Electronic Structure of Mechanically Relaxed Incommensurate Materials using Momentum Space

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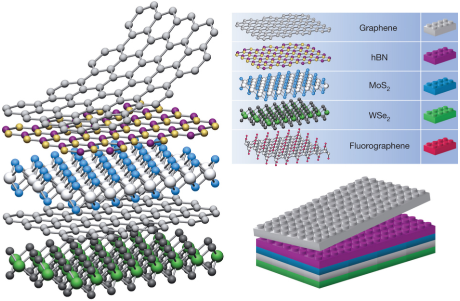
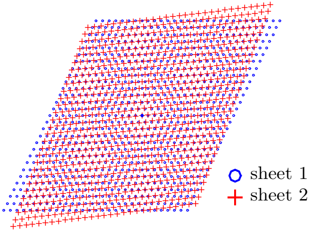
- Periodic structures studied via Bloch theory.
- Incommensurate stacking breaks periodicity.
- One approach is via configuration space (real space) techniques.
- Here we focus on momentum space techniques ($k.p$).

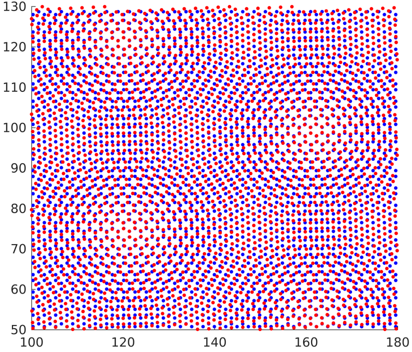


[Geim, 2013]

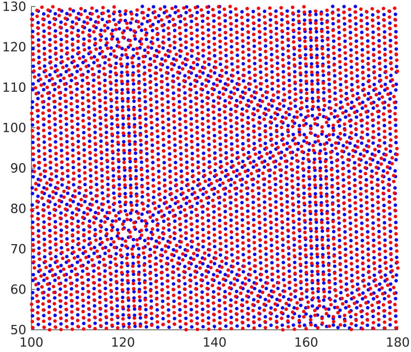


Incommensurate bilayer

- When two layers are closely aligned, they form large-scale moiré patterns.
- This leads to slowly varying long-range atomistic relaxation.



Unrelaxed

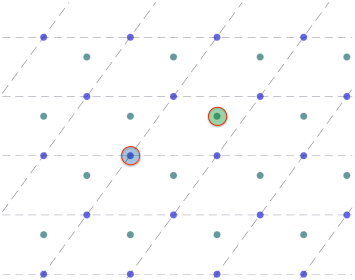


Relaxed

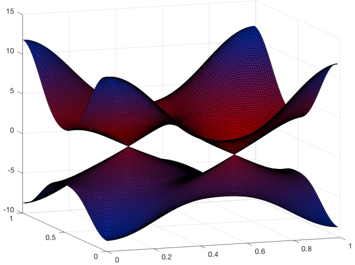
- Real space tight-binding models are effective at understanding a wide variety of systems.
- Momentum space ($k.p$) bring important physical insight and frequently are less expensive.
- We introduce a framework to rigorously build momentum space models by direct transformation of real space models.
- We show observables like density of states (DoS) are identical under transformation from real space to momentum space.
- We apply this technique to generate a momentum space model for mechanically relaxed incommensurate bilayer systems.

- In a tight-binding framework, there are a finite number of degrees of freedom, or orbitals $\mathcal{A}$, per unit cell.
- For Bravais lattice $\mathcal{R} := \{An : n \in \mathbb{Z}^2\}$ (A is a 2×2 matrix), we have degree of freedom space $\Omega := \mathcal{R} \times \mathcal{A}$.

- Hamiltonian is exponentially localized in hopping and periodic.
- $[H\psi]_{R\alpha} = \sum_{R\alpha \in \Omega} H_{R\alpha, R'\alpha'} \psi_{R'\alpha'}$.
- $H_{R\alpha, R'\alpha'} = h_{\alpha\alpha'}(R - R')$.
- $|h_{\alpha\alpha'}(R)| \lesssim e^{-\gamma|R|}$.



- We Bloch transform real space to momentum space. Γ^* is the reciprocal lattice unit cell.
- $[\mathcal{G}\psi]_\alpha(q) = |\Gamma^*|^{-1/2} \sum_{R\in\mathcal{R}} \psi_{R\alpha} e^{-iq\cdot R}$.
- $H\psi = h * \psi$, so $[\mathcal{G}H\psi](q) = |\Gamma^*|^{1/2}[\mathcal{G}h](q)[\mathcal{G}\psi](q)$.
- The transformed H is given by $\mathcal{G}H\mathcal{G}^* = \mathcal{G}h$.
- $\sigma_{\text{cont}}(H) = \bigcup_{q\in\Gamma^*} \sigma_{\text{point}}\big([\mathcal{G}h](q)\big)$.



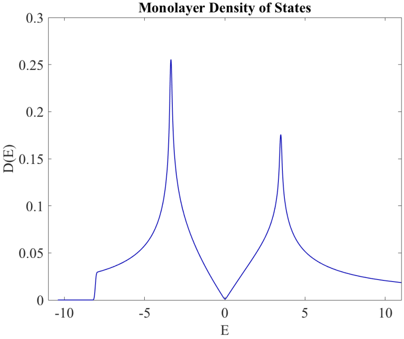
Density of States, Monolayer

- Heuristically, density of states of a system is a normalized trace, $D(E) = \bar{\text{Tr}}\delta(E - H)$.
- Instead of using the delta function, we use arbitrary polynomial function g, typically a thin Chebyshev polynomial approximation to a Gaussian. By periodicity we have

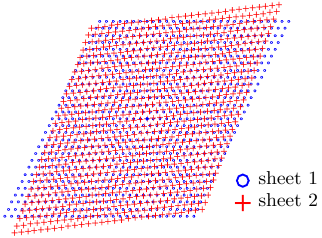
$$\int g(E)D(E)dE = |\mathcal{A}|^{-1} \sum_{\alpha\in\mathcal{A}} e_{0\alpha}^* g(H) e_{0\alpha}.$$

- $e_{0\alpha} \in \ell^1(\Omega)$ is the standard basis vector centered at $0\alpha \in \Omega$.

- We can transform observables, here density of states used as an example, to momentum space directly.
- We use that $\mathcal{G}^*\mathcal{G} = I$ is the identity over $\ell^2(\Omega)$.
- $e_{0\alpha}^* g(H) e_{0\alpha} = (\mathcal{G}e_{0\alpha})^* g(\mathcal{G}H\mathcal{G}^*)(\mathcal{G}e_{0\alpha}) = \fint_{\Gamma*} e_\alpha^* g(\mathcal{G}h(q))e_\alpha dq.$
- Hence $\int g(E)D(E)dE = |\mathcal{A}|^{-1}\sum_{\alpha\in\mathcal{A}} \fint_{\Gamma*} e_\alpha^* g(\mathcal{G}h(q))e_\alpha dq.$



- For bilayer, we have $\Omega_j = \mathcal{R}_j \times \mathcal{A}_j$.
- $\Omega = \Omega_1 \cup \Omega_2$.
- $H : \ell^1(\Omega) \to \ell^1(\Omega)$.
- $H_{R\alpha, R'\alpha'} = h_{\alpha\alpha'}(R - R')$.
- $h_{\alpha\alpha'} : \mathbb{R}^2 \to \mathbb{C}$, $|h_{\alpha\alpha'}(x)| \lesssim e^{-\gamma'|x|}$.



Incommensurate bilayer

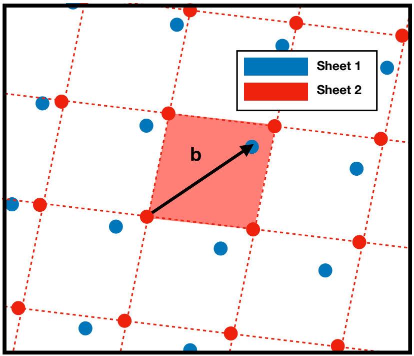
- Let $\mathcal{R}_1^*$ and $\mathcal{R}_2^*$ denote reciprocal lattices.
- We assume both the lattices and reciprocal lattices are incommensurate, i.e.

$$\mathcal{R}_1 \cup \mathcal{R}_2 + v = \mathcal{R}_1 \cup \mathcal{R}_2 \text{ iff } v = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$

$$\mathcal{R}_1^* \cup \mathcal{R}_2^* + v = \mathcal{R}_1^* \cup \mathcal{R}_2^* \text{ iff } v = \begin{pmatrix} 0 \\ 0 \end{pmatrix}.$$

Configuration

- Here we discuss configuration for the purpose of understanding mechanical relaxation.
- We let $\Gamma_j := A_j[0,1)^2$ be sheet j's unit cell, where A_j are the 2×2 lattice matrices.
- All lattice sites can be parameterized by configuration (b), or over compact Γ_1 and Γ_2.
- Example: $R \in \mathcal{R}_1$ parameterized by $b = R - R' \in \Gamma_2$, $R' \in \mathcal{R}_2$.



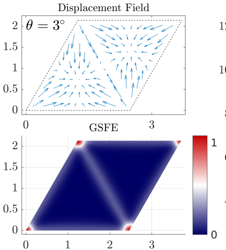
- Using a linear elastic model, we can find functions

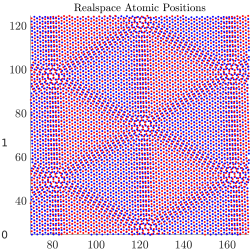
$$u_1 \in C_{\mathrm{per}}(\Gamma_2; \mathbb{R}^2) \qquad\qquad u_2 \in C_{\mathrm{per}}(\Gamma_1; \mathbb{R}^2).$$

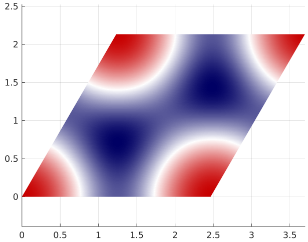
such that $\mathcal{R}_j \to \mathcal{R}_j + u_j(R_j)$. We periodically extend u_1 and u_2.

- For $R\alpha \in \Omega_k$ and $R'\alpha' \in \Omega_j$, we have

$$[H^u]_{R\alpha,R'\alpha'} = h_{\alpha\alpha'}(R + u_k(R) - R' - u_j(R')).$$







- Define the Bloch transform for each sheet:

$$[\widehat{\mathcal{G}}_j\psi_j]_\alpha(q) = |\Gamma_j^*|^{-1/2} \sum_{R_j \in \mathcal{R}_j} \psi_{R_j\alpha} e^{-iR_j\cdot q}.$$

- For $\psi = (\psi_1, \psi_2) \in \ell^1(\Omega)$,

$$\mathcal{G}_1\psi = (\widehat{\mathcal{G}}_1\psi_1, 0), \qquad \mathcal{G}_2\psi = (0, \widehat{\mathcal{G}}_2\psi_2).$$

- Define the bilayer Bloch transform as

$$\mathcal{G}\psi := \mathcal{G}_1\psi + \mathcal{G}_2\psi = (\widehat{\mathcal{G}}_1\psi_1, \widehat{\mathcal{G}}_2\psi_2).$$

- Let $\mathcal{P}_j$ be the projection onto sheet j.
- Let $H_{ij} = \mathcal{P}_i H \mathcal{P}_j$. Note that $\sum_{ij} H_{ij} = H$.

Proposition

We have for $\psi \in \ell^1(\Omega)$ *that*

$$\begin{aligned}
\mathcal{G}_1 H_{11}\psi(q) &= c_1 \mathcal{G}_1 h(q) \mathcal{G}_1 \psi(q), \\
\mathcal{G}_2 H_{22}\psi(q) &= c_2 \mathcal{G}_2 h(q) \mathcal{G}_2 \psi(q), \\
\mathcal{G}_1 H_{12}\psi(q) &= \sum_{K \in \mathcal{R}_1^*} c_0 \hat{h}^{12}(q+K) \mathcal{G}_2 \psi(q+K), \\
\mathcal{G}_2 H_{21}\psi(q) &= \sum_{K \in \mathcal{R}_2^*} c_0 \hat{h}^{21}(q+K) \mathcal{G}_1 \psi(q+K),
\end{aligned}$$

where $c_j = |\Gamma_j^*|^{1/2}, \qquad c_0 = c_1 \cdot c_2.$

- Let T_K be the translation by $K \in \mathcal{R}_j^*$. Recall $\mathcal{G}H\mathcal{G}^*$ defined over $\bigotimes_{j=1}^2 C_{\text{per}}(\Gamma_j^*; \mathbb{C}^{\mathcal{A}_j})$.

$$\mathcal{G}H\mathcal{G}^* = \begin{pmatrix} c_1\mathcal{G}h_{11} & 0 \\ 0 & c_2\mathcal{G}h_{22} \end{pmatrix} + \sum_{K\in\mathcal{R}_1^*} H_{K2}T_K + \sum_{K\in\mathcal{R}_2^*} H_{K1}T_K.$$

- H_{Kj}'s are the hopping terms:

$$H_{K1}(q) = \begin{pmatrix} 0 & 0 \\ c_0\hat{h}^{21}(q+K) & 0 \end{pmatrix}, \qquad H_{K2}(q) = \begin{pmatrix} 0 & c_0\hat{h}^{12}(q+K) \\ 0 & 0 \end{pmatrix}.$$

Momentum Space Hamiltonian

- Define the momentum Hamiltonian centered at wavenumber $q \in \Gamma_j^*$:

$$[\widehat{H}(q)]_{K\alpha,K'\alpha'} = c_0 \hat{h}_{\alpha\alpha'}(q + K + K'), \qquad \text{for interlayer,}$$
$$[\widehat{H}(q)]_{K\alpha,K\alpha'} = c_j \mathcal{G}_j h_{\alpha\alpha'}(q + K), \qquad \text{for intralayer.}$$

- We then have the following theorem based on the ergodicity property:

Theorem

For incommensurate system, we have

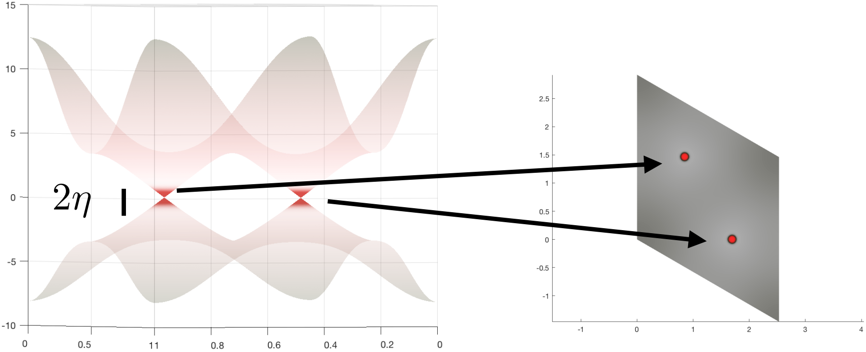
$$\int g(E)D(E)dE = \nu^* \sum_{j=1}^{2} \sum_{\alpha\in\mathcal{A}_j} \int_{\Gamma_j^*} e_\alpha^* g\big(\widehat{H}(q)\big) e_\alpha dq.$$

where ν^* is the normalization constant.

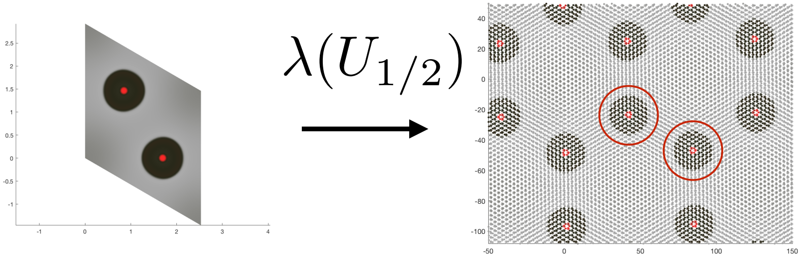
Hamiltonian cut-off

- Here we wish to find a reduced matrix approximation.
- We define maximum interlayer coupling η (for graphene, let $E_0 = 0$):

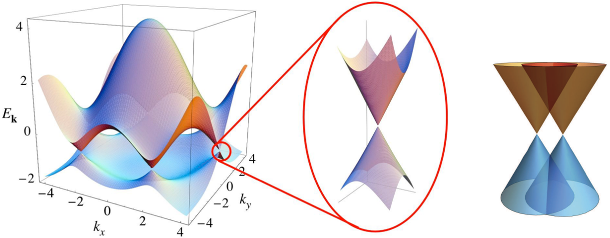
$$\eta = \|\widehat{H}_{12}\|_{\mathrm{op}},$$
$$U_0 := \{q' \in \mathbb{R}^2 : \eta^{-1}\|E_0 I - \mathcal{G}_k h(q')\|_{\mathrm{op}} < \beta < 1\},$$
$$U_r := U_0 + B_r(0).$$



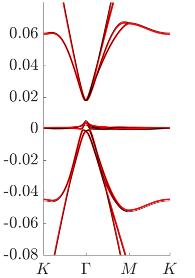
- $\widehat{H}(q)$ has diagonal blocks of the form $\mathcal{G}_j h(q+K)$.
- Under the assumption $\theta := \|A_2^{-T} - A_1^{-T}\|_{\text{op}} \ll 1$, diagonal blocks vary slowly.
- This gives us mapping λ (q-dependent) from $\mathbb{R}^2$ to subsets of Ω^*.
- Restrict $\widehat{H}(q)$ to small number of reciprocal lattice sites.



Graphene Band Structure



[*The electronic properties of graphene*, Neto, Geim, et al, *Rev. Mod. Phys*]
[*Correlated insulator behaviour at half-filling in magic-angle graphene superlattices*, Cao, Jarillo-Herroro, et al, *Nature*]



1.085° degree twist, magic angle

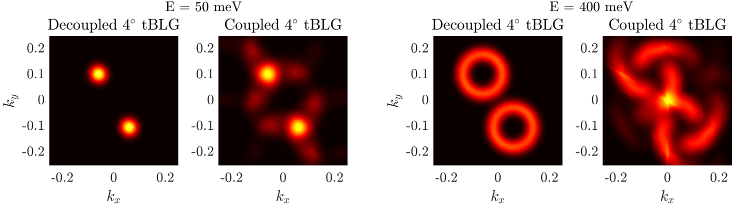
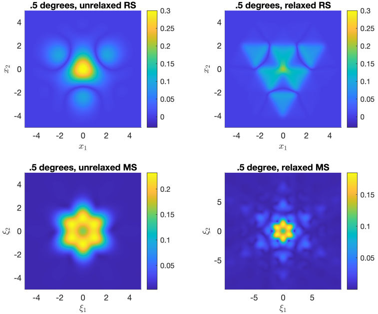


Figure: tBLG, plotting $e_\alpha^* \delta(E - \widehat{H}(q)) e_\alpha$.

- We wish to find $\mathcal{G} H^u \mathcal{G}^*$ to get relaxed system.
- If we ignore relaxation effects on intralayer, then it is easy.
- $h^u_{\alpha\alpha'}(x) = h_{\alpha\alpha'}(x + u_1(x) - u_2(-x))$ for sheet $2 \to 1$ *real space* interlayer coupling.
- $c_0 \hat{h}^u_{\alpha\alpha'}(\xi)$ for *momentum space* interlayer coupling.



- Intralayer terms can be modeled to first order as $[H^u]_{R\alpha,R'\alpha'} = h_{\alpha\alpha'}(u_j(R) - u_j(R') + (R - R'))$.
- This form won't Bloch transform however, so we find an appropriate approximate system:

$$
\begin{aligned}
[H^u]_{R\alpha,R'\alpha'} &\approx [\tilde{H}^u]_{R\alpha,R'\alpha'} \\
[\tilde{H}^u]_{R\alpha,R'\alpha'} &:= h_{\alpha\alpha'}(R - R') + \\
&\qquad \nabla h_{\alpha\alpha'}(R - R') \cdot \underbrace{\nabla u_j(R)}_{\text{breaks periodicity}} \Lambda_j(R - R').
\end{aligned}
$$

- $\Lambda_1 = I - A_2 A_1^{-1}$ and $\Lambda_2 = I - A_1 A_2^{-1}$.
- Here intralayer terms produce scattering.

- Let $P_1 = 2$ and $P_2 = 1$.
- We can take a Fourier expansion of $u_j(b)$, $K \in \mathcal{R}_{P_j}$:

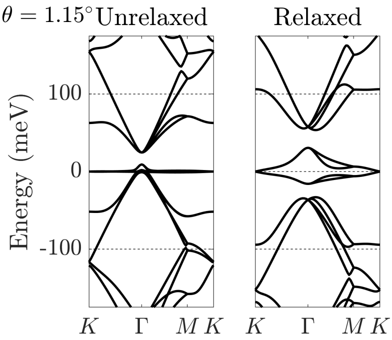
$$\hat{u}_j^K = |\Gamma_{P_j}^*|^{-1/2} \int_{\Gamma_{P_j}^*} u_j(b) e^{iK\cdot b},$$

$$-i\hat{u}_j^K \otimes K = |\Gamma_{P_j}^*|^{-1/2} \int_{\Gamma_{P_j}^*} \nabla u_j(b) e^{iK\cdot b}.$$

- $[\tilde{H}^u]_{R\alpha,R'\alpha'} = h_{\alpha\alpha'}(R - R') - \sum_{K\in\mathcal{R}_{P_j}^*} ie^{-iK\cdot R}(\hat{u}_j^K \otimes K) : s_{\alpha\alpha'}^j(R - R')$.
- $s_{\alpha\alpha'}^j(R - R') := \nabla h_{\alpha\alpha'}(R - R') \otimes \Lambda_j(R - R')$.

Approximate Momentum Space Relaxed Hamiltonian

- Mechanical relaxation opens the gaps.
- This approximate Hamiltonian has corresponding momentum space Hamiltonian $\widehat{H}^u$ defined below.



$$[\widehat{H}^u(q)]_{K\alpha,K'\alpha'} = c_0 \hat{h}^u_{\alpha\alpha'}(q+K+K'), \qquad \text{for interlayer,}$$

$$[\widehat{H}^u(q)]_{K\alpha,K'\alpha'} = c_j \mathcal{G}_j h_{\alpha\alpha'}(q+K)\delta_{KK'} - i c_j \hat{u}_j^{K+K'} \otimes (K+K') : \mathcal{G}_j s_{\alpha\alpha'}(q+K+K'), \qquad \text{for intralayer.}$$

Conclusion

- Real space systems are easier to model and are versatile, while momentum space give more physical information and are often computationally faster.
- We introduce a methodology for transforming real space systems into momentum space and show observables such as density of states are preserved.
- We apply this approximately to mechanically relaxed bilayers.
- This can be extended to more complex systems such as multilayers and observables such as conductivity.