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ELECTRONIC DENSITY OF STATES FOR INCOMMENSURATE LAYERS*

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Abstract. We prove that the electronic density of states (DOS) for two-dimensional incommensurate layered structures, where Bloch theory does not apply, is well-defined as the thermodynamic limit of finite clusters. In addition, we obtain an explicit representation formula for the DOS as an integral over local configurations. Next, based on this representation formula, we propose a novel algorithm for computing electronic structure properties in incommensurate heterostructures, which overcomes limitations of the common approach to artificially strain a large supercell and then apply Bloch theory.

Key words. incommensurate, two-dimensional, density of states

AMS subject classification. 81V70

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1. Introduction.

Bloch theory provides an elegant solution for describing the electronic structure of periodic materials. However, there has been a lot of focus recently on the study of *incommensurate* layers of two-dimensional (2D) crystal structures [18, 19]. In the absence of periodicity, computing the electronic structure of such materials becomes more challenging.

A common approach to approximate the electronic properties of such a system is to artificially strain it to obtain periodicity on a large supercell and then apply Bloch theory to this periodic system [4, 9, 10, 13, 18]. Commensurate approximations to an incommensurate system are computationally expensive, and their approximation error is unclear. Here we introduce a new method for computing a class of observables derived from the density of states for multilayer incommensurate heterostructures *without* requiring an artificial strain in the system.

To approximate an observable of an infinite incommensurate system, we approximate local lattice site contributions to the observable. We observe that a site is uniquely defined by its local geometry. Using an equidistribution theorem, there is a predictable distribution of local geometries and hence site contributions. Consequently, we can express observables in incommensurate heterostructures in terms of an integral over a unit cell, in a fashion rather similar to Bloch theory. This unit cell classification of local configurations is related to Bellisard's noncommutative Brillouin zone for aperiodic solids [1]. Prodan used the Bellisard formalism to compute electronic properties for periodic materials with on-site defects modeled by a tight-binding model [15]. Here we consider the density of states and related observables for incommensurate multilayers.

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While the methodology is in principle generic, our derivation and analysis focuses on tight-binding models, which are commonly employed for computing the electronic structure of 2D materials [2, 8]. We consider the density of states and related observables for incommensurate multilayers. We use Chebyshev polynomial methods to approximate the density of states as a function [6, 14, 16, 17, 21], and from this function any observable can be computed.

Outline. In section 2 we introduce the results for the bilayer case and briefly discuss their extension to the multilayer case. In section 2.1 we introduce incommensurate systems and the equidistribution result. In section 2.2 we specify the details of our model problem, and in section 2.3 we show how to compute the local density of states. In section 2.4 we prove the infinite system is well posed and express the observables as an integral over local observables.

In section 3 we describe an approximation scheme and present numerical results. In section 3.1 we discuss the integral discretization. In section 3.2 we introduce a Chebyshev kernel polynomial method, and in section 3.4 we present numerical results. In section 4 we present the details of the proofs.

2. Main results.

2.1. Incommensurate heterostructures. Consider two periodic atomic sheets in parallel 2D planes separated by a constant distance. Each individual sheet can be described as a Bravais lattice embedded in $\mathbb{R}^2$ by neglecting the out of plane distance. This coordinate is not relevant for classifying the aperiodicity and will be incorporated in section 2.2. For sheet $j \in \{1, 2\}$, we define the Bravais lattice

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

where A_j is a 2×2 invertible matrix. We define the *unit cell* for sheet j as

$$\Gamma_j = \{A_j \alpha : \alpha \in [0, 1)^2\}.$$

Each individual sheet is trivially periodic, since

$$\mathcal{R}_j = A_j n + \mathcal{R}_j \qquad \text{for } n \in \mathbb{Z}^2.$$

However, the combined system $\mathcal{R}_1 \cup \mathcal{R}_2$ need not be periodic (Figure 1(a)). (Note that here $\mathcal{R}_1 \cup \mathcal{R}_2$ is only considered to describe geometry, not as an indexing of the atoms as it would have the failure of identifying the origins from each lattice.)

Since we are interested in aperiodic systems, we consider *incommensurate* systems, which we define next.

DEFINITION 2.1. *Lattices* $\mathcal{L}_1$ *and* $\mathcal{L}_2$ *are* incommensurate *if*

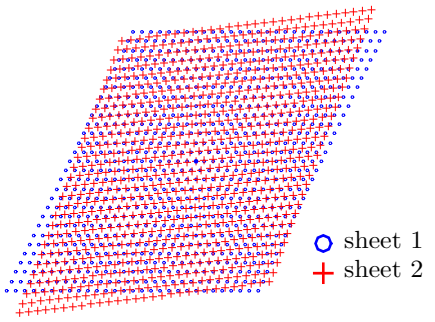
$$v + \mathcal{L}_1 \cup \mathcal{L}_2 = \mathcal{L}_1 \cup \mathcal{L}_2 \quad \Leftrightarrow \quad v = \begin{pmatrix} 0 \\ 0 \end{pmatrix}.$$

We define the reciprocal lattice of $\mathcal{R}_j$ as

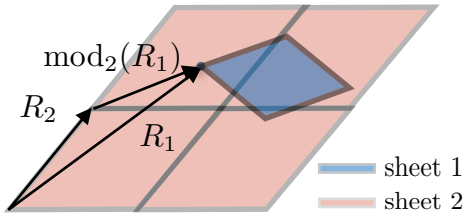
$$\mathcal{R}_j^* := \left\{2\pi (A_j^{-1})^* n : n \in \mathbb{Z}^2\right\}.$$

We will use the following assumption.

Assumption 2.1. The dual lattices $\mathcal{R}_1^*$ and $\mathcal{R}_2^*$ are incommensurate.

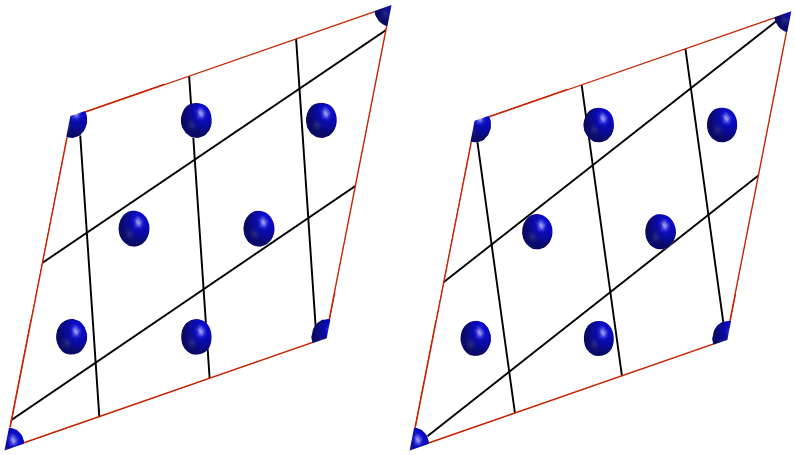


(a) An incommensurate hexagonal bilayer. Sheet 1 is rotated by $\theta = 6^\circ$ relative to sheet 2.



(b) $\mathrm{mod}_2(R_1)$ is the shift of the first lattice relative to the second lattice.

FIG. 1. *Visualization of incommensurate bilayer geometry.*



(a) Incommensurate cell

(b) Commensurate cell approximation

FIG. 2. (A) *Two lattices (spheres and lines) that are incommensurate.* (B) *The sphere lattice is slightly rotated to obtain a commensurate cell approximation.*

Since the majority of material simulation tools rely on periodicity, the most common method at present to simulate incommensurate layers is to adjust one of the two layers slightly in order to make the system commensurate on some larger supercell (Figure 2). In contrast we take advantage of an equidistribution of local geometries.

To parameterize the local geometries, we define the modulation operator $\mathrm{mod}_j : \mathbb{R}^2 \to \Gamma_j$ on sheet j for position $u \in \mathbb{R}^2$:

$$\mathrm{mod}_j(u) := u + R_j, \quad \text{where } R_j \in \mathcal{R}_j \text{ such that } u + R_j \in \Gamma_j.$$

Then the relative shift of site $R_1 \in \mathcal{R}_1$ is $\mathrm{mod}_2(R_1) \in \Gamma_2$ (See Figure 1(b)). The local geometry of site $R_1 \in \mathcal{R}_1$ is defined by

$$\mathcal{R}_1 \cup \mathcal{R}_2 - R_1 = \mathcal{R}_1 \cup (\mathcal{R}_2 - R_1) = \mathcal{R}_1 \cup (\mathcal{R}_2 - \mathrm{mod}_2(R_1)).$$

Hence, the local geometry is determined by the relative shift $\mathrm{mod}_2(R_1)$. The same argument holds for relative configurations around a site on sheet two. A fundamental

idea in this method is that the distribution of $\mathrm{mod}_1(R_2) \in \Gamma_1$ and $\mathrm{mod}_2(R_1) \in \Gamma_2$ are uniform in the sense of Theorem 2.1 below.

We let

$$B_r = \{y \in \mathbb{R}^2 : |y| < r\} \qquad \text{for } r > 0.$$

For $j \in \{1, 2\}$, we let P_j be the transposition, that is, $P_1 = 2$ and $P_2 = 1$.

THEOREM 2.1. *Consider $\mathcal{R}_1$ and $\mathcal{R}_2$ incommensurate lattices embedded in $\mathbb{R}^2$ (i.e., satisfying Assumption* 2.1*). Then for $g \in C_{per}(\Gamma_{P_j})$, we have*

$$\frac{1}{\#\mathcal{R}_j \cap B_r} \sum_{\ell \in \mathcal{R}_j \cap B_r} g(\ell) \to \frac{1}{|\Gamma_{P_j}|} \int_{\Gamma_{P_j}} g(b)db, \qquad \text{as } r \to \infty. \tag{2.1}$$

In particular, local geometries around sheet 1 sites can be parameterized by Γ_2, while local geometries around sheet 2 sites can be parameterized by Γ_1.

Theorem 2.1 suggests the following strategy for defining *and computing* electronic structure properties in incommensurate heterostructures: (1) split an observable into local contributions from each atomic site (we will employ the local density of states); (2) employ Theorem 2.1 to demonstrate that the thermodynamic limit from finite clusters exist (observe that (2.1) is a sum over a finite cluster); (3) use the right-hand side of (2.1) to compute the limit quantity.

2.2. Tight-binding model. Electronic structure is governed by solutions to the Schrödinger eigenproblem. It is typically approximated using methods such as the Kohn–Sham DFT (KS-DFT) model or the Hartree–Fock approximation [8, 12]. For systems in the thousands of atoms, however, the standard KS-DFT calculation becomes intractable. The tight-binding (TB) model applies further approximations and as a result can treat larger systems ranging in the millions of atoms.

Let $\mathcal{A}_i$ denote the set of indices of orbitals associated with each unit cell of sheet i. We assume that $\mathcal{A}_i$ are finite and that $\mathcal{A}_1 \cap \mathcal{A}_2 = \emptyset$. Then the full degree of freedom space is

$$\Omega = (\mathcal{R}_1 \times \mathcal{A}_1) \cup (\mathcal{R}_2 \times \mathcal{A}_2).$$

The interaction between orbitals indexed by $R\alpha, R'\alpha' \in \Omega$ is denoted by $h_{\alpha\alpha'}(R - R')$, where $h_{\alpha\alpha'} \in C(\mathbb{R}^2)$. Although the sheets have a vertical displacement between them, this distance is constant and hence can be encoded into $h_{\alpha\alpha'}$ (using the assumption that $\mathcal{A}_1 \cap \mathcal{A}_2 = \emptyset$). We will further use the following assumption.

Assumption 2.2. Orbital interactions $h_{\alpha\alpha'}$ are uniformly continuous on $\mathbb{R}^2$ and decay exponentially, that is, $\forall \alpha, \alpha' \in \mathcal{A}_1 \cup \mathcal{A}_2$

$$|h_{\alpha\alpha'}(x)| \leq Ce^{-\tilde{\gamma}|x|} \qquad \text{for } x \in \mathbb{R}^2.$$

This applies in most scenarios, since in most tight-binding models the orbitals are *tightly bound* around the atomic sites [8] or are exponentially decaying. We then formally define a matrix H such that

$$H_{R\alpha, R'\alpha'} = h_{\alpha\alpha'}(R - R').$$

This is an infinite matrix, and hence the eigenproblem

$$H\psi = E\psi$$

for $\psi \in \mathbb{C}^\Omega$ cannot be solved directly. Instead, we will define a class of observables for the infinite system by first defining them for finite subsystems and then passing to the limit in section 2.4.

For $\tilde{\Omega} \subset \Omega$ with $\#\tilde{\Omega} = n$ the associated Hamiltonian is $\tilde{H} = (H_{ij})_{i,j\in\tilde{\Omega}} \in M_n(\mathbb{C})$, where $M_n(\mathbb{C})$ denotes the set of $n \times n$ Hermitian matrices over $\mathbb{C}$. The *density of states* for $\tilde{\Omega}$ can be defined via its action on test functions, or, observables g, by

$$\mathcal{D}[\tilde{H}](g) = \frac{1}{n}\mathrm{Tr}[g(\tilde{H})], \quad g \in C(\mathbb{R}).$$

(We will later slightly extend the space of observables.) For example, we can consider the bond energy $\mathcal{D}[\tilde{H}](U_T)$, where $U_T(\epsilon) = \epsilon F_T(\epsilon)$ and $F_T(\epsilon) = (1 + e^{(\epsilon-\mu)/kT})^{-1}$ is the Fermi function. Formally, the value of the observable for the infinite system Ω is the limit of $\mathcal{D}[\tilde{H}](g)$ as $\tilde{\Omega} \uparrow \Omega$.

For future reference we remark that, since H is defined in terms of the lattices R_j and the hopping functions $h_{\alpha\alpha'}$, we will say "H satisfies Assumptions 2.2 and 2.1" to mean that "R_1, R_2 satisfy Assumption 2.1 and $h_{\alpha\alpha'}$ satisfy Assumption 2.2."

2.3. Local density of states. The next step is to define the local density of states distribution, which will allow us to identify local site contribution to an observable. Consider a finite subsystem $\tilde{\Omega} \subset \Omega$ with associated Hamiltonian $\tilde{H} \in M_n(\mathbb{C})$; then the local density of states (LDoS) distribution is defined as

$$\mathcal{D}_k[\tilde{H}](g) = [g(\tilde{H})]_{kk}, \qquad k \in \tilde{\Omega},\ g \in C(\mathbb{R}).$$

Note that

$$\frac{1}{n}\sum_{k\in\tilde{\Omega}} \mathcal{D}_k[\tilde{H}](g) = \mathcal{D}[\tilde{H}](g).$$

This reformulation puts us very close to the setting of Theorem 2.1. It remains to control the dependence of $\mathcal{D}_k[\tilde{H}](g)$ on $\tilde{\Omega}$, which we will achieve in the next section by fixing k and letting $\tilde{\Omega} \uparrow \Omega$ while controlling the error.

Toward that end we now specify a sequence of local degree of freedom spaces,

$$\Omega_r = \Big[\big[\mathcal{R}_1 \cap B_r\big]\times\mathcal{A}_1\Big] \cup \Big[\big[\mathcal{R}_2 \cap B_r\big]\times\mathcal{A}_2\Big] \qquad \text{for } r > 0;$$

see also Figure 3. For $r > 0$ and $b \in \Gamma_{P_j}$ we define $H_{r,j}(b) \in M_{|\Omega_r|}(\mathbb{C})$ by

$$[H_{r,j}(b)]_{R\alpha,R'\alpha'} = h_{\alpha\alpha'}\big(b(\delta_{\alpha\in\mathcal{A}_{P_j}} - \delta_{\alpha'\in\mathcal{A}_{P_j}}) + R - R'\big)$$

for $R\alpha, R'\alpha' \in \Omega_r$. Physically, $H_{r,j}(b)$ describes a cluster of radius r of the bilayer system in which the sheet P_j is shifted by b. If the LDoS is well-defined in the thermodynamic limit, we expect it to be approximated by the local cluster around the site of interest, i.e.,

$$\mathcal{D}_{0\alpha}[H_{r,j}(\mathrm{mod}_{P_j}(R))] \approx \mathcal{D}_{R\alpha}[H], \quad R\alpha \in \mathcal{R}_j \times \mathcal{A}_j$$

in a suitable weak sense. Since the two layers are incommensurate, the set of shifts b that occur in this way are dense. Hence, we consider the local density of states

$$\mathcal{D}_\alpha[H_{r,j}(b)] := \mathcal{D}_{0\alpha}[H_{r,j}(b)] \quad \text{for} \quad \alpha \in \mathcal{A}_j \tag{2.2}$$

for arbitrary shifts b.

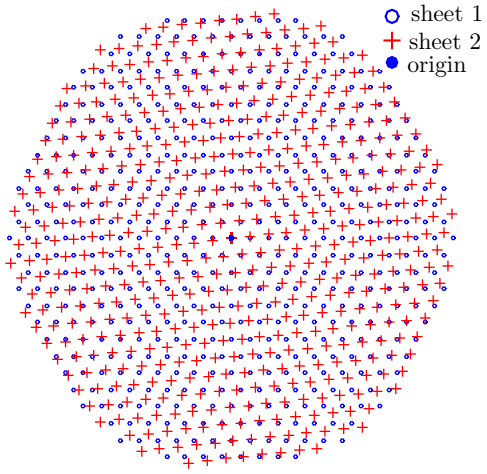


FIG. 3. *All the sites in Ω_r for a hexagonal bravais lattice. The central site for sheet 1 is highlighted.*

2.4. Thermodynamic limit. We now consider the limit as $r \to \infty$ of the LDoS, which will allow us to define the DoS for the infinite system. Let

$$E[H] := \sup_{r>0,\, j\in\{1,2\}} \left[\sup_{b\in\Gamma_j} \|H_{r,j}(b)\|_2 \right] < \infty,$$

where $\|\tilde{H}\|_2 := \sup_{\psi\in\mathbb{C}^n\setminus\{0\}} \|\tilde{H}\psi\|_2/\|\psi\|_2$ for $\tilde{H} \in M_n(\mathbb{C})$. $E[H]$ is finite because of the exponential localization from Assumption 2.2. Then the local density of states distribution will be supported on the interval

$$S[H] = [-E[H], E[H]].$$

Since the spectrum is only defined on $S[H]$, we are only interested in observables $g \in C(S[H])$. We supply this space with the norm

$$\|g\|_\infty := \sup_{x\in S[H]} |g(x)| \qquad \text{for } g \in C(S[H]).$$

For $U \subset \mathbb{C}$, we define the distance

$$d(U, S[H]) = \inf_{z\in U, z'\in S[H]} |z - z'|.$$

This is a bound on the distance between U and the spectrum. To pass to the limit in the LDoS and later in the DoS, we narrow down admissible test functions. We define

$$\Lambda_{\tilde{d}} := \left\{ g \in C(\mathbb{R}) \mid \ g \text{ admits an analytic extension to } \{z \in \mathbb{C} : d(z, S[H]) < \tilde{d}\} \right\}.$$

Then our admissible test functions are

$$\Lambda := \bigcup_{\tilde{d}>0} \Lambda_{\tilde{d}}.$$

While we consider the thermodynamic limit on the space Λ, we afterward extend to $g \in C(S[H])$ using the fact that Λ is dense in $C(S[H])$.

THEOREM 2.2. (1) *Suppose that H satisfies Assumptions* 2.1 *and* 2.2. *Then, for $\alpha \in \mathcal{A}_j$, there exists a function $\mathcal{D}_\alpha[H] : \Gamma_{P_j} \times C(S[H]) \to \mathbb{C}$ such that, for $g \in \Lambda$,*

$$\mathcal{D}_\alpha[H_{r,j}(b)](g) \to \mathcal{D}_\alpha[H](b,g) \quad \text{as } r \to \infty.$$

(The distribution $\mathcal{D}_\alpha[H](b,g)$ is the local density of states for the infinite system.)

(2) *The map $g \mapsto \mathcal{D}_\alpha[H](b,g)$ is a bounded linear functional, more precisely,*

$$\left|\mathcal{D}_\alpha[H](b,g)\right| \le \|g\|_\infty \qquad \text{for } g \in C(S[H]).$$

(3) *There exist constants $C, \gamma' > 0$ such that, for $\tilde{d} > 0$ and $g \in \Lambda_{\tilde{d}}$,*

$$|\mathcal{D}_\alpha[H](b,g) - \mathcal{D}_\alpha[H_{r,j}(b)](g)| \le C\tilde{d}^{-6} \sup_{d(z,S[H])<\tilde{d}} \left[|g(z)|e^{-\gamma'\tilde{d}r}\right].$$

We next analyze the regularity of the map $b \mapsto \mathcal{D}_\alpha[H](b,g)$ for fixed g, which will allow us to integrate with respect to b.

THEOREM 2.3. *Suppose $h_{\alpha\alpha'} \in C^n(\mathbb{R}^2)$ for $n \in \mathbb{N} \cup \{0,\infty\}$, $\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}$ is uniformly continuous for $m + m' \le n$, and there exist constants $C_{mm'}, \gamma_{mm'} > 0$ such that*

$$|\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}(r)| \le C_{mm'} e^{-\gamma'_{mm'} r}.$$

Then, for $\alpha \in \mathcal{A}_j$ and $g \in \Lambda$,

$$\mathcal{D}_\alpha[H](\cdot, g) \in C^n_{\mathrm{per}}(\Gamma_{P_j}).$$

Our next objective is to rigorously define the density of states distribution for the infinite incommensurate bilayer system H. Taking a sequence of finite incommensurate clusters surrounded by a vacuum that grow toward infinity and combining our results on the equidistribution of local configurations with the convergence of the local density of states we obtain the following representation formula.

THEOREM 2.4. *Suppose that H satisfies Assumptions* 2.1 *and* 2.2. *Then there exists a bounded linear functional $\mathcal{D}[H] : C(S[H]) \to \mathbb{C}$ such that, for $g \in \Lambda$, we have*

$$\mathcal{D}[H_{r,j}(0)](g) \to \mathcal{D}[H](g) \quad \text{as } r \to \infty \quad \text{for } j = 1, 2.$$

Further, we have an explicit expression for the limit $\mathcal{D}[H](g)$ in terms of the LDoS operators:

$$\mathcal{D}[H](g) = \nu \sum_{j=1}^{2} \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H](b,g)db, \tag{2.3}$$

where

$$\nu = \frac{1}{|\mathcal{A}_2| \cdot |\Gamma_1| + |\mathcal{A}_1| \cdot |\Gamma_2|}.$$

Note that the right-hand side of (2.3) *is well-defined for $g \in \Lambda$ by using Theorem* 2.2 *and Theorem* 2.3.

This suggests a natural approximation to the DoS using the LDoS operators,

$$\nu \sum_{j=1}^{2} \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H_{r,j}(b)](g)db.$$

THEOREM 2.5. *Suppose that H satisfies Assumptions* 2.1 *and* 2.2. *If $g \in \Lambda_{\tilde{d}}$, then we have the explicit error bound on the LDoS approximation*

$$\left| \mathcal{D}[H](g) - \nu \sum_{j=1}^{2} \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H_{r,j}(b)](g) db \right| \leq C\tilde{d}^{-6} \sup_{d(z,S[H])<\tilde{d}} \left[|g(z)| e^{-\gamma \tilde{d} r} \right],$$

where C, γ are independent of $r, \tilde{d}$, and g.

Proof. This follows trivially from Theorem 2.2. ∎

Remark 2.1. The finite systems employed in the thermodynamic limit are defined by the matrices $H_{r,j}(0)$ for $j = 1, 2$. They represent finite incommensurate clusters surrounded by a vacuum. Since the boundary Hamiltonian entries are not chosen by DFT calculations or experimental values they will not be accurate. However, as long as the boundary coefficients satisfy Assumption 2.2, the limit of the density of states $\mathcal{D}[H_{r,j}(0)]$ will be independent of the choice of boundary terms.

Remark 2.2. For the sake of convenience, we have chosen a circular shape for the approximating domains. Weaker requirements can be readily formulated, e.g., domains $\widetilde{\Omega}$ should *contain* balls centered at the origin with radii growing to infinity, while at the same time keeping a suitable bound on the surface area to volume ratio.

Remark 2.3. The Riesz–Markov–Kakutani representation theorem states that the dual space of the continuous compact functions are the Radon measures. Since all our density of states and local density of states operators are continuous linear functionals over the space of compact continuous functions, they are all Radon measures.

Remark 2.4. This methodology can easily be extended to three or more incommensurate layers, but at the cost of higher dimensional integration, since one must integrate over all relative shifts between the different layers. The local density of states, on the other hand, where there is no integration, can be easily analyzed for multiple layers without adding much to the cost.

3. Numerical simulations.

3.1. Quadrature. To compute the integrals occuring in Theorem 2.5 numerically, we can use the smoothness properties from Theorem 2.3, which can be strengthened further by assuming analyticity on $h_{\alpha\alpha'}$.

THEOREM 3.1. *Assume $h_{\alpha\alpha'}$ is analytic and satisfies Assumption* 2.2. *Let*

$$S_j = \left\{ A_j \begin{pmatrix} i_1/N_{disc} \\ i_2/N_{disc} \end{pmatrix} : 0 \leq i_1, i_2 < N_{disc} \right\}$$

be the uniform discretization sample points. Then we have

$$\left| \frac{|\Gamma_{P_j}|}{N_{disc}^2} \sum_{b \in S_{P_j}} \sum_{\alpha \in \mathcal{A}_j} \mathcal{D}_\alpha[H](b, g) - \sum_{\alpha \in \mathcal{A}_j} \int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H](b, g) db \right| \leq C\tilde{d}^{-1} \sup_{d(z,S[H])<\tilde{d}} \left[|g(z)| e^{-\gamma'' \tilde{d} N_{disc}} \right]$$

for some $\gamma'' > 0$.

Remark 3.1. In practice, $h_{\alpha\alpha'}$ has a finite cutoff and hence cannot be analytic. However, we can think of it as an approximation to an "exact" analytic $\bar{h}_{\alpha\alpha'}$. Preasymptotically, it is therefore useful to treat $h_{\alpha\alpha'}$ as if it were itself analytic.

3.2. Kernel polynomial method approximation. A complete eigensolve on $H_{r,j}(b)$ for each quadrature point b is computationally expensive, with scaling $O(r^6)$. Instead we use a Chebyshev kernel polynomial method (KPM) to compute the density of states [21]. This method scales as $O(r^2)$, where the constant depends on the desired accuracy. It yields the density of states operator as a smooth function from which multiple observables can then be computed.

LEMMA 3.1. *Assume that H satisfies Assumptions* 2.2 *and* 2.1 *and that $f \in C(\mathbb{R}\times\mathbb{R};\mathbb{C})$ and $g \in \Lambda$; then*

$$\int \mathcal{D}[H]\big(f(\varepsilon,\cdot)\big)g(\varepsilon)d\varepsilon = \mathcal{D}[H]\bigg(\int f(\varepsilon,\cdot)g(\varepsilon)d\varepsilon\bigg).$$

Proof. This result follows immediately from Remark 2.3 and Fubini's theorem. ∎

We note that $|\mathcal{D}[H](g)| \le \|g\|_\infty$, and hence

$$\bigg|\mathcal{D}[H]\bigg(\int f(\varepsilon,\cdot)g(\varepsilon)d\varepsilon\bigg) - \mathcal{D}[H](g)\bigg| \le \bigg\|\int f(\varepsilon,\cdot)g(\varepsilon)d\varepsilon - g\bigg\|_\infty. \tag{3.1}$$

Note that this bound trivially extends from Λ to $C(S[H])$. Moreover, if $f(\varepsilon,e) \approx \delta(\varepsilon - e)$, then the smooth function

$$D_f(\varepsilon) := \mathcal{D}[H](f(\varepsilon,\cdot)) \approx \mathcal{D}[H]$$

in the sense of (3.1). We now choose a convenient f.

Recall that the Chebyshev polynomials are a basis defined recursively by

$$T_0(e) = 1, \qquad T_1(e) = e, \qquad \text{and} \qquad T_{n+1}(e) = 2eT_n(e) - T_{n-1}(e). \tag{3.2}$$

The polynomials are orthogonal in the sense that

$$\int_{-1}^{1} \frac{1}{\pi\sqrt{1-e^2}} T_n(e)T_m(e)de = \frac{1+\delta_{0n}}{2}\delta_{nm}.$$

An approximation to the shifted delta function $\delta(e-\varepsilon)$, at $\varepsilon \in (-1,1)$, is given by

$$\hat{\chi}_p(\varepsilon,e) = \frac{1}{\pi\sqrt{1-\varepsilon^2}} \sum_{m\le p} g_m^p T_m(\varepsilon)T_m(e), \qquad e,\varepsilon \in (-1,1),$$

where

$$g_m^p = (2-\delta_{m0})\frac{(p-m+1)\cos(\frac{\pi m}{p+1}) + \sin(\frac{\pi m}{p+1})\arctan(\frac{\pi}{p+1})}{p+1}$$

are the so-called Jackson coefficients designed to remove the Gibbs phenomenon [21].

To approximate the density of states on the interval $S[H] = [-E[H], E[H]]$, we rescale

$$\chi_p(\varepsilon,e) := \eta\hat{\chi}_p(\eta\varepsilon,\eta e), \qquad e,\varepsilon \in (-1/\eta, 1/\eta),$$

where η is a positive constant selected so that $E[H] \le 1/\eta$.

We approximate $\mathcal{D}[H]$ by

$$D_{\chi_p}(\varepsilon) = \nu\sum_{j=1}^{2}\sum_{\alpha\in\mathcal{A}_j}\int_{\Gamma_{P_j}} \mathcal{D}_\alpha[H](b,\chi_p(\varepsilon,\cdot))db$$

and subsequently approximate the integrand $\mathcal{D}_\alpha[H](b, \chi_p(\varepsilon,\cdot))$ by

$$\begin{aligned}\mathcal{D}_\alpha[H_{r,j}(b)](\chi_p(\varepsilon,\cdot)) &= [\chi_p(\varepsilon, H_{r,j}(b))]_{0\alpha,0\alpha} \\ &= \frac{\eta}{\pi\sqrt{1-(\eta\varepsilon)^2}} \sum_{m\le p} g_m^p T_m(\eta\varepsilon)\big[\eta T_m(H_{r,j}(b))\big]_{0\alpha,0\alpha}.\end{aligned} \tag{3.3}$$

Note that for all ε, the calculation requires the same $[T_m(\eta H_{r,j}(b))]_{0\alpha,0\alpha}$ coefficients, which is the core of our Algorithm A.

Algorithm A: Approximate DoS

Step 1: Choose quadrature parameter $N_{\mathrm{disc}} \in \mathbb{N}$ and domain truncation radius $r > 0$. For each $j \in \{1, 2\}$ and $b \in S_{P_j}$ construct the matrix $H_{r,j}(b)$.

Step 2: Let $e_i \in \mathbb{R}^{|\Omega_r|}$ such that $[e_i]_j = \delta_{ij}$ is the i^{th} coordinate vector. Using the recursion (3.2) we compute, for $\alpha \in \mathcal{A}_j$,

$v_0 = e_{0\alpha}$

$v_1 = \eta H_{r,j}(b) e_{0\alpha}$

store: $[T_0(\eta H_{r,j}(b))]_{0\alpha,0\alpha} = e_{0\alpha} \cdot v_0$ and $[T_1(\eta H_{r,j}(b)]_{0\alpha,0\alpha} = e_{0\alpha} \cdot v_1$

for loop: $1 \le m \le p-1$

$v_{m+1} = 2\eta H_{r,j}(b) v_m - v_{m-1}$

store: $[T_{m+1}(\eta H_{r,j}(b))]_{0\alpha,0\alpha} = e_{0\alpha} \cdot v_{m+1}$

This yields the coefficients $[T_m(\eta H_{r,j}(b))]_{0\alpha,0\alpha}$ for (3.3).

Step 3: Compute the expression

$$\mathcal{D}_\alpha[H_{r,j}(b)](\chi_p(\varepsilon,\cdot)) = \frac{\eta}{\pi\sqrt{1-(\eta\varepsilon)^2}} \sum_{m\le p} g_m^p T_m(\eta\varepsilon)[T_m(\eta H_{r,j}(b))]_{0\alpha,0\alpha}.$$

This yields a local density of states approximation, which is interesting in its own right.

Step 4: The total density of states approximation is obtained by evaluating

$$D(\varepsilon) := \frac{\nu}{N_{\mathrm{disc}}^2} \sum_{j=1}^{2} \sum_{\alpha\in\mathcal{A}_j} \sum_{b\in S_{P_j}} |\Gamma_{P_j}| \cdot \mathcal{D}_\alpha[H_{r,j}(b)](\chi_p(\varepsilon,\cdot))$$

for all desired ε.

The approximation error for the output $D(\varepsilon)$ of Algorithm A is estimated in the following result.

THEOREM 3.2. *Suppose that H satisfies Assumptions* 2.1 *and* 2.2, *and then for* $g \in \Lambda_{\tilde{d}}$,

$$\begin{aligned}\left|\mathcal{D}[H](g) - \int D(\varepsilon) g(\varepsilon) d\varepsilon\right| &\\ \leq \underbrace{C\tilde{d}^{-6} \sup_{d(z,S[H])<\tilde{d}} \left[|g(z)| e^{-\gamma \tilde{d} r}\right]}_{\textit{Truncation Error}} &\\ + \underbrace{C\tilde{d}^{-1} \sup_{d(z,S[H])<\tilde{d}} \left[|g(z)| e^{-\gamma' \tilde{d} N_{disc}}\right]}_{\textit{Discretization Error}} + \underbrace{C' \left\| g - \int \chi_p(\varepsilon, \cdot) g(\varepsilon) d\varepsilon \right\|_\infty}_{\textit{Kernel Polynomial Method Error}}&.\end{aligned}$$

Here $\gamma, \gamma' > 0$ are independent of the choice of $\tilde{d}$.

Proof. The truncation error follows from Theorem 2.2, the discretization error from Theorem 3.1, and the kernel polynomial error from (3.1). ∎

Remark 3.2. If we do not assume that $h_{\alpha\alpha'}$ is analytic and use $h_{\alpha\alpha'} \in C_0^n(\mathbb{R}^2)$ instead, the discretization error above is replaced with the standard periodic discretization error [20, Theorem 1], but the bound does not give the dependence of N_{disc} on $\tilde{d}$.

3.3. Convergence rates. We briefly discuss a heuristic to choose the approximation parameters $p, N_{\text{disc}} \in \mathbb{N}$ and $r > 0$. In practice, one is interested in calculating the density of states at a point or in calculating an observable $\mathcal{D}[H](g)$ for $g \in \Lambda_{\tilde{d}}$.

For the first case, we note that χ_p acts similar to a Gaussian approximation to the identity of width proportional to p^{-1} [21] with well preserved regularity because of the Jackson coefficients. For analytic purposes, we can therefore consider

$$\chi_p(\varepsilon, e) \sim \phi_p(\varepsilon - e), \tag{3.4}$$

for

$$\phi_p(e) = \frac{p}{\sqrt{2\pi^3}} e^{-(ep/\pi)^2}$$

as demonstrated in [21]. An approximation of the density of states at a given energy point ε is given by $\mathcal{D}[H](\chi_p(\varepsilon, \cdot)) \sim \mathcal{D}[H](\phi_p)$. To approximate $\mathcal{D}[H](\chi_p(\varepsilon, \cdot))$, we consider the error bounds in Theorem 3.2. We have

$$\text{truncation error} \sim \tilde{d}^{-2} \sup_{d(z,S[H])<\tilde{d}} \left[|\chi_p(\varepsilon, z)| e^{-\gamma \tilde{d} r}\right].$$

If we let $\tilde{d} \sim p^{-1}$ and we use (3.4), we have $\sup_{d(z,S[H])<p^{-1}} [\chi_p(\varepsilon, z)] \sim p$. Then

$$\text{truncation error} \sim p^3 e^{-\gamma p^{-1} r}.$$

Therefore $r \sim p \log(p)$. The same argument holds for the discretization error, yielding

$$r \sim N_{\text{disc}} \sim p \log(p) \tag{3.5}$$

Suppose the density of states is a Lipschitz continuous function, i.e.,

$$\mathcal{D}[H](g) = \int \text{DoS}(\epsilon) g(\epsilon) d\epsilon,$$

where DoS has Lipschitz constant M. Then we can estimate

$$|\mathrm{DoS}(\varepsilon) - \mathcal{D}[H](\chi_p(\varepsilon,\cdot)| \leq Mp^{-1},$$

where we have used that $\chi_p(\varepsilon,\cdot)$ acts like a Gaussian with width proportional to p^{-1}. We then obtain

$$|D(\varepsilon) - \mathrm{DoS}(\varepsilon)| \leq C'\left(pe^{-\gamma'\frac{N_{\mathrm{disc}}}{p}} + p^2 e^{-\gamma\frac{r}{p}} + Mp^{-1}\right).$$

If the constants in (3.5) are chosen sufficiently small, we have

$$|D(\varepsilon) - \mathrm{DoS}(\varepsilon)| \leq (M+C)p^{-1}, \tag{3.6}$$

where $C > 0$ is independent of smoothness properties of DoS.

If the DoS is C^2 at a point ε of interest, then we may expect even stronger convergence. Using that $D(\varepsilon) \sim \int \mathrm{DoS}(e)\phi_p(\varepsilon - e)de$, we can do a Taylor expansion up to the second order around ε for $|e| \ll 1$ to obtain

$$\mathrm{DoS}(\varepsilon + e) = \mathrm{DoS}(\varepsilon) + e\mathrm{DoS}'(\varepsilon) + O(e^2).$$

This gives

$$|\mathrm{DoS}(\varepsilon) - D(\varepsilon)| = \left|\mathrm{DoS}(\varepsilon) - \int \phi_p(e)\Big(\mathrm{DoS}(\varepsilon) + e\mathrm{DoS}'(\varepsilon) + O(e^2)\Big)de\right|.$$

Using that $\int e\phi_p(e) = 0$, we get

$$|D(\varepsilon) - \mathrm{DoS}(\varepsilon)| \leq Cp^{-2}. \tag{3.7}$$

For the second case, when the observable $g \in \Lambda$ is fixed (no polynomial degree approximation parameter p), we have *in principle* exponential decay of the error in r and N_{disc}. This seems to imply that it would be optimal to calculate the observable directly using an eigensolve, thus avoiding the slower decay in p. However, the decay rate in r is strongly coupled to the value of $\tilde{d}$ from Theorem 2.2, which is fairly small for interesting observables. Therefore, the involved matrices are typically quite large, rendering direct eigensolves impractical.

3.4. Numerical results. We test our approximation scheme using a tight-binding model for twisted bilayer graphene [5] with a relative twist angle of 6°. We fix an $\alpha \in \mathcal{A}_1$ and then verify numerically the following two results:

1. As predicted in Theorem 2.2, $\mathcal{D}_\alpha[H_{r,1}(b)](\chi_p(\varepsilon,\cdot)) \to \mathcal{D}_\alpha[H](b,\chi_p(\varepsilon,\cdot))$ as $r \to \infty$ with exponential rate proportional to p^{-1}: see Figure 4.
2. As predicted by Theorem 3.2 and (3.7), $D \to \mathrm{DoS}$ pointwise as $p, r, N_{\mathrm{disc}} \to \infty$ with quadratic rate: see Figure 5.

Furthermore, we demonstrate the practicality of Algorithm A by reproducing twisted bilayer effects in the density of states of two stacked graphene sheets with a relative twist of 6° as predicted in [5] (see Figure 6). We included the DoS for monolayer graphene for comparison. The conical region near the $-.6$ energy region is called the Dirac cone. When the two layers interact, the curve splits near the cone tip (the Dirac point) forming two Van Hove singularities (VHS) on either side of the tip. In practice the VHS needs higher resolution. We will explore how to achieve high resolutions in a future work.

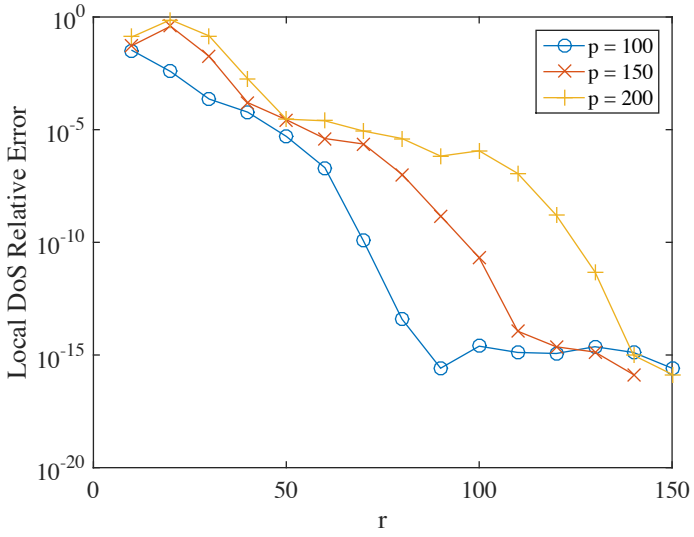


FIG. 4. *Relative error of* $\mathcal{D}_\alpha[H_{r,1}(0)](\chi_p(0,\cdot))$ *converging to* $\mathcal{D}_\alpha[H](\chi_p(0,\cdot))$ *for increasing values of* p. *Here the reference value is taken at* $r = 250$.

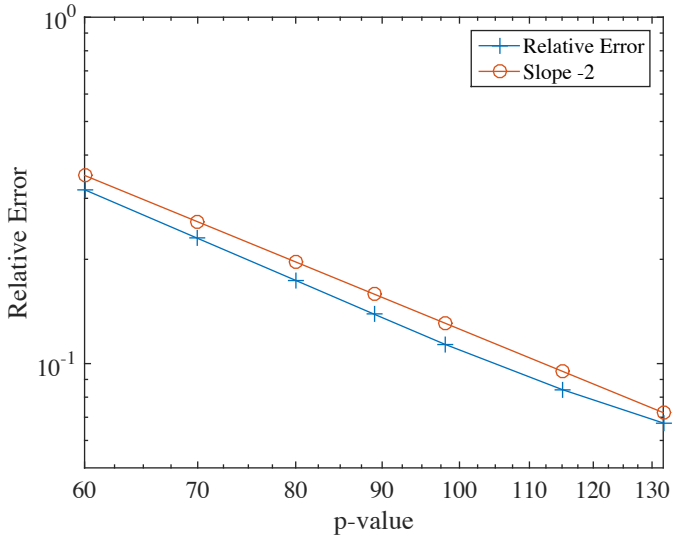


FIG. 5. *Relative error of* $D(0) \to DoS(0)$ *pointwise, where* r *and* N_{disc} *scale as in* (3.5). *The slope is* $-1.98 \approx -2$, *as predicted in* (3.7). *Here the reference value is taken at* $p = 196$ *with proportional choice of* N_{disc} *and* r. *The ratio in* (3.5) *is chosen such that the first point corresponds to* $p = 60$, $r = 20$, *and* $N_{disc} = 2$.

4. Proofs. To attain bounds on the density of states objects, we will use resolvent bounds as introduced in [3]. We denote $\mathcal{C}$ a contour around $S[H]$, which contains the spectrum. We can write for $\tilde{\Omega} \subset \Omega$ finite, $\tilde{H} \in M_{|\tilde{\Omega}|}(\mathbb{C})$, $k \in \Omega$, and g analytic

$$[g(\tilde{H})]_{kk} = \frac{1}{2\pi i} \oint_{\mathcal{C}} g(z)[(z - \tilde{H})^{-1}]_{kk} dz.$$

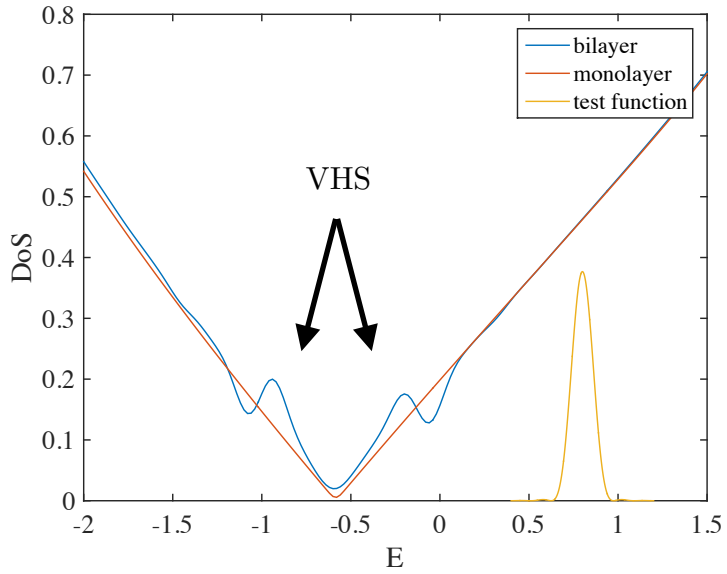


FIG. 6. *Approximation of the DoS using Algorithm A with* $r = 180$, $p = 700$, *and* $N_{disc} = 4$. *We can see VHS forming near the Dirac Point, agreeing with theoretical results* [5]. *We include the test function* $(\chi_p(\varepsilon, \cdot))$, *which is to scale in the E-axis, but not in the DoS-axis.*

We will then rely on decay estimates for $[(z - \tilde{H})^{-1}]_{kk}$ as $\tilde{\Omega} \uparrow \Omega$. We will vary our choice of $\mathcal{C}$ to tune the error bounds.

4.1. Proof of Theorem 2.1. Although this result is conceptually close to the equidistribution theorem [22], our specific statement of the result seems to be unavailable. Hence we prefer to give a complete proof. Without loss of generality, we let $j = 1$ and hence $P_j = 2$. Then we wish to show for $g \in C_{\text{per}}(\Gamma_2)$, we have

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

We let $V_r = \#\mathbb{Z}^2 \cap A_1^{-1} B_r$. We then wish to show for $g \in C_{\text{per}}(\Gamma_2)$ that

$$\frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} g(A_1 n) \to \frac{1}{|\Gamma_2|} \int_{\Gamma_2} g(b)db. \tag{4.1}$$

Since $C^\infty_{\text{per}}(\Gamma_2)$ is dense in $C_{\text{per}}(\Gamma_2)$, we assume $g \in C^\infty_{\text{per}}(\Gamma_2)$. On expanding g into Fourier modes, it suffices to show (4.1) for an arbitrary fourier mode $g(x) = e^{2\pi i m \cdot A_2^{-1} x}$, where $m \in \mathbb{Z}^2$.

If $m = (0, 0)$, then the left-hand side and right-hand side of (4.1) are 1.

For $m \neq (0, 0)$, the right-hand side of (4.1) vanishes, so we need to prove that $\frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} g(A_1 n) \to 0$ as $r \to \infty$. We first rewrite

$$\frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} g(A_1 n) = \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} e^{2\pi i m^t A_2^{-1} A_1 n} = \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} e^{2\pi i a \cdot n},$$

where $a = (a_1, a_2) = m^t A_2^{-1} A_1$. If both a_1 and a_2 were rational, then this would

contradict Assumption 2.1, since

$$(A_1^{-1})^* n + (A_2^{-1})^* m = (0,0)$$

for $n, m \in \mathbb{Z}^2$ if and only if $n = m = (0,0)$. Hence we assume, without loss of generality, that $a_2 \notin \mathbb{Q}$.

Let $c > 0$ such that

$$n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r \quad \Rightarrow \quad n_1 \in [-cr, cr].$$

Moreover, for $n_1 \in [-cr, cr] \cap \mathbb{Z}^2$ let $f_-(n_1), f_+(n_1) \in \mathbb{Z}^2$ such that $(n_1, n_2) \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r$ if and only if $f_-(n_1) \le n_2 \le f_+(n_1)$.

We can now compute

$$\begin{aligned}\frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} e^{2\pi i a \cdot n} &= \frac{1}{V_r} \sum_{n_1 \in [-cr,cr] \cap \mathbb{Z}^2} e^{2\pi i a_1 n_1} \sum_{n_2 = f_-(n_1)}^{f_+(n_1)} e^{2\pi i a_2 n_2} \\ &= \frac{1}{V_r} \sum_{n_1 \in [-cr,cr] \cap \mathbb{Z}^2} e^{2\pi i a_1 n_1} \frac{e^{2\pi i a_2 (f_-(n_1)+1)} - e^{2\pi i a_2 (f_+(n_1)+1)}}{1 - e^{2\pi i a_2}}.\end{aligned}$$

Since a_2 is irrational, $1 - e^{2\pi i a_2} \neq 0$, and hence we can estimate

$$\left| \frac{1}{V_r} \sum_{n \in \mathbb{Z}^2 \bigcap A_1^{-1} B_r} e^{2\pi i a \cdot n} \right| \le \frac{4cr}{|1 - e^{2\pi i a_2}| V_r} \le C r^{-1},$$

which vanishes in the limit $r \to \infty$, as required. This completes the proof of Theorem 2.1.

4.2. Proof of Theorem 2.2. Recall that

$$\Lambda := \bigcup_{\tilde{d} > 0} \Lambda_{\tilde{d}}.$$

In particular, note that Λ is dense in $C(S[H])$, in the sense that for any $f \in C(S[H])$ and $\epsilon > 0$, there exists $g \in \Lambda$ such that

$$\|g|_{S[H]} - f\|_\infty < \epsilon.$$

This will be useful for extending the density of states operators from Λ to $C(S[H])$.

LEMMA 4.1. *Suppose $\tilde{H} \in M_n(\mathbb{C})$, and $y : \{1, 2, \dots, n\} \to \mathbb{R}^2$ such that*

$$|\tilde{H}_{k\ell}| \le C e^{-\tilde{\gamma}|y(k) - y(\ell)|}$$

for some $\tilde{\gamma} > 0$. Suppose that there exists $N \in \mathbb{N}$, $r' > 0$ such that for all $x \in \mathbb{R}^2$, $|\#\{y(j) : y(j) \in B_{r'}(x)\}| < N$. Then there exists $\gamma > 0$ such that, for all $z \in \mathbb{C}$, $\operatorname{dist}(z, S[H]) \ge \tilde{d}/2$,

$$\left| [(z - \tilde{H})^{-1}]_{k\ell} \right| \le C' \tilde{d}^{-1} e^{-\gamma(1+\kappa)\tilde{d}|y(k) - y(\ell)|}.$$

Here C' and γ are dependent on $\tilde{\gamma}, N, r'$, and C, and $\kappa > 0$, $\kappa \ll 1$.

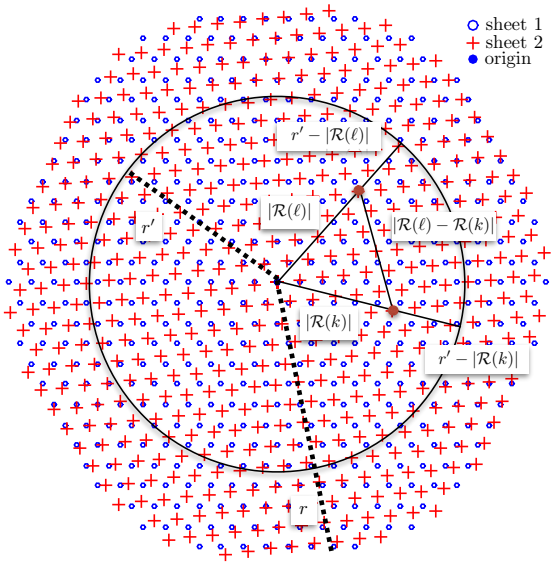


FIG. 7. *For given sites ℓ and k, we plot the relevant distances in solid lines and system radii in dotted lines for considering resolvent error in Lemma* 4.2.

Proof. This is a version of [3, Lemma 2.2]. Note that we don't include a lower bound on the distance between sites $y(j)$. We do not need this condition since we don't allow singularities in our coupling functions $h(x)$. This is reasonable for interlayer terms since $x \to 0$ does not correspond to orbitals moving arbitrary close together, as we assumed a fixed distance between the sheets. As for intralayer, the orbitals are fixed on the Bravais lattice. ∎

In particular, the previous lemma applies to the matrices $H_{r,j}(b)$. To apply it we will set $y = \mathfrak{R}$, where in the following we define

$$\mathfrak{R} : \Omega \to \mathbb{R}^2, \qquad \mathfrak{R}(R\alpha) = R.$$

For the next lemma, recall the definition of $H_{r',j}(b)$ from (2.2).

LEMMA 4.2. *Suppose that H satisfies Assumptions* 2.2 *and* 2.1. *Let $\tilde{\Omega} \subset \Omega$ be a set of indices and $\tilde{H}_j(b)$ be the matrix defined over $\tilde{\Omega}$ with shift b relative to sheet j, that is,*

$$[\tilde{H}_j(b)]_{R\alpha,R'\alpha'} = h_{\alpha\alpha'}\big(b(\delta_{\alpha\in\mathcal{A}_{P_j}} - \delta_{\alpha'\in\mathcal{A}_{P_j}}) + R - R'\big).$$

We let $k = R\alpha$ and $\ell = R'\alpha'$. Suppose that $r' > 0$ such that $\Omega_{r'} \subset \tilde{\Omega}$ and $\tilde{d} > 0$ such that $d(z, S[H]) > \tilde{d}$. Then

$$\left|\left[(z - \tilde{H}_j(b))^{-1}\right]_{k\ell} - \left[(z - H_{r',j}(b))^{-1}\right]_{k\ell}\right| \leq C\tilde{d}^{-6} \min\left\{e^{-\gamma\tilde{d}|\mathfrak{R}(k)-\mathfrak{R}(\ell)|}, r' e^{-\gamma\tilde{d}\max\{r'-|\mathfrak{R}(k)|, r'-|\mathfrak{R}(\ell)|\}}\right\},$$

where C and γ are independent of $\tilde{\Omega}$ and r' (see Figure 7*).*

Proof. We define the matrix $\tilde{H}_j^{r'}(b) \in M_{|\tilde{\Omega}|}(\mathbb{C})$ such that

$$[\tilde{H}_j^{r'}(b)]_{k\ell} = \begin{cases} H_{r',j}(b) & \text{if } k,\ell \in \Omega_{r'} \\ 0 & \text{otherwise.} \end{cases}$$

We note

$$(z - \tilde{H}_j(b))^{-1} - (z - \tilde{H}_j^{r'}(b))^{-1} = (z - \tilde{H}_j(b))^{-1}(\tilde{H}_j(b) - \tilde{H}_j^{r'}(b))(z - \tilde{H}_j^{r'}(b))^{-1}.$$

Now $[\tilde{H}_j(b) - \tilde{H}_j^{r'}(b))]_{ts}$ is only nonzero if t or $s \notin \Omega_{r'}$. We use the definition

$$\tilde{\Omega} \setminus \Omega_{r'} := \{x : x \in \tilde{\Omega}, x \notin \Omega_{r'}\}.$$

From Lemma 4.1, we have

$$|[(z - \tilde{H}_j(b))^{-1}]_{st}| \le \tilde{C}\tilde{d}^{-1}e^{-\gamma\tilde{d}(1+\kappa)|\Re(s)-\Re(t)|}$$

and

$$|[(z - \tilde{H}_j^{r'}(b))^{-1}]_{st}| \le \tilde{C}\tilde{d}^{-1}e^{-\gamma\tilde{d}(1+\kappa)|\Re(s)-\Re(t)|}.$$

Therefore, we obtain the bound

$$\begin{aligned}
&\left|[(z - \tilde{H}_j(b))^{-1} - (z - \tilde{H}_j^{r'}(b))^{-1}]_{k\ell}\right| \\
&\quad\le \sum_{t\in\tilde{\Omega}} \sum_{s\in\tilde{\Omega}\setminus\Omega_{r'}} \left|[(z - \tilde{H}_j(b))^{-1}]_{kt}[\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)]_{ts}[(z - \tilde{H}_j^{r'}(b))^{-1}]_{s\ell}\right| \\
&\qquad + \sum_{s\in\tilde{\Omega}} \sum_{t\in\tilde{\Omega}\setminus\Omega_{r'}} \left|[(z - \tilde{H}_j(b))^{-1}]_{kt}[\tilde{H}_j(b) - \tilde{H}_j^{r'}(b)]_{ts}[(z - \tilde{H}_j^{r'}(b))^{-1}]_{s\ell}\right| \\
&\quad\le C\tilde{d}^{-2} \sum_{t\in\tilde{\Omega}} \sum_{s\in\tilde{\Omega}\setminus\Omega_{r'}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)} \\
&\qquad + C\tilde{d}^{-2} \sum_{s\in\tilde{\Omega}} \sum_{t\in\tilde{\Omega}\setminus\Omega_{r'}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)}.
\end{aligned}$$

Here we consider one of the terms on the right-hand side. First, we observe

$$\sum_{t\in\tilde{\Omega}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|} \le C' \int_{\mathbb{R}^2} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-y|+|y-\Re(s)|)}dy.$$

Using alternative elliptic coordinates, letting $a = \frac{1}{2}|\Re(k) - \Re(s)|$, we get

$$\begin{aligned}
&\sum_{t\in\tilde{\Omega}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|)} \\
&\quad\le Ca^2 \int_{-1}^{1}\int_{1}^{\infty} \frac{\sigma^2-\tau^2}{\sqrt{(\sigma^2-1)(\tau^2-1)}} e^{-2a\gamma\tilde{d}(1+\kappa)\sigma} d\sigma d\tau \\
&\quad\le C'a^2\left(\int_0^\infty \frac{(\sigma+1)^2}{\sqrt{\sigma(\sigma+2)}} e^{-2a\gamma\tilde{d}(1+\kappa)\sigma}\right) d\sigma e^{-2a\gamma\tilde{d}(1+\kappa)} \\
&\quad\le C''a^2\left(\int_0^1 \sigma^{-1/2}e^{-2a\gamma\tilde{d}\sigma}d\sigma + \int_1^\infty \sigma e^{-2a\gamma\tilde{d}\sigma}d\sigma\right) e^{-2a\gamma\tilde{d}(1+\kappa)} \\
&\quad\le C'''(a^{3/2}\tilde{d}^{-1/2} + \tilde{d}^{-2})e^{-2a\gamma\tilde{d}(1+\kappa)} \\
&\quad\le C'''(|\Re(k) - \Re(s)|^{3/2}\tilde{d}^{-1/2} + \tilde{d}^{-2})e^{-\gamma(1+\kappa)\tilde{d}|\Re(k)-\Re(s)|} \\
&\quad\le C''''\tilde{d}^{-2}e^{-\gamma\tilde{d}|\Re(k)-\Re(s)|}.
\end{aligned}$$

We find some κ_0 such that $(1+\kappa_0)^2 < (1+\kappa)$. Then using the above argument twice using κ_0 in place of κ, we have

$$\sum_{t\in\tilde{\Omega}}\sum_{s\in\tilde{\Omega}\setminus\Omega_{r'}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)} \le \tilde{C}\tilde{d}^{-4}e^{-\gamma\tilde{d}|\Re(k)-\Re(\ell)|}.$$

Alternatively, if we use it once, then we obtain

$$\begin{aligned}
&\sum_{t\in\tilde{\Omega}}\sum_{s\in\tilde{\Omega}\setminus\Omega_{r'}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)}\\
&\quad\le C'''\tilde{d}^{-2}\sum_{s\in\tilde{\Omega}\setminus\Omega_{r'}} e^{-\gamma\tilde{d}(|\Re(k)-\Re(s)|+|\Re(s)-\Re(\ell)|)}\\
&\quad\le \tilde{C}'\tilde{d}^{-2}\int_{|y|>r'} e^{-\gamma\tilde{d}(|\Re(k)-y|+|\Re(\ell)-y|)}dy\\
&\quad\le \tilde{C}'\tilde{d}^{-2}\int_{|y|>r'} e^{-\gamma\tilde{d}|x-y|}dy, \qquad \text{for } x=\Re(k) \text{ or } x=\Re(\ell)\\
&\quad\le \tilde{C}'\tilde{d}^{-2}\int_{|y|>r'} e^{-\gamma\tilde{d}(|y|-|x|)}dy.
\end{aligned}$$

Therefore, we deduce that

$$\begin{aligned}
&\sum_{t\in\tilde{\Omega}}\sum_{s\in\tilde{\Omega}\setminus\Omega_{r'}} e^{-\gamma\tilde{d}(1+\kappa)(|\Re(k)-\Re(t)|+|\Re(t)-\Re(s)|+|\Re(s)-\Re(\ell)|)}\\
&\qquad\le \tilde{C}''\tilde{d}^{-4}r'e^{-\gamma\max\{\tilde{d}(r'-|\Re(k)|),\tilde{d}(r'-|\Re(\ell)|)\}}.
\end{aligned}$$

Finally, we can conclude that

$$\begin{aligned}
&\left|[(z-\tilde{H}_j(b))^{-1}]_{k\ell} - [(z-H_{r',j}(b))^{-1}]_{k\ell}\right|\\
&\qquad\le C'\tilde{d}^{-6}\min\{e^{-\gamma\tilde{d}|\Re(k)-\Re(\ell)|}, r'e^{-\gamma\tilde{d}\max\{|r'-|\Re(k)|,r'-|\Re(\ell)|\}}\}.
\end{aligned}$$ ∎

Lemma 4.2 shows that the resolvent difference is bounded by the site distances from the edge of the first cutoff region (the circle with radius r') and the distance between the two sites. This is consistent with Lemma 4.1.

Let $\mathcal{C}$ be a contour around $S[H]$ such that $\tilde{d}/2 < d(\mathcal{C}, S[H]) < \tilde{d}$. By Lemma 4.2, we have for $g\in\Lambda_{\tilde{d}}$ and $r<r'$ that

$$\begin{aligned}
&|\mathcal{D}_\alpha[H_{r,j}(b)](g) - \mathcal{D}_\alpha[H_{r',j}(b)](g)|\\
&\quad= \left|\frac{1}{2\pi i}\oint_{\mathcal{C}} g(z)\Big([(z-H_{r,j}(b))^{-1}]_{0\alpha,0\alpha} - [(z-H_{r',j}(b))^{-1}]_{0\alpha,0\alpha}\Big)dz\right|\\
&\quad\le C'\tilde{d}^{-6}r'\sup_{z\in\mathcal{C}}|g(z)|e^{-\gamma\tilde{d}r'}.
\end{aligned}$$

Hence $\{\mathcal{D}_\alpha[H_{r_n,j}(b)]\}_n$ is a Cauchy sequence for $r_n\to\infty$, which therefore has some limit $\mathcal{D}_\alpha[H](b,g)$. $\mathcal{D}_\alpha[H]$ is linear in g, since each element of the Cauchy sequence is linear. Further, we have the error bound

$$|\mathcal{D}_\alpha[H](b,g) - \mathcal{D}_\alpha[H_{r,j}(b)](g)| \le C'\tilde{d}^{-6}r\sup_{z\in\mathcal{C}}|g(z)|e^{-\gamma\tilde{d}r}.$$

Since the linear functional $\mathcal{D}_\alpha[H_{r,j}(b)]$ is bounded by $\|\mathcal{D}_\alpha[H_{r,j}(b)]\|\le 1$ we also obtain that $\mathcal{D}_\alpha[H](b,\cdot)$ is a bounded linear functional and so has a unique extension to a bounded linear functional on the space $C(S[H])$.

This completes the proof of Theorem 2.2.

4.3. Proof of Theorem 2.3.

LEMMA 4.3. *Suppose $h_{\alpha\alpha'} \in C^n(\mathbb{R}^2)$ for $n \in \mathbb{N} \cup \{\infty\}$ and $\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}$ is uniformly continuous for $m + m' \le n$. We further assume the decay estimate*

$$|\partial_{b_1}^m \partial_{b_2}^{m'} h_{\alpha\alpha'}(x)| \le C_{mm'} e^{-\gamma'_{mm'}|x|}. \tag{4.2}$$

Then for $k = 0\alpha$ and $d(\{z\}, S[H]) > 0$, we have $b \mapsto [(z - H_{r,j}(b))^{-1}]_{kk} \in C^n_{\mathrm{per}}(\Gamma_{P_j})$ for all $b \in \mathbb{R}^2$ and fixed $r > 0$, and we have the limit

$$b \mapsto \lim_{r\to\infty} [(z - H_{r,j}(b))^{-1}]_{kk} \in C^n_{\mathrm{per}}(\Gamma_{P_j})$$

for all $b \in \mathbb{R}^2$. Furthermore, for all $b \in \mathbb{R}^2$, $z \mapsto [(z - H_{r,j}(b))^{-1}]_{kk}$ is analytic in $\mathbb{C} \setminus S[H]$.

Proof. We will only consider the derivative ∂_{b_1}; the treatment of higher (and lower) order derivatives follow the same line of argument but are more cumbersome. We drop the subscript of γ'_{10} for brevity. Let $k = 0\alpha$ for some $\alpha \in \mathcal{A}_j$, and then

$$\partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk} = \sum_{s,\ell\in\Omega_r} [(z - H_{r,j}(b))^{-1}]_{ks} [\partial_{b_1} H_{r,j}(b)]_{s\ell} [(z - H_{r,j}(b))^{-1}]_{\ell k}.$$

Lemma 4.2 implies that, for $r > r' > 0$,

$$\begin{aligned} R(r, r', k, s) &:= \left| [(z - H_{r,j}(b))^{-1}]_{ks} - [(z - H_{r',j}(b))^{-1}]_{ks} \right| \\ &\le C \min\{e^{-\gamma|\Re(k)-\Re(s)|}, r' e^{-\gamma \min\{r' - |\Re(k)|, r' - |\Re(s)|\}}\}, \end{aligned}$$

where C and γ are independent of r. Since we are trying to define the limit and are not interested in the convergence rate, we let $\tilde{d} = 1$ in our use of Lemma 4.2. Note also that, for $s, \ell \in \Omega_{r'}$, we have

$$\partial_{b_1}[H_{r,j}(b)]_{s\ell} = \partial_{b_1}[H_{r',j}(b)]_{s\ell}.$$

Recalling that $\Re(k) = \Re(0\alpha) = 0$ and employing (4.2), we estimate

$$\begin{aligned} &\left|\partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk} - \partial_{b_1}[(z - H_{r',j}(b))^{-1}]_{kk}\right| \\ &\le C \Bigg(\sum_{s,\ell\in\Omega_{r'}} \left(R(r, r', k, s) e^{-\gamma|\Re(\ell)-\Re(k)|} + R(r, r', \ell, k) e^{-\gamma|\Re(s)-\Re(k)|} \right) |\partial_{b_1}[H_{r,j}(b)]_{s\ell}| \\ &\qquad + \sum_{s\in\Omega_r, \ell\in\Omega_r\setminus\Omega_{r'}} \left| [(z - H_{r,j}(b))^{-1}]_{ks} [\partial_{b_1} H_{r,j}(b)]_{s\ell} [(z - H_{r,j}(b))^{-1}]_{\ell k} \right| \Bigg) \\ &\le C' \left(\sum_{s,\ell\in\Omega_{r'}} \left(R(r, r', k, s) e^{-\gamma|\Re(\ell)|} + R(r, r', \ell, k) e^{-\gamma|\Re(s)|} \right) e^{-\gamma'|\Re(s)-\Re(\ell)|} + r' e^{-\gamma r'} \right). \end{aligned}$$

Here the bound on the second term comes from the same argument as in Lemma 4.2. We then have

$$\begin{aligned} &\left|\partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk} - \partial_{b_1}[(z - H_{r',j}(b))^{-1}]_{kk}\right| \\ &\quad \le C'' r' \sum_{s,\ell\in\Omega_{r'}} e^{-\gamma(r' - |\Re(s)|) - \gamma|\Re(\ell)| - \gamma'|\Re(s)-\Re(\ell)|} + C' r' e^{-\gamma r'} \\ &\quad \le C''' r'^2 \sum_{s\in\Omega_{r'}} e^{-\gamma(r' - |\Re(s)|) - \min\{\gamma,\gamma'\}|\Re(s)|} + C' r' e^{-\gamma r'} \\ &\quad \le \tilde{C} e^{-\gamma'' r'} \end{aligned}$$

for any choice of $\gamma'' < \min\{\gamma, \gamma'\}$, where $\tilde{C}$ depends on the choice of γ''.

Therefore, as $r_n \to \infty$, $\partial_{b_1}[(z - H_{r_n,j}(b))^{-1}]_{kk}$ forms a Cauchy sequence and in particular has a limit

$$L_1(b) := \lim_{r\uparrow\infty} \partial_{b_1}[(z - H_{r,j}(b))^{-1}]_{kk}.$$

Likewise, using that $[(z - H_{r,j}(b))^{-1}_{kk}$ forms a Cauchy sequence, we define

$$L(b) := \lim_{r\uparrow\infty}[(z - H_{r,j}(b))^{-1}]_{kk}.$$

We need to show that $\partial_{b_1} L$ exists and satisfies

$$\partial_{b_1} L = L_1.$$

We denote for fixed $r > 0$

$$\mathrm{Res}_r(b) = [(z - H_{r,j}(b))^{-1}]_{kk}.$$

Since $\partial_{b_1} h_{\alpha\alpha'}$ is uniformly continuous there exists a modulus of continuity ω such that $|\partial_{b_1} h(b) - \partial_{b_1} h(b')| \le \omega(|b - b'|)$. We then observe that, for $\epsilon > 0$, $\epsilon \ll |\mathrm{Im}(z)|$, and $e_1 = (1, 0)$

$$\delta H_r = H_{r,j}(b + \epsilon) - H_{r,j}(b) = O(\epsilon\omega(\epsilon)),$$

where the remainder is r-independent. Then we have

$$\begin{aligned}
&\frac{1}{\epsilon}\Big(\mathrm{Res}_r(b + \epsilon e_1) - \mathrm{Res}_r(b)\Big) \\
&\quad = \frac{1}{\epsilon}\left[(z - H_{r,j}(b))^{-1}\left(\sum_{n\ge 0}(\delta H_r(z - H_{r,j}(b))^{-1})^n - 1\right)\right]_{kk} \\
&\quad = \frac{1}{\epsilon}\left[(z - H_{r,j}(b))^{-1}\sum_{n\ge 1}(\delta H_r(z - H_{r,j}(b))^{-1})^n\right]_{kk}.
\end{aligned}$$

Using the ℓ^2 norm, we have

$$\begin{aligned}
\left|\frac{1}{\epsilon}\Big(\mathrm{Res}_r(b + \epsilon e_1) - \mathrm{Res}_r(b)\Big)\right| &\le \frac{1}{\epsilon}\|(z - H_{r,j}(b))^{-1}\|_2 \sum_{n\ge 1}\|\delta H_r\|_2^n \|(z - H_{r,j}(b))^{-1}\|_2^n \\
&\le O\left(\sum_{n\ge 1}|\mathrm{Im}(z)|^{-1-n}\epsilon^{n-1}\omega(\epsilon)^n\right) \\
&\le O(|\mathrm{Im}(z)|^{-1}\omega(\epsilon)).
\end{aligned}$$

This bound is independent of r. Hence

$$\frac{1}{\epsilon}\Big([L(b + \epsilon e_1) - L(b)\Big) = L_1(b) + O(\omega(\epsilon)).$$

Letting $\epsilon \to 0$ shows that $L \in C^{(1,0)}_{\mathrm{per}}(\Gamma_j)$ and $\partial_{b_1} L = L_1$, which is the desired result.

Continuity of L_1 with respect to b follows the same argument. Analyticity with respect to z follows from [7, section 5.2]. ∎

Theorem 2.3 follows immediately from Lemma 4.3.

4.4. Proof of Theorem 2.4. Without loss of generality, let $j = 1$. Fix $g \in \Lambda, r > 0$ and $\eta < 1$. Then we have

$$\begin{aligned}\mathcal{D}[H_{r,1}(0)](g) &= \frac{1}{|\Omega_r|}\sum_{k\in\Omega_r}\mathcal{D}_k[H_{r,1}(0)](g)\\ &= \frac{1}{|\Omega_r|}\left(\sum_{k\in\Omega_r\setminus\Omega_{\eta r}}\mathcal{D}_k[H_{r,1}(0)](g) + \sum_{k\in\Omega_{\eta r}}\mathcal{D}_k[H_{r,1}(0)](g)\right).\end{aligned}$$

We define $\mathfrak{A} : \Omega \to \mathcal{A}_1 \cup \mathcal{A}_2$ such that $\mathfrak{A}(R\alpha) = \alpha$. By Lemma 4.2, we have for $k = R\alpha \in \Omega_{\eta r}$ and $\alpha \in \mathcal{A}_j$ that

$$|\mathcal{D}_k[H_{r,1}(0)](g) - \mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}\circ\mathfrak{R}(k), g)| \le C\sup_{z\in\mathcal{C}}|g(z)|e^{-\gamma r(1-\eta)}.$$

The site k is at least a distance $r(1-\eta)$ from the boundary of Ω_r.

Consider the distribution

$$\mathcal{D}[H](g) = \nu\sum_{j=1}^{2}\sum_{\alpha\in\mathcal{A}_j}\int_{\Gamma_{P_j}} D_\alpha[H](b,g)db.$$

Since the integrand is continuous with respect to b (see Theorem 2.3) the integration is well-defined. We now estimate

$$\begin{aligned}&|\mathcal{D}[H](g) - \mathcal{D}[H_{r,1}(0)](g)|\\ &\quad\le \left|\frac{1}{|\Omega_r|}\sum_{k\in\Omega_r\setminus\Omega_{\eta r}}\mathcal{D}_k[H_{r,1}(0)](g)\right|\\ &\qquad + \left|\mathcal{D}[H](g) - \frac{1}{|\Omega_{\eta r}|}\sum_{j=1}^{2}\sum_{R\alpha\in\Omega_{\eta r}:\alpha\in\mathcal{A}_j}\mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}(R), g)\right|\\ &\qquad + \left|\frac{1}{|\Omega_{\eta r}|}\sum_{j=1}^{2}\sum_{R\alpha\in\Omega_{\eta r}:\alpha\in\mathcal{A}_j}\mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}(R), g)\right.\\ &\qquad\qquad\left. - \frac{1}{|\Omega_{\eta r}|}\sum_{k\in\Omega_{\eta r}}\mathcal{D}_k[H_{r,1}(0)](g)\right|\\ &\qquad + \left(1 - \frac{|\Omega_{\eta r}|}{|\Omega_r|}\right)\frac{1}{|\Omega_{\eta r}|}\left|\sum_{k\in\Omega_{\eta r}}\mathcal{D}_k[H_{r,1}(0)](g)\right|.\end{aligned}$$

The first and fourth terms are easily seen to be bounded by $O(1-\eta^2)$. For the second term, we let

$$\Omega_{\eta r}^j = \{R\alpha : R\alpha \in \Omega_{\eta r}, \alpha \in \mathcal{A}_j\}.$$

Then

$$\frac{1}{|\Omega_{\eta r}|}\sum_{j=1}^{2}\sum_{R\alpha\in\Omega_{\eta r}:\alpha\in\mathcal{A}_j}\mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}(R), g) = \frac{1}{|\Omega_{\eta r}|}\sum_{j=1}^{2}\sum_{R\alpha\in\Omega_{\eta r}^j}\mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}(R), g).$$

We then use $\frac{|\Omega^1_{\eta r}|}{|\Omega^2_{\eta r}|} \to \frac{|\Gamma_2|\cdot|\mathcal{A}_1|}{|\Gamma_1|\cdot|\mathcal{A}_2|}$, $|\Omega_{\eta r}| = |\Omega^1_{\eta r}| + |\Omega^2_{\eta r}|$, and Theorem 2.1 to get

$$\frac{1}{|\Omega_{\eta r}|}\sum_{j=1}^{2}\sum_{R\alpha\in\Omega_{\eta r}:\alpha\in\mathcal{A}_j}\mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}(R),g) \to \mathcal{D}[H](g),$$

and hence the second term goes to zero as $r\to\infty$. Finally, the third term can be estimated by

$$\left|\frac{1}{|\Omega_{\eta r}|}\sum_{j=1}^{2}\sum_{R\alpha\in\Omega_{\eta r}:\alpha\in\mathcal{A}_j}\mathcal{D}_\alpha[H](\mathrm{mod}_{P_j}(R),g) - \frac{1}{|\Omega_{\eta r}|}\sum_{R\alpha\in\Omega_{\eta r}}\mathcal{D}_{R\alpha}[H_{r,1}(0)](g)\right| \le C\sup_{z\in\mathcal{C}}|g(z)|e^{-\gamma r(1-\eta)}.$$

Therefore if we choose a pair of sequences $(\eta_j),(r_j)$ such that $\eta_j\uparrow 1$, $r_j\uparrow\infty$, and $r_j(1-\eta_j)\to\infty$, we conclude that

$$\mathcal{D}[H_{r,1}(0)](g)\to\mathcal{D}[H](g).$$

Since $\mathcal{D}[H]$ is a bounded linear functional, it can be extended as before to be a bounded linear functional over $C(S[H])$.

4.5. Proof of Theorem 3.1. We denote $\tilde z=(\tilde z_1,\tilde z_2)\in\mathbb{C}^2$. Let $z\in\mathbb{C}$. Then if $c>0$ is sufficiently small and $\mathrm{Im}(\tilde z_1),\mathrm{Im}(\tilde z_2)\in(-c,c)$, we have

$$\|z-H_{r,j}(\tilde z)\|_2>0,$$

and hence $\oint_{\mathcal{C}} g(z)[(z-H_{r,j}(\tilde z))^{-1}]_{0\alpha,0\alpha}$ is analytic at $\tilde z$ satisfying $\mathrm{Im}(\tilde z_1),\mathrm{Im}(\tilde z_2)\in(-c,c)$. We pick a contour $\mathcal{C}$ enclosing $S[H]$ such that $\tilde d/2<d(\mathcal{C},S[H])<\tilde d$ and then chose $c=\kappa\tilde d$ for $\kappa>0$ sufficiently small and independent of $\tilde d$ such that

$$\|H_{r,j}(\tilde z)-H_{r,j}(\mathrm{Re}(\tilde z))\|<\tilde d/4$$

by analyticity of $H_{r,j}$ (see Figure 8). To see that we still maintain analyticity of $(z-H_{r,j}(\tilde z))^{-1}$ for this choice of c and $z\in\mathcal{C}$, we note that it suffices to show

$$\|z-H_{r,j}(\tilde z)\|_2>\tilde d/4.$$

However,

$$\|z-H_{r,j}(\tilde z)\|_2\ge\|z-H_{r,j}(\mathrm{Re}(\tilde z))\|_2-\|H_{r,j}(\mathrm{Re}(\tilde z))-H_{r,j}(\tilde z)\|_2>\tilde d/4.$$

Hence, we have analyticity. Since $\int_{\mathcal{C}} g(z)[(z-H_{r,j}(\tilde z))^{-1}]_{0\alpha,0\alpha}dz$ is analytic with respect to $\tilde z$, we can apply Theorem 9.28 of [11] twice, with respect to b_1 and b_2, to deduce

$$\left|\int_{\Gamma_{P_j}}\oint_{\mathcal{C}} g(z)[(z-H_{r,j}(b))^{-1}]_{0\alpha,0\alpha}dbdz-\frac{|\Gamma_{P_j}|}{N^2_{\mathrm{disc}}}\sum_{b\in S_{P_j}}\sum_{\alpha\in\mathcal{A}_j}\mathcal{D}_\alpha[H](b,g)\right| < C\tilde d^{-1}\sup_{d(z,S[H])<\tilde d}\left[|g(z)|e^{-\gamma''\tilde d N_{\mathrm{disc}}}\right]$$

for some $C>0$ independent of r. The result follows.

5. Conclusion. The main result of this work, Theorem 2.4, is a representation formula for the thermodynamic limit of the electronic structure of incommensurate layered heterostructures. The result is reminiscent of Bellisard's noncommutative

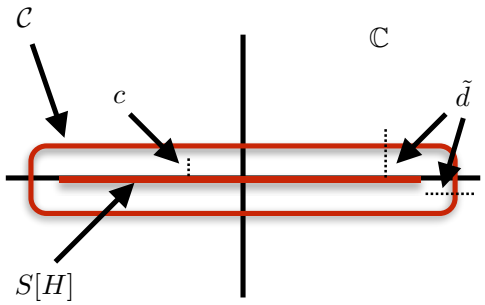


FIG. 8. *The highlighted region shows the region* $\mathbb{R} \times (-c, c)$ *enclosing* $S[H]$. *The contour* $\mathcal{C}$ *has a distance to* $S[H]$ *bounded above by* $\tilde{d}$ *and below by* $\tilde{d}/2$. *We also have* $c \sim \tilde{d}$.

Brillouin zone for aperiodic solids [1], replacing on-site randomness with a number-theoretic equidistribution theorem.

Crucially, our representation formula lends itself to numerical approximation. In section 3 we formulate, and analyze at a heuristic level, an efficient kernel polynomial method to approximately compute the density of states in twisted bilayer graphene. This preliminary exploration provides not only quantitative confirmation of our analytical results but also demonstrates the utility of our approach for applications to real material models.

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REFERENCES

[1] J. BELLISSARD, *Coherent and dissipative transport in aperiodic solids: An overview*, Dynamics of Dissipation, Springer, Berlin, 2002, pp. 413–485, https://doi.org/10.1007/3-540-46122-1_18.
[2] A. H. CASTRO NETO, F. GUINEA, N. M. R. PERES, K. S. NOVOSELOV, AND A. K. GEIM, *The electronic properties of graphene*, Rev. Modern Phys., 81 (2009), pp. 109–162, https://doi.org/10.1103/RevModPhys.81.109.
[3] H. CHEN AND C. ORTNER, *QM/MM Methods for Crystalline Defects. Part* 1, Multiscale Model. Simul., 14 (2016), pp. 232–264.
[4] A. EBNONNASIR, B. NARAYANAN, S. KODAMBAKA, AND C. V. CIOBANU, *Tunable MoS*2 *bandgap in MoS*2*-graphene heterostructures*, Appl. Phys. Lett., 105 (2014), 031603, https://doi.org/10.1063/1.4891430.
[5] S. FANG, R. KUATE DEFO, S. N. SHIRODKAR, S. LIEU, G. A. TRITSARIS, AND E. KAXIRAS, *Ab initio tight-binding Hamiltonian for transition metal dichalcogenides*, Phys. Rev. B, 92 (2015), 205108, https://doi.org/10.1103/PhysRevB.92.205108.
[6] C. HUANG, A. VOTER, AND D. PEREZ, *The kernel polynomial method*, Rev. Modern Phys., 78 (2006).
[7] T. KATO, *Perturbation Theory for Linear Operators*, Classics Math., Springer, Berlin, 1995.
[8] E. KAXIRAS, *Atomic and Electronic Structure of Solids*, Cambridge University Press, Cambridge, 2003.
[9] D. S. KODA, F. BECHSTEDT, M. MARQUES, AND L. K. TELES, *Coincidence lattices of* 2*D crystals: Heterostructure predictions and applications*, J. Phys. Chem. C, 120 (2016), pp. 10895–10908, https://doi.org/10.1021/acs.jpcc.6b01496.
[10] H.-P. KOMSA AND A. V. KRASHENINNIKOV, *Electronic structures and optical properties of realistic transition metal dichalcogenide heterostructures from first principles*, Phys. Rev. B, 88 (2013), 085318, https://doi.org/10.1103/PhysRevB.88.085318.
[11] R. KRESS, *Numerical Analysis*, Springer, Berlin, 1998.
[12] C. LE BRIS, *Computational chemistry from the perspective of numerical analysis*, Acta Numer., 14 (2005), pp. 363–444, https://doi.org/10.1017/S096249290400025X.

[13] G. C. Loh and R. Pandey, *A graphene-boron nitride lateral heterostructure—A first-principles study of its growth, electronic properties, and chemical topology*, J. Mater. Chem. C, 3 (2015), pp. 5918–5932, https://doi.org/10.1039/C5TC00539F.

[14] G. Mazzi and B. J. Leimkuhler, *Dimensional reductions for the computation of time–dependent quantum expectations*, SIAM J. Sci. Comput., 33 (2011), pp. 2024–2038.

[15] E. Prodan, *Quantum transport in disordered systems under magnetic fields: A study based on operator algebras*, Appl. Math. Res. Express AMRX, 2013, pp. 176–255.

[16] H. Röder, R. N. Silver, D. A. Drabold, and J. J. Dong, *Kernel polynomial method for a nonorthogonal electronic-structure calculation of amorphous diamond*, Phys. Rev. B, 55 (1997), pp. 15382–15385.

[17] R. N. Silver, H. Roeder, A. F. Voter, and J. D. Kress, *Kernel polynomial approximations for densities of states and spectral functions*, J. Comput. Phys., 124 (1996), pp. 115–130.

[18] H. Terrones and M. Terrones, *Bilayers of transition metal dichalcogenides: Different stackings and heterostructures*, J. Mat. Res., 29 (2014), pp. 373–382, https://doi.org/10.1557/jmr.2013.284.

[19] G. A. Tritsaris, S. N. Shirodkar, E. Kaxiras, P. Cazeaux, M. Luskin, P. Plecháč, and E. Cancès, *Perturbation theory for weakly coupled two-dimensional layers*, J. Mat. Res., 31 (2016), pp. 959–966.

[20] J. A. C. Weideman, *Numerical integration of periodic functions: A few examples*, Amer. Math. Monthly, 109 (2002), pp. 21–36.

[21] A. Weisse, G. Wellein, A. Alvermann, and H. Fehske, *The kernel polynomial method*, Rev. Modern Phys., 78 (2006), pp. 275–306, https://doi.org/10.1103/RevModPhys.78.275.

[22] A. Zorzi, *An elementary proof for the equidistribution theorem*, Math. Intelligencer, 37 (2015), pp. 1–2, https://doi.org/10.1007/s00283-014-9505-x.