Electronic Structure of Mechanically Relaxed Incommensurate Materials using Momentum Space

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Incommensurate 2D Materials

- 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.
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 $2 \times 2$ matrix), we have degree of freedom space $\Omega := \mathcal{R} \times \mathcal{A}$.

Hamiltonian is exponentially localized in hopping and periodic.

\[
\begin{align*}
[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)| &\leq e^{-\gamma|R|}.
\end{align*}
\]
We Bloch transform real space to momentum space. $\Gamma^*$ 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 $\mathcal{G}H\mathcal{G}^* = \mathcal{G} h$.

\[
\sigma_{\text{cont}}(H) = \bigcup_{q \in \Gamma^*} \sigma_{\text{point}}([\mathcal{G} h](q)).
\]
Heuristically, density of states of a system is a normalized trace,
\[ D(E) = \overline{\text{Tr}} \delta(E - H). \]

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

\( e_{0\alpha} \in \ell^1(\Omega) \) 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 $\ell^2(\Omega)$.

$e_{0\alpha}^* g(H)e_{0\alpha} = (\mathcal{G} e_{0\alpha})^* g(\mathcal{G} H \mathcal{G}^*) (\mathcal{G} e_{0\alpha}) = \int_{\Gamma^*} e_{\alpha}^* g(\mathcal{G} h(q)) e_{\alpha} dq$.

Hence $\int g(E) D(E) dE = |\mathcal{A}|^{-1} \sum_{\alpha \in \mathcal{A}} \int_{\Gamma^*} e_{\alpha}^* g(\mathcal{G} h(q)) e_{\alpha} dq$. 

![Monolayer Density of States](image)
Incommensurate Bilayer

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

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

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

$$\mathcal{R}_1^* \cup \mathcal{R}_2^* + \nu = \mathcal{R}_1^* \cup \mathcal{R}_2^* \text{ iff } \nu = \begin{pmatrix} 0 \\ 0 \end{pmatrix}.$$
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 $2 \times 2$ lattice matrices.

All lattice sites can be parameterized by configuration $(b)$, or over compact $\Gamma_1$ and $\Gamma_2$.

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

\[ u_1 \in C_{\text{per}}(\Gamma_2; \mathbb{R}^2) \quad u_2 \in C_{\text{per}}(\Gamma_1; \mathbb{R}^2). \]

such that \( R_j \rightarrow R_j + u_j(R_j) \). 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:

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

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

\[
\mathcal{G}_1 \psi = (\hat{G}_1 \psi_1, 0), \quad \mathcal{G}_2 \psi = (0, \hat{G}_2 \psi_2).
\]

Define the bilayer Bloch transform as

\[
\mathcal{G} \psi := \mathcal{G}_1 \psi + \mathcal{G}_2 \psi = (\hat{G}_1 \psi_1, \hat{G}_2 \psi_2).
\]
Let \( P_j \) be the projection onto sheet \( j \).

Let \( H_{ij} = P_i H P_j \). Note that \( \sum_{ij} H_{ij} = H \).

**Proposition**

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

\[
\begin{align*}
G_1 H_{11} \psi(q) &= c_1 G_1 h(q) G_1 \psi(q), \\
G_2 H_{22} \psi(q) &= c_2 G_2 h(q) G_2 \psi(q), \\
G_1 H_{12} \psi(q) &= \sum_{K \in R_1^*} c_0 \hat{h}^{12}(q + K) G_2 \psi(q + K), \\
G_2 H_{21} \psi(q) &= \sum_{K \in R_2^*} c_0 \hat{h}^{21}(q + K) G_1 \psi(q + K),
\end{align*}
\]

where \( c_j = |\Gamma_j^*|^{1/2} \), \( c_0 = c_1 \cdot c_2 \).
Let $T_K$ be the translation by $K \in \mathcal{R}_j^*$. Recall $GHG^*$ defined over $\bigotimes_{j=1}^2 C_{\text{per}}(\Gamma_j^*; \mathbb{C} A_j)$.

\[
GHG^* = \begin{pmatrix} c_1G h_{11} & 0 \\ 0 & c_2G h_{22} \end{pmatrix} + \sum_{K \in \mathcal{R}_1^*} H_{K_2} T_K + \sum_{K \in \mathcal{R}_2^*} H_{K_1} T_K.
\]

$H_{K_j}$’s are the hopping terms:

\[
H_{K_1}(q) = \begin{pmatrix} 0 & 0 \\ c_0 \hat{h}_{21}(q + K) & 0 \end{pmatrix}, \quad H_{K_2}(q) = \begin{pmatrix} 0 & c_0 \hat{h}_{12}(q + K) \\ 0 & 0 \end{pmatrix}.
\]
Define the momentum Hamiltonian centered at wavenumber \( q \in \Gamma_j^* \):

\[
\begin{align*}
[\hat{H}(q)]_{K_\alpha, K'_{\alpha'}} &= c_0 \hat{h}_{\alpha'\alpha} (q + K + K'), \\
[\hat{H}(q)]_{K_\alpha, K_{\alpha'}} &= c_j G_j \hat{h}_{\alpha'\alpha} (q + K),
\end{align*}
\]

for interlayer, and

for intralayer.

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

**Theorem**

For incommensurate system, we have

\[
\int g(E) D(E) dE = \nu^* \sum_{j=1}^{2} \sum_{\alpha \in A_j} \int_{\Gamma_j^*} e_{\alpha}^* g(\hat{H}(q)) e_{\alpha} dq.
\]

where \( \nu^* \) is the normalization constant.
Here we wish to find a reduced matrix approximation.

- We define maximum interlayer coupling $\eta$ (for graphene, let $E_0 = 0$):
  \[
  \eta = \| \hat{H}_{12} \|_{\text{op}},
  \]
  \[
  U_0 := \{ q' \in \mathbb{R}^2 : \eta^{-1} \| E_0 I - G_k h(q') \|_{\text{op}} < \beta < 1 \},
  \]
  \[
  U_r := U_0 + B_r(0).
  \]
\( \hat{H}(q) \) has diagonal blocks of the form \( G_j h(q + K) \).

Under the assumption \( \theta := \| A_2^T - A_1^T \|_{op} \ll 1 \), diagonal blocks vary slowly.

This gives us mapping \( \lambda \) (\( q \)-dependent) from \( \mathbb{R}^2 \) to subsets of \( \Omega^* \).

Restrict \( \hat{H}(q) \) 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-Herrero, et al, Nature]

1.085° degree twist, magic angle

Figure: tBLG, plotting $e_{\alpha}^{*} \delta(E - \hat{H}(q)) e_{\alpha}$. 
We wish to find $G H^u G^*$ to get relaxed system.

If we ignore relaxation effects on intralayer, then it is easy.

$$h_{\alpha\alpha'}^u(x) = h_{\alpha\alpha'}(x + u_1(x) - u_2(-x))$$ for sheet $2 \rightarrow 1$ real space interlayer coupling.

$c_0 \hat{h}_{\alpha\alpha'}^u(\xi)$ for momentum space interlayer coupling.
Intralayer terms

- 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:

\[
[H^u]_{R\alpha, R'\alpha'} \approx [\tilde{H}^u]_{R\alpha, R'\alpha'} \\
[\tilde{H}^u]_{R\alpha, R'\alpha'} := h_{\alpha\alpha'}(R - R') + (R - R') \cdot \nabla u_j(R) \cdot \Lambda_j(R - R').
\]

- \( \Lambda_1 = I - A_2A_1^{-1} \) and \( \Lambda_2 = I - A_1A_2^{-1} \).

- Here intralayer terms produce scattering.
Let $P_1 = 2$ and $P_2 = 1$.

We can take a Fourier expansion of $u_j(b)$, $K \in \mathcal{R}_{P_j}$:

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

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

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

$$s^j_{\alpha\alpha'}(R - R') := \nabla h_{\alpha\alpha'}(R - R') \otimes \Lambda_j(R - R').$$
Mechanical relaxation opens the gaps.

This approximate Hamiltonian has corresponding momentum space Hamiltonian \( \hat{H}^u \) defined below.

\[
\begin{align*}
[\hat{H}^u(q)]_{K\alpha,K'\alpha'} & = c_0 \hat{h}^u_{\alpha\alpha'}(q + K + K'), \\
[\hat{H}^u(q)]_{K\alpha,K'\alpha'} & = c_j \mathcal{G}_j h_{\alpha\alpha'}(q + K) \delta_{KK'} \\
& - ic_j \hat{u}^K_{j} \otimes (K + K') : \mathcal{G}_j \mathcal{s}_{\alpha\alpha'}(q + K + K'),
\end{align*}
\]

for interlayer, for intralayer.
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.