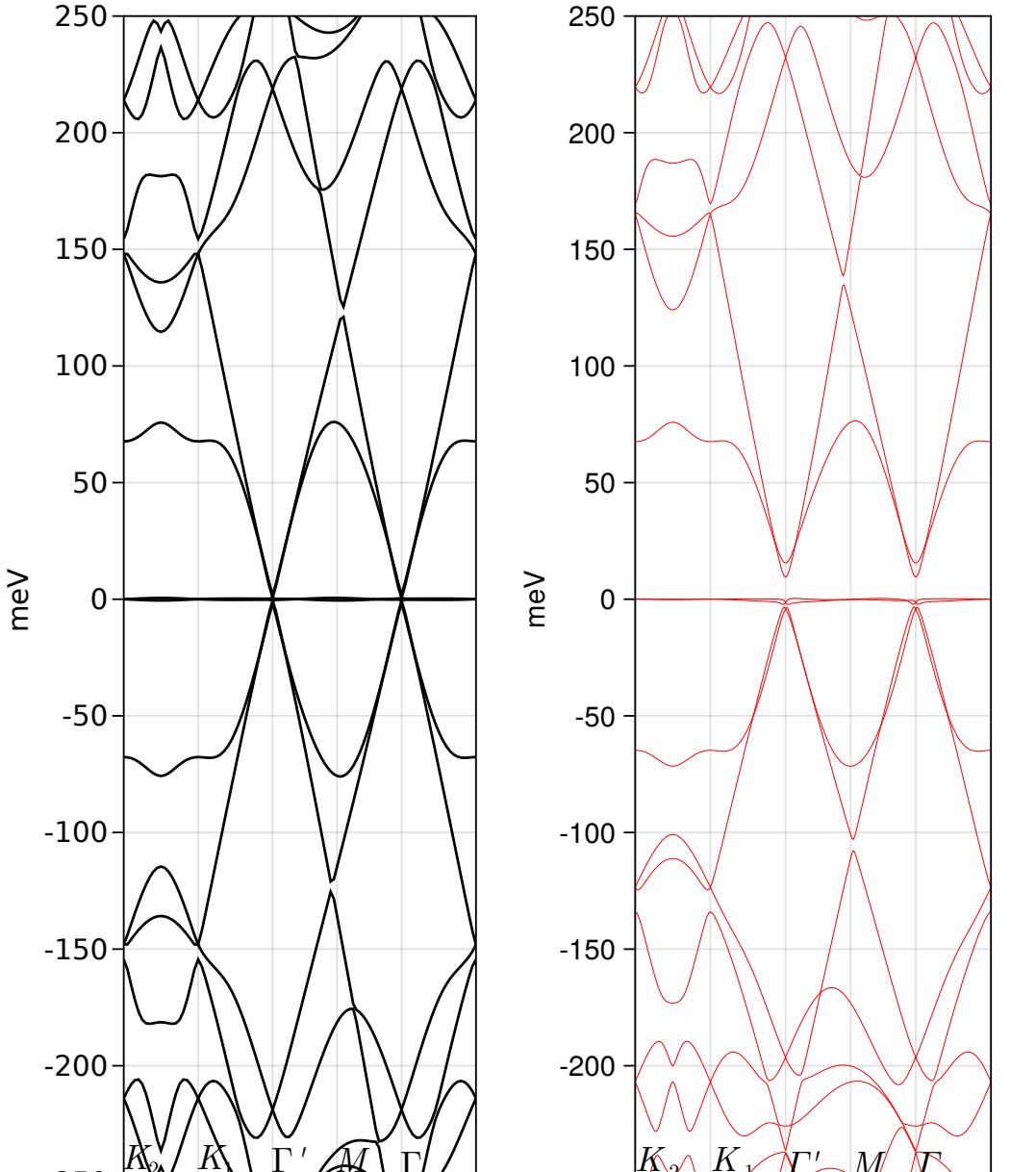
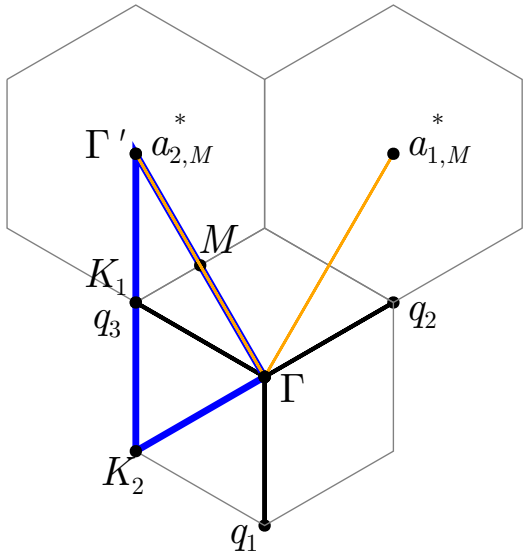
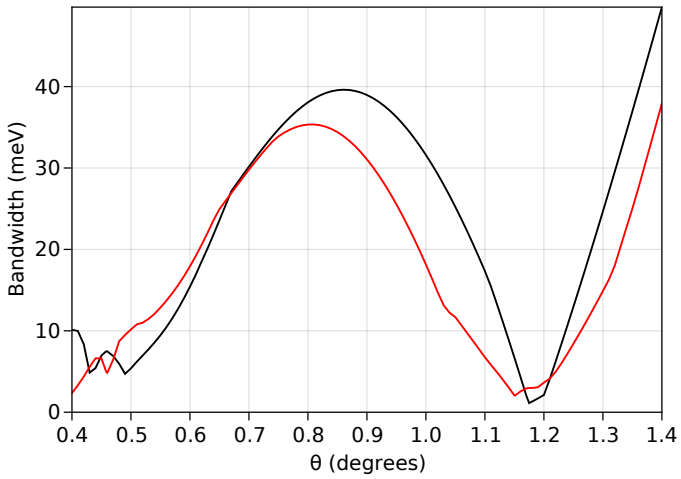
3. **assuming that the functions** $\mathbb{W}_d^{\pm}(\mathbf{X}) - \delta\mu_{\mathrm{F}} I_2$ **are very small**
4. **neglecting the term** $-\frac{\varepsilon_\theta}{2}\Delta$

Band diagrams (left: BM, right: our model)

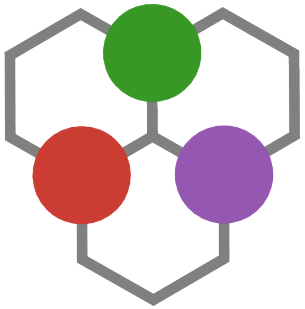
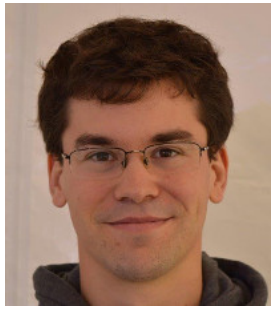






Computation done with DFTK (Density-Functional ToolKit)

- **planewave DFT package in julia (2019-), MIT license**
- **outcome of the EMC2 ERC Synergy project**
- **main developers: Michael Herbst and Antoine Levitt**

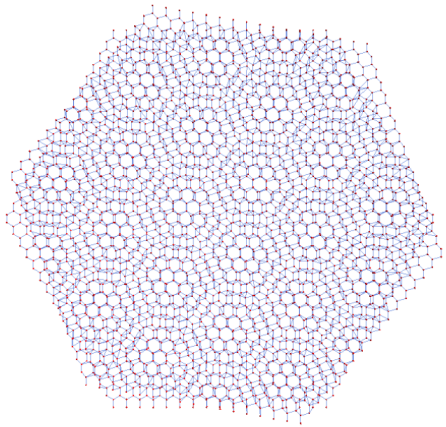




- **supports mathematical developments (low entrance barrier, ∼7k lines of code) and scale-up to relevant applications (∼ 1,000 electrons)**
- **fully composable with julia ecosystem**
  - **arbitrary precision (32bit, 64bit...)**
  - **algorithmic differentiation**
  - **numerical error control**
  - **...**

2 - Mathematical analysis of KS Hamiltonians for moiré materials

Atomic-scale Hamiltonian

$$H_{d,\varepsilon} = -\frac{1}{2}\Delta + v_d(\mathbf{x} - c(\varepsilon)\varepsilon\mathbf{x}, \varepsilon\mathbf{x}, z), \qquad \varepsilon = 2\sin\frac{\theta}{2}, \quad c(\varepsilon) = \frac{1-\sqrt{1-\varepsilon^2}}{\varepsilon} \sim \frac{\varepsilon}{2}$$



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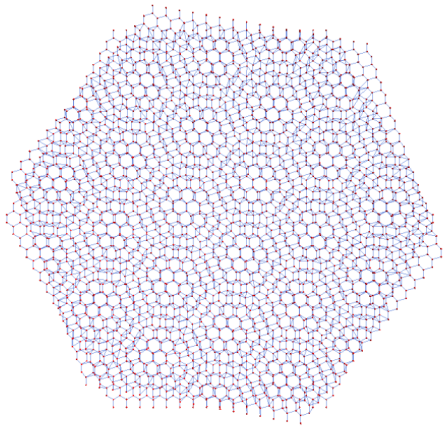
$$H_{d,\varepsilon} = -\frac{1}{2}\Delta + v_d(\mathbf{x} - c(\varepsilon)\varepsilon\mathbf{x}, \varepsilon\mathbf{x}, z), \qquad \varepsilon = 2\sin\frac{\theta}{2}, \quad c(\varepsilon) = \frac{1-\sqrt{1-\varepsilon^2}}{\varepsilon} \sim \frac{\varepsilon}{2}$$



DOS in the negative energy window: **positive Borel measure $\nu_{H_{d,\varepsilon}}$ on $(-\infty, 0)$ such that for $f \in C_c^\infty(\mathbb{R}; \mathbb{R}_+)$ with support in $(-\infty, 0)$,**

$$\underline{\mathrm{Tr}}[f(H_{d,\varepsilon})] = \int_{(-\infty,0)} f(E)\, d\nu_{H_{d,\varepsilon}}(E)$$

Basic idea inspired by Dimassi '99 and Panati-Teufel '03, based on many works going back to Balezard-Konlein '85: approximate $\underline{\mathrm{Tr}}[f(H_{d,\varepsilon})]$ for ε small using semiclassical analysis with operator-valued symbols

Dimassi: ε-expansion of $\mathrm{Tr}(f(H_\varepsilon))$ for

$$H_\varepsilon = \frac{1}{2}(-i\nabla + \mathbf{A}(\varepsilon\mathbf{r}))^2 + V_{\mathrm{per}}(\mathbf{r}) + W(\varepsilon\mathbf{r}) \quad \text{on } L^2(\mathbb{R}^d)$$

with $V_{\mathrm{per}} \in C^\infty(\mathbb{R}^d;\mathbb{R})$ periodic, and e.g. $\mathbf{A} \in C^\infty_{\mathrm{b}}(\mathbb{R}^d;\mathbb{R}^d)$, $W \in C^\infty_{\mathrm{c}}(\mathbb{R};\mathbb{R})$, and f supported in a spectral gap of the periodic operator $-\frac{1}{2}\Delta + V_{\mathrm{per}}$

Panati-Teufel: quantum dynamics in periodic media

$$H_\varepsilon = \frac{1}{2}(-i\nabla + \mathbf{A}(\varepsilon\mathbf{r}))^2 + V_{\mathrm{per}}(\mathbf{r}) + W(\varepsilon\mathbf{r}) \quad \text{on } L^2(\mathbb{R}^d)$$

$\longrightarrow$ **Semiclassical dynamics on Bloch bands with Berry curvature terms**

Atomic-scale Hamiltonian

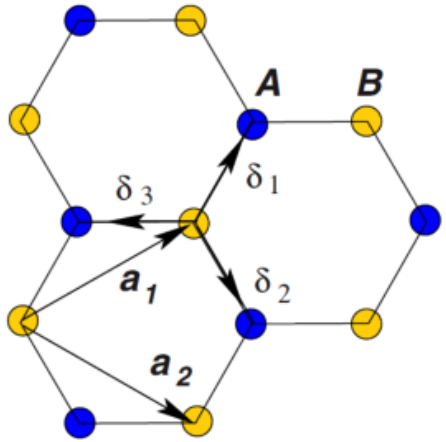
$$H_{d,\varepsilon} = -\frac{1}{2}\Delta + v_d(\mathbf{x} - c(\varepsilon)\varepsilon\mathbf{x}, \varepsilon\mathbf{x}, z), \qquad \varepsilon = 2\sin\frac{\theta}{2}, \quad c(\varepsilon) = \frac{1-\sqrt{1-\varepsilon^2}}{\varepsilon} \sim \frac{\varepsilon}{2}$$

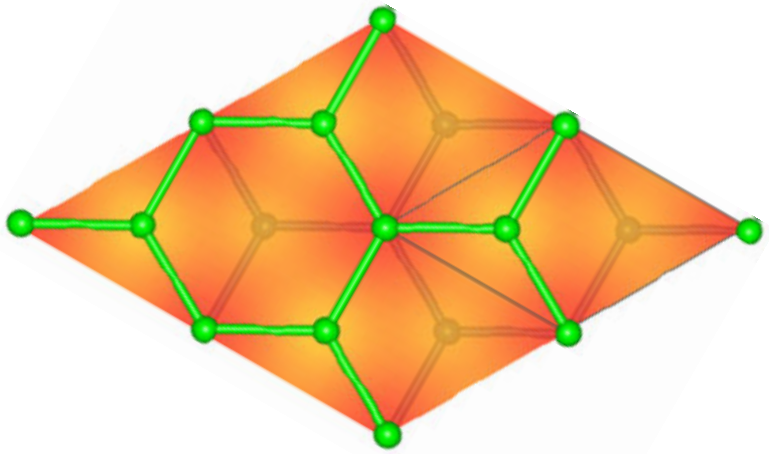
The Hamiltonian

$$h_{d,\mathbf{X}} = -\frac{1}{2}\Delta_{\mathbf{x},z} + v_d(\mathbf{x}, \mathbf{X}, z) \quad \textbf{acting on} \quad L^2(\mathbb{R}^2_{\mathbf{x}} \times \mathbb{R}_z; \mathbb{C})$$

describes an untwisted bilayer graphene with disregistry $\mathbf{y} = J\mathbf{X}$

$\mathbf{y} = 0$**: A-A stacking** $\mathbf{y} = \frac{1}{3}(\mathbf{a}_1 + \mathbf{a}_2)$**: A-B stacking**





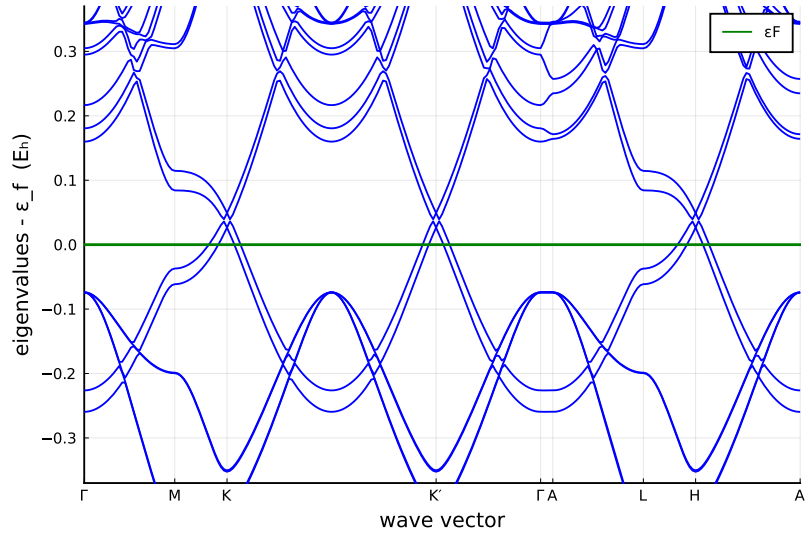
Longitudinal Bloch transform of the $\mathbb{L}$-periodic Hamiltonian $h_{d,\mathbf{X}}$

$$h_{d,\mathbf{X}} = -\frac{1}{2}\Delta_{\mathbf{x},z} + v_d(\mathbf{x},\mathbf{X},z) \quad \textbf{acting on} \quad L^2(\mathbb{R}^2_{\mathbf{x}} \times \mathbb{R}_z;\mathbb{C})$$

$$= \mathcal{U}^{-1}\left(\fint_{\mathrm{BZ}}^{\oplus} h_{d,0}(\mathbf{k},\mathbf{X})\,d\mathbf{k}\right)\mathcal{U} \quad \textbf{with} \quad h_{d,0}(\mathbf{k},\mathbf{X}) := \frac{1}{2}\left(-i\nabla_{\mathbf{x}} + \mathbf{k}\right)^2 - \frac{1}{2}\partial_z^2 + v_d(\cdot,\mathbf{X},\cdot)$$

For each $(\mathbf{k},\mathbf{X}) \in \mathbb{R}^2 \times \mathbb{R}^2$**,**

- $h_{d,0}(\mathbf{k},\mathbf{X})$ **is an operator on** $L^2_{\mathrm{per}} := L^2((\mathbb{R}^2_{\mathbf{x}}/\mathbb{L}) \times \mathbb{R}_z)$
- $\sigma_{\mathrm{c}}(h(\mathbf{k},X)) = \mathbb{R}_+$



Band spectrum of $h(\cdot,\mathbf{X})$ **in the negative energy window**

Semiclassical description of the atomic scale Hamiltonian

$$H_{d,\varepsilon} = -\frac{1}{2}\Delta + v_d(\mathbf{x} - c(\varepsilon)\varepsilon\mathbf{x}, \varepsilon\mathbf{x}, z), \qquad \varepsilon = 2\sin\frac{\theta}{2}, \quad c(\varepsilon) = \frac{1-\sqrt{1-\varepsilon^2}}{\varepsilon} \sim \frac{\varepsilon}{2}$$

Introducing the operator-valued symbol

$$h_{d,\varepsilon}(\mathbf{k}, \mathbf{X}) := \frac{1}{2}\left(-i\nabla_{\mathbf{x}} + \mathbf{k}\right)^2 - \frac{1}{2}\partial_z^2 + v_d(\cdot - c(\varepsilon)\mathbf{X}, \mathbf{X}, \cdot), \qquad (\mathbf{k}, \mathbf{X}) \in \mathbb{R}^2 \times \mathbb{R}^2$$

($h_{d,\varepsilon}(\mathbf{k}, \mathbf{X})$ self-adjoint operator on L^2_{per}), we have

$$\boxed{H_{d,\varepsilon} = \mathcal{U}^{-1}\mathrm{Op}_\varepsilon(h_{d,\varepsilon})\mathcal{U}}$$

where

$$[\mathrm{Op}_\varepsilon(a)\phi]_{\mathbf{k}}(\mathbf{x}, z) = \frac{1}{(2\pi\varepsilon)^2}\int_{\mathbb{R}^2\times\mathbb{R}^2}\left[a\left(\frac{\mathbf{k}+\mathbf{k}'}{2}, \mathbf{X}\right)\phi_{\mathbf{k}'}\right](\mathbf{x}, z)\, e^{-i\frac{(\mathbf{k}-\mathbf{k}')\cdot\mathbf{X}}{\varepsilon}}\, d\mathbf{k}'\, d\mathbf{X}$$

Weyl quantization rule for operator-valued symbols

$$[\mathrm{Op}_\varepsilon(a)\phi]_{\mathbf{k}}(\mathbf{x},z) = \frac{1}{(2\pi\varepsilon)^2}\int_{\mathbb{R}^2\times\mathbb{R}^2}\left[a\left(\frac{\mathbf{k}+\mathbf{k}'}{2},\mathbf{X}\right)\phi_{\mathbf{k}'}\right](\mathbf{x},z)\, e^{-i\frac{(\mathbf{k}-\mathbf{k}')\cdot\mathbf{X}}{\varepsilon}}\, d\mathbf{k}'\, d\mathbf{X}$$

Remarks:

- **compare with usual Weyl quantization formula for symbols in** $C^\infty(\mathbb{R}^d\times\mathbb{R}^d;\mathbb{C})$

$$[\mathrm{Op}_\varepsilon(a)\varphi](\mathbf{r}) = \frac{1}{(2\pi\varepsilon)^d}\int_{\mathbb{R}^d\times\mathbb{R}^d} a\left(\frac{\mathbf{r}+\mathbf{r}'}{2},\mathbf{p}\right)\varphi(\mathbf{r}')\, e^{i\frac{\mathbf{p}\cdot(\mathbf{r}-\mathbf{r}')}{\varepsilon}}\, d\mathbf{r}\, d\mathbf{p}$$

- **assuming that** v_d **is smooth, the operator-valued symbol**

$$h_{d,\varepsilon}(\mathbf{k},\mathbf{X}) := \frac{1}{2}\left(-i\nabla_{\mathbf{x}}+\mathbf{k}\right)^2 - \frac{1}{2}\partial_z^2 + v_d(\cdot - c(\varepsilon)\mathbf{X},\mathbf{X},\cdot), \qquad (\mathbf{k},\mathbf{X})\in\mathbb{R}^2\times\mathbb{R}^2$$

  is in $S^\omega(\mathcal{L}(H^2_{\mathrm{per}};L^2_{\mathrm{per}}))$ **with order function** $\omega(\mathbf{k},\mathbf{X}) = 1+|\mathbf{k}|^2$

- **it is** τ**-equivariant in the sense that it satisfies the property**

$$\forall(\mathbf{k},\mathbf{X})\in\mathbb{R}^2\times\mathbb{R}^2,\quad \forall\mathbf{G}\in\mathbb{L}^*,\quad h_{d,\varepsilon}(\mathbf{k}+\mathbf{G},\mathbf{X}) = \tau_{\mathbf{G}}h_{d,\varepsilon}(\mathbf{k},\mathbf{X})\tau_{-\mathbf{G}}$$

  where

$$\forall u\in H^s_{\mathrm{per}},\quad (\tau_{\mathbf{G}}u)(\mathbf{x},z) = e^{i\mathbf{G}\cdot\mathbf{x}}u(\mathbf{x},z)$$

Theorem (C, Meng, in preparation). Under suitable assumptions on V and V_{int}, for all $f \in C_{\mathrm{c}}^{\infty}(\mathbb{R};\mathbb{R}_+)$ with compact support included in $(-\infty,0)$, it holds

$$\underline{\mathrm{Tr}}[f(H_{d,\varepsilon})] = \sum_{j=0}^{n} \frac{\varepsilon^j}{|\Omega|} \fint_{J\Omega} \fint_{\Omega^*} \mathrm{Tr}_{L^2_{\mathrm{per}}}[f_{d,j}(k,X)]\, dk\, dX + \mathcal{O}(\varepsilon^{n+1})$$

with

$$f_{d,0}(k,X) := f(h_{d,0}(k,X))$$

$$f_{d,1}(k,X) := \frac{i}{2\pi} \int_{\mathbb{C}} \overline{\partial}\widetilde{f}(\zeta) \left[\{(\zeta - h_{d,0})^{-1}, (\zeta - h_{d,0})\}(\zeta - h_{d,0})^{-1}\right](k,X)\, d^2\zeta$$

$$\dots$$

where $\widetilde{f} : \mathbb{C} \to \mathbb{C}$ is any almost analytic extension of f

Tools for the proof: semiclassical analysis with operator-valued symbols, Helffer-Sjöstrand formula, (degenerate) perturbation theory, "twisted" Weyl calculus to deal with the $-c(\varepsilon)\mathrm{X}$ term in $v_d(\cdot - c(\varepsilon)\mathrm{X}, \mathrm{X}, \cdot)$

Theorem (C, Meng, in preparation). Under suitable assumptions on V and V_{int}, for all $f \in C_{\mathrm{c}}^{\infty}(\mathbb{R};\mathbb{R}_+)$ with compact support included in $(-1,1)$, $\delta,\varepsilon>0$ small enough, $d>0$ large enough,

$$\underline{\mathrm{Tr}}\left(f\left(\frac{H_{d,\varepsilon}-\mu_{\mathrm{F}}}{\delta}\right)\right) = 2\underline{\mathrm{Tr}}\left(f\left(\frac{T_{d,K,\varepsilon}}{\delta}\right)\right) + \mathcal{O}(\delta^{-1}\varepsilon) + \delta^{1-}e_d + \mathcal{O}(\delta^{3-})$$

with

$$T_{d,\mathbf{K},\varepsilon} := T(-i\nabla_x - \mathbf{K}) + \mathcal{V}_{d,\mathbf{K}}(\varepsilon\mathbf{x}) \quad \textbf{acting on } L^2(\mathbb{R}^2;\mathbb{C}^4),$$

where

$$T(\mathbf{k}) := \begin{pmatrix} v_{\mathrm{F}}\boldsymbol{\sigma}\cdot\mathbf{k} & 0 \\ 0 & v_{\mathrm{F}}\boldsymbol{\sigma}\cdot\mathbf{k}\end{pmatrix}, \qquad \mathcal{V}_{d,\mathbf{K}}(\mathbf{X}) := \begin{pmatrix} \mathbb{W}^{+}_{d,\mathbf{K}}(\mathbf{X}) & \mathbb{V}_{d,\mathbf{K}}(\mathbf{X}) \\ \mathbb{V}_{d,\mathbf{K}}(\mathbf{X})^* & \mathbb{W}^{-}_{d,\mathbf{K}}(\mathbf{X})\end{pmatrix}$$

and $e_d = f(V, V_{\mathrm{int}}, u_j, d)$ computable, converging to zero when $d \to +\infty$

Conclusion and perspectives

Towards first-principle models for moiré materials

- first attempt to derive an effective model for moiré materials directly from Kohn-Sham Hamiltonians
- bypass tight-binding models and use instead tools from multiscale analysis

Extensions (work in progress)

- mathematical analysis with semiclassical techniques
- lattice relaxation: straightforward if the displacement field is given
- coupling with phonons