Electronic Density of States for Incommensurate Layers

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2D materials can be stacked and rotated.

This leads to incommensurate systems.

Our new method can approximate the Density of States for incommensurate structures that cannot be approximated by supercells (such as low angle rotations).
Commensurate Approximation

Incommensurate system.

Rotate blue lattice to make commensurate.
Main Results

- Rigorously define the Density of States (DoS) for incommensurate systems.
- Derive an efficient algorithm to calculate the DoS and Local Density of States (LDoS) for incommensurate systems.
- Derive error bounds, which allows controlling parameter selection to optimize the accuracy and efficiency of the approximation.
Lattices defined by $2 \times 2$ invertible matrices $A_j$:

$$\mathcal{R}_j = \{ A_j n : n \in \mathbb{Z}^2 \}.$$ 

We assume $\mathcal{R}_1$ and $\mathcal{R}_2$ are *incommensurate*, or for $v \in \mathbb{R}^2$,

$$v + \mathcal{R}_1 \cup \mathcal{R}_2 = \mathcal{R}_1 \cup \mathcal{R}_2$$

$$\iff v = \begin{pmatrix} 0 \\ 0 \end{pmatrix}.$$ 

We will be interested when the reciprocal lattices are incommensurate,

$$\mathcal{R}_j^* = \{ 2\pi A_j^{-T} n : n \in \mathbb{Z}^2 \}.$$
• $\mathcal{A}_j$ are orbital index sets.
• $\Omega = (\mathcal{R}_1 \times \mathcal{A}_1) \cup (\mathcal{R}_2 \times \mathcal{A}_2)$.
• $H_{\mathcal{R}\alpha, \mathcal{R}'\alpha'} = h_{\alpha\alpha'}(R - R')$.
• Orbital interactions $h_{\alpha\alpha'}$ are uniformly continuous on $\mathbb{R}^2$.
• They decay exponentially $(r \in \mathbb{R}^2)$:
  \[ |h_{\alpha\alpha'}(r)| \leq C e^{-\tilde{\gamma}|r|}. \]
Finite Matrix Approximation

- Let $\tilde{\Omega} \subset \Omega$ be finite.
- The associated hamiltonian is $\tilde{H} = (H_{ij})_{i,j \in \tilde{\Omega}}$. 
Density of States (DoS)

- For eigenvalues \( \{ \epsilon_s \}_{s \in \tilde{\Omega}} \), the *density of states* is
  \[
  \text{DoS}(\epsilon) = \frac{1}{\#\tilde{\Omega}} \sum_s \delta(\epsilon - \epsilon_s).
  \]

- DoS can be defined weakly, \( g \) analytic:
  \[
  \mathcal{D}[\tilde{H}](g) = \frac{1}{\#\tilde{\Omega}} \text{Tr}[g(\tilde{H})] = \int g(\epsilon)\text{DoS}(\epsilon)d\epsilon.
  \]

- The thermodynamic limit is weakly defined by
  \[
  \mathcal{D}[H](g) = \lim_{\tilde{\Omega} \uparrow \Omega} \mathcal{D}[\tilde{H}](g).
  \]

- DoS can be broken into site contributions:
  \[
  \mathcal{D}[\tilde{H}](g) = \frac{1}{\#\tilde{\Omega}} \sum_{k \in \tilde{\Omega}} [g(\tilde{H})]_{kk}.
  \]
Local Configuration Sampling
The local geometry of site $R_1 \in \mathcal{R}_1$ is defined by
\[ \mathcal{R}_1 \cup \mathcal{R}_2 - R_1 = \mathcal{R}_1 \cup (\mathcal{R}_2 - R_1) = \mathcal{R}_1 \cup (\mathcal{R}_2 - \text{mod}_2(R_1)). \]

$\text{mod}_2(R_1) \in \Gamma_2 := \{A_2 \alpha : \alpha \in [0, 1)^2\}$. 
Equidistribution of Local Configurations

Theorem

Consider $\mathcal{R}_1$ and $\mathcal{R}_2$ such that their reciprocal lattices are incommensurate. Then for $g \in C_{\text{per}}(\Gamma_2)$, we have

$$\frac{1}{\#(\mathcal{R}_1 \cap B_r)} \sum_{\ell \in \mathcal{R}_1 \cap B_r} g(\ell) \to \frac{1}{|\Gamma_2|} \int_{\Gamma_2} g(b) db.$$  \hspace{1cm} (1)

- We will calculate site contribution to Density of States.
- Integrate over $\Gamma_j$'s for all site contributions.
DoS Approximation

- $H_{r,1}(b)$ is defined over $\mathcal{R}_1 \cap B_r$ and $\mathcal{R}_2 \cap B_r + b$, $b \in \Gamma_1$.
- LDoS for sheet 2 can be defined as

$$D_\alpha[H](b, g) = \lim_{r \to \infty} [g(H_{r,1}(b))]_{0,0\alpha}, \quad \alpha \in A_2.$$ 

**Theorem**

$$D[H](g) = \nu \left( \sum_{\alpha \in A_1} \int_{\Gamma_2} D_\alpha[H](b, g) \, db + \sum_{\alpha \in A_2} \int_{\Gamma_1} D_\alpha[H](b, g) \, db \right)$$

where

$$\nu = \frac{1}{|A_2| \cdot |\Gamma_1| + |A_1| \cdot |\Gamma_2|}.$$
The Approximation

- For $g_{\epsilon, \eta}(\xi) = \frac{1}{\sqrt{2\pi \eta}} e^{-(\xi-\epsilon)^2/2\eta^2}$, we have the final approximation

$$D_\eta(\epsilon) := \nu \left( \sum_{\alpha \in A_1} \int_{\Gamma_2} [g_{\epsilon, \eta}(H_{r,2}(b))]_{0\alpha,0\alpha} db + \sum_{\alpha \in A_2} \int_{\Gamma_1} [g_{\epsilon, \eta}(H_{r,1}(b))]_{0\alpha,0\alpha} db \right).$$

- For $h$ smooth, we uniformly discretize the integrals.
Resolvent bounds yield an exponential decay rate:

**Theorem**

For $\alpha \in A_1$ and $h_{\alpha \alpha'}$ exponentially decaying, we have

$$|D_{\alpha}[H](b, g_{\epsilon, \eta}) - [g_{\epsilon, \eta}(H_{r,1}(b))]_{0\alpha, 0\alpha}| \lesssim \eta^{-7} e^{-\gamma \eta r}.$$  

Then the error is optimized for $r \sim \eta^{-1} \log(\eta^{-1})$.

$\eta$ is chosen small so $g_{\epsilon, \eta} \sim \delta(\epsilon - \cdot)$. 
Kernel Polynomial Method to approximate $g_{\epsilon, \eta}(\cdot) \sim \delta(\epsilon - \cdot)$.

With more cost, can calculate DoS for multi-layers.

graphene bilayer with $6^\circ$ twist DoS and monolayer graphene DoS.
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Electronic Density of States for Incommensurate Layers.

E. Cancès, P. Cazeaux, M. Luskin
Generalized Kubo Formulas for the Transport Properties of
Incommensurate 2D Atomic Heterostructures.

Twistronics: Manipulating the electronic properties of two-dimensional
layered structures through their twist angle.