Interlayer coupling in rotationally faulted multilayer graphenes

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Abstract
This paper reviews progress in the theoretical modelling of the electronic structure of rotationally faulted multilayer graphenes. In these systems the crystallographic axes of neighbouring layers are misaligned so that the layer stacking does not occur in the Bernal structure observed in three-dimensional graphite and frequently found in exfoliated bilayer graphene. Notably, rotationally faulted graphenes are commonly found in other forms of multilayer graphene including epitaxial graphenes thermally grown on SiC (000¨1), graphenes grown by chemical vapour deposition, folded mechanically exfoliated graphenes, and graphene flakes deposited on graphite. Rotational faults are experimentally associated with a strong reduction of the energy scale for coherent single particle interlayer motion. The microscopic basis for this reduction and its consequences have attracted significant theoretical attention from several groups that are highlighted in this review.

1. Introduction

Coherent interlayer motion in multilayer graphenes plays a crucial role in determining their low energy electronic properties. In single-layer graphene the absence of the layer degree of freedom cleanly exposes the geometrical structure of its low energy electronic physics. This is controlled by single particle spectra containing linearly dispersing bands around singular points at its inequivalent zone corners, described by a pair of valley-polarized two-dimensional massless Dirac Hamiltonians [1, 2]. The physics is very different for graphene bilayers that are stacked in the Bernal geometry with the ‘A’ sublattice of one layer eclipsed with the ‘B’ sublattice of its neighbour (AB stacking). Here the effects of coherent interlayer coupling are quite strong and the low energy sector is described instead by a different class layer-coherent chiral fermions with a quadratic dispersion and a Berry’s phase of $2\pi$ for reciprocal space orbits that encircle the point of degeneracy [3]. This physics is readily understood from the experimentally known strength of the interlayer tunnelling amplitude at eclipsed sites and it can be generalized to describe the low energy physics of multilayer graphenes where the crystallographic axes of neighbouring layers are rotated by special angles $\theta = n\pi/3$ [4–7].

Surprisingly, experimental work over the last five years has revealed a family of multilayer graphenes that show only weak (if any) effects of their interlayer coupling. This family includes graphenes that are grown epitaxially on the SiC (000¨1) surface [8–10], CVD grown graphenes [11] and some forms of exfoliated graphene [12–14]. A common structural attribute of these systems is a rotational misorientation (a twist) of their neighbouring layers at angles of $\theta \neq n\pi/3$. The layer decoupling has been inferred from the measurements of the magnetotransport [8, 9], of the Landau level spectra observed in scanning tunnelling spectroscopy [14, 15] and perhaps most clearly in angle-resolved photoemission spectra [16]. These experimental observations are attracting significant theoretical attention. The layer decoupling in twisted multilayers is frequently attributed to a kinematical effect whereby the layer projections of the zone corner crystal momenta are misaligned by the rotation, preventing momentum-conserving interlayer motion at sufficiently low energy [17]. In this scenario the low energy theory is described by four separate valley- and layer-polarized Dirac cones. These fermions are then recoupled at a crossover energy scale where pairs of individual Dirac cones from the two layers merge and hybridize thus changing the band topology [18]. The simplest version of this theory predicts that the residual low energy effect of the twist is to reduce the Fermi velocities by an angle-dependent factor where the smallest velocities are expected for small rotation angles. These theoretical predictions have provided a taking off point...
for further investigations of this problem using a variety of methods ranging from microscopic atomistic calculations to continuum models designed to capture selected elements of the microscopic physics. Currently there is a lively discussion concerning the theoretical interpretation of the electronic physics in twisted graphenes: what is the appropriate long wavelength theory? How can one distinguish between the electronic physics for ‘small’ and ‘large’ rotation angles? How does the interlayer coherence scale depend on the fault angle? What are the experimental consequences of the reduced interlayer coherence? It is fair to say that the one-electron physics of these systems is proving to be unexpectedly rich and it has so far eluded a satisfactory theoretical description. In this paper we briefly highlight some recent theoretical progress on this problem and focus on some of the major unresolved issues.

Section 2 presents a discussion of the geometric properties of rotationally faulted bilayers which are generally useful for analysing their structural and electronic properties. The results presented here provide a foundation for a theoretical analysis we have presented earlier [19] although these details have not been published previously. Sections 3–5 briefly review the existing theoretical approaches that have been developed for describing the electronic structure of these systems. Section 3 reviews the essential features of a long wavelength theory of layer that illustrates the physics of layer decoupling by ‘rotational mismatch’ [17]. Section 4 presents the wavelength theory of layer that illustrates the physics of layer stacking. The unit cells of these structures have the same area so that these structures are related. We refer to these structures as commensuration partners. The simplest example of such a pair occurs trivially for \( \theta = 0 \) and \( \bar{\theta} = 60^\circ - \theta \) which correspond to the smallest possible unit cells with \( AB \) (Bernal) stacking and \( AA \) (perfectly eclipsed) stacking. The unit cells of these structures have the same area but they have different sublattice symmetries. Importantly, all commensurate rotations share this property: they occur in partners where the sum of the rotation angles is \( 60^\circ \) and their unit cells have the same area. The commensuration indices \((m, n)\) and \((\bar{m}, \bar{n})\) of the partners are related

\[
(\bar{m}, \bar{n}) = (-1, 1) \begin{pmatrix} m \\ n \end{pmatrix}
\]

eliminating common divisors by 3 from the result. Figure 1 illustrates this situation where the structure in the left panel corresponds to \((m, n) = (1, 3)\), \( \theta = 32.204^\circ \) and on the right \((\bar{m}, \bar{n}) = (2, 5)\), \( \bar{\theta} = 27.796^\circ \). Partner commensurations can also be transformed into each other by a rigid translation \( \Delta \) keeping the rotation angle \( \theta \) constant, demonstrating the invariance of the primitive cell area. The structure at \( \theta = \bar{\theta} = 30^\circ \) is its own commensuration partner and corresponds to an elementary two-dimensional quasicrystalline lattice. Note also that the form of equation (1) demonstrates that the indices \((m, n)\) generally provide a more useful specification of the structure than the fault angle \( \theta \). Indeed nearby rotation angles can have very different fault indices and therefore describe crystalline structures with vastly different periods and different physical properties. A plot of the commensuration periods \(|A|\) as a function of rotation angle \( \theta \) shows a complex distribution of allowed periods which is bounded from below. This lower bound has a smooth dependence on \( \theta \), diverges as \( \theta \to 0^\circ \) and \( \theta \to 60^\circ \) and is symmetric around the self dual state at \( \theta = 30^\circ \). Commensuration partners are distinguished by their sublattice exchange parity. A commensuration is sublattice exchange ‘even’ if the commensuration cell contains an \( A \) and a \( B \) sublattice site in each layer that are coincident with atomic sites in the neighbouring layer. A commensuration is sublattice exchange ‘odd’ if only one sublattice site in the commensuration cell is eclipsed. (Fixing the rotation centre of the twist at an atom site guarantees that there will be at least one coincident site.) The sublattice exchange parity can be
described from the translation indices \((m, n)\). It is convenient to label the eclipsed sites at the origin as the \(A\) sublattice, a nearest neighbour bond vector \(\mathbf{\tau}\) and its partner in the rotated layer \(\mathbf{\tau}'\). Then the condition for a second coincident site on the \(B\) sublattice is

\[
\mathbf{T} + \mathbf{\tau} = \mathbf{T}' + \mathbf{\tau}'
\]

(5)

for some possible choice of \(\mathbf{T}(\mathbf{T}')\) in the set of lattice translations in the reference (rotated) layers. Since the \(\mathbf{T}\)’s are both lattice translations, this requires integer \((p, q)\) solutions to

\[
e^{i3\phi_{\text{ans}}} = \frac{1 + \sqrt{3}(pe^{i\pi/6} + qe^{-i\pi/6})}{1 + \sqrt{3}(pe^{-i\pi/6} + qe^{i\pi/6})} = \frac{me^{i\pi/6} + ne^{-i\pi/6}}{me^{-i\pi/6} + ne^{i\pi/6}}
\]

(6)

which can be expressed

\[
p = \frac{m - n + 3mq}{3n}.
\]

(7)

Equation (7) has integer solutions only when \(m - n\) is divisible by 3. When this is satisfied the coincident sites occur at special high symmetry points in the cell \(\mathcal{A}_c = \pm \mathcal{A}_{\text{ans}}/3\) (with only one sign per structure) which identify high symmetry positions along the diagonal of the rhombus shown in the right-hand panel of figure 1. When \(m - n\) is not divisible by 3 the only coincident site occurs at the centre of rotation and its supercell translates.

2.2. Reciprocal space

Similar considerations apply to the momentum space representation of the twisted bilayer for which figure 2 gives a map illustrating the structure of its reciprocal space. The reciprocal lattice of the commensuration supercell can be treated as a conventional triangular lattice spanned by two primitive vectors \(2\pi(\hat{e}_x \times \mathbf{A}_1)/\mathcal{A}\) where \(\mathcal{A} = |\mathbf{A}_1 \times \mathbf{A}_2|\) is the area of the commensuration supercell and \(\hat{e}_c\) is the layer normal. However, it is often useful to use the fact that the real space lattice translations of the supercell are \textit{coincident lattice translations} of each of the layers. Thus one can index its reciprocal lattice vectors with \textit{four} integer indices describing all linear combinations of the primitive reciprocal lattice translations of of the reference \((\mathbf{G})\) and rotated \((\mathbf{G}')\) vectors in the manner

\[
\mathbf{g}_{p,q,p',q'} = p\mathbf{G}_1 + q\mathbf{G}_2 + p'\mathbf{G}'_1 + q'\mathbf{G}'_2.
\]

(8)

Thus that the single-layer primitive \(\mathbf{G}\)’s and \(\mathbf{G}'\)’s and all possible combinations \(\mathbf{G} + \mathbf{G}'\) are in the reciprocal lattice of the faulted structure. The smallest nonzero combinations of these vectors have length \(4\pi/\sqrt{3(m^2 + mn + n^2)}\) spanning the first star of reciprocal lattice vectors of the commensuration supercell.

A relative rotation of the neighbouring layers displaces the momenta of the zone corners \(\mathbf{K}\) and \(\mathbf{K}'\) to new positions \(\mathbf{K}(\theta)\) and \(\mathbf{K}'(\theta)\). A critical question is whether the offsets between these momenta \(\mathbf{K}(\theta) - \mathbf{K}\) or \(\mathbf{K}(\theta) - \mathbf{K}'\) are also in the reciprocal lattice of the commensuration cell. For the

**Figure 1.** Two lattice structures for rotationally faulted graphene bilayers at complementary rotation angles. Red and blue dots denote atomic positions in different layers. The highlighted rhombus is a primitive commensuration cell. The figure compares the stacking patterns for commensuration pairs that are related by \(\theta = 60^\circ - \theta\). The structures are (left) \(\theta = 32.204^\circ (m = 1, n = 3)\) and (right) \(27.796^\circ (m = 2, n = 5)\). The commensuration cells are the same for the partner structures but the point symmetry is different.

**Figure 2.** A reciprocal space map for a twisted graphene bilayer illustrating the rotation of the first star of reciprocal lattice vectors (open dots) to a star of rotated reciprocal lattice vectors (filled blue dots), and a corresponding rotation of the zone corner \(\mathbf{K}\) points (red dots). The offset points \(\mathbf{K}\) and \(\mathbf{K}(\theta)\) become coincident in the extended zone after translations by a particular pair of reciprocal lattice vectors.
former situation this is the question of whether
\[ \vec{K}(\theta) - \vec{K} = \vec{g}_{p,q,p',q'} = p\vec{G}_1 + q\vec{G}_2 + p'\vec{G}'_1 + q'\vec{G}'_2 \]  
(9)
for some choice of integers \((p, q, p', q')\). Representing these two-dimensional vectors by complex numbers one finds that equation (9) can be expressed
\[ e^{i\theta_m} = \frac{1 + \sqrt{3}(p e^{i\pi/6} + q e^{-i\pi/6})}{1 + \sqrt{3}(p e^{-i\pi/6} + q' e^{i\pi/6})} \]  
(10)
where \(\theta_m\) is given by equation (2). Nontrivial solutions invert the indices \(p' = q\) and \(q' = p\) and lead to the matching condition
\[ p = \frac{m - n - 3mq}{3n}. \]  
(11)
Thus \(\vec{K}(\theta) - \vec{K}\) is in the reciprocal lattice only for supercommensurate structures where \(m - n\) is a multiple of 3. Equation (11) is identical to equation (7) that identifies the even sublattice exchange commensurations, so that sublattice ‘even’ structures always allow intravalley interlayer coupling. For example, when \((m, n) = (2, 5)\) we have \(\theta = 27.796^\circ\) and the first integer solutions to equation (10) occur for \(q = 3\) for which \(p = 1\) and \((p', q') = (3, 1)\). The existence of this solution implies that these \(K\) points are coincident in the extended zone after translations by \(p\vec{G}_1 + q\vec{G}_2\) and \(p'\vec{G}'_1 + q'\vec{G}'_2\) as illustrated in the right panel of figure 3.

One can also ask about the possibility of commensurability for intravalley momentum transfer \(K(\theta) - K'\), namely
\[ \vec{K}(\theta) - \vec{K}' = \vec{g}_{p,q,p',q'} = p\vec{G}_1 + q\vec{G}_2 + p'\vec{G}'_1 + q'\vec{G}'_2. \]  
(12)
Following a similar line of analysis one finds a different set of commensurability conditions
\[ p = \frac{m(q + 1) - nq}{m + 2n} \]
\[ p' = \frac{mq - (1 + q)n}{m + 2n} \]
\[ q' = \frac{(2m + n)q + m}{m + 2n}. \]  
(13)
Thus for example, \(m = 1, n = 3\) gives a rotation angle \(\theta = 32.204^\circ\) which has its first integer solution when \(\vec{q} = q = 4\), giving \((p, q) = (-1, 4)\) and \((p', q') = (1, 3)\). Note the asymmetry between the values of \((p, q)\) and \((p', q')\): the scattering between inequivalent Dirac cones requires different umklapp terms when indexed to the individual reciprocal lattices of the two layers. The indices would be reversed by considering \(K \rightarrow K'(\theta)\) couplings. This matching rule implies that \(\vec{K}(\theta)\) and \(\vec{K}'\) are coincident in after translations by \(p\vec{G}_1 + q\vec{G}_2\) and \(p'\vec{G}'_1 + q'\vec{G}'_2\) as illustrated in the left panel of figure 3.

One can prove that equations (9) and (12) cannot be simultaneously satisfied for a common rotation angle. For example if \(m = 3\mu + 1\) and \(n = 3\nu + 1\) then \(m - n\) is a multiple of 3 and intravalley couplings are in the reciprocal lattice. In this situation the commensurability condition for intervalley coupling requires an integer solution to equation (13)
\[ p = \frac{3(\mu q - \nu q + \mu) + 1}{3(\mu + 2\nu + 1)} \]  
(14)
which is impossible since the numerator is never divisible by 3 so that intervalley coupling is excluded. On the other hand, when \(m = 3\mu ± 1\) and \(\nu = 3\nu ± 1\) intravalley is excluded and
\[ p = \frac{3(\mu - \nu)q + \mu + 2q + 1}{3(\mu + 2\nu + 1)} \]  
(15)
which can be satisfied for integer \(p\) by appropriate choice of integer \(q\) so that intervalley coupling is allowed. These two possibilities are thus complementary and mutually exclusive: one or the other must occur if the rotation is commensurate. Using the indexing rule equation (4), one can easily show that partner commensurations realize complementary commensuration conditions: one member admits only the intravalley interlayer coupling while the other allows only the analogous intervalley scattering. Thus the valley structure of the interlayer couplings are specified by the sublattice exchange symmetry of the structure.

3. Layer decoupling by rotational mismatch

Early work on the electronic properties of twisted graphene bilayers recognized that small angle rotational faults are inevitably described by large period commensuration cells that make atomistic calculations impractical. Instead it is useful to develop a long wavelength description that captures the effect of rotation on the low energy electronic structure. The essential physics in this treatment is the momentum offset of the Dirac nodes produced by the rotation [17] as illustrated in figure 2.

The starting point of the continuum theory is the long wavelength theory appropriate to single-layer graphene. The effective mass theory for electrons in each valley introduces two Dirac Hamiltonians for their smoothly varying pseudospinor fields
\[ H_K = -i\hbar v_F \sigma \cdot \nabla; \quad H_{K'} = \sigma_L H_K \sigma_L \]  
(16)
where the \(\sigma_s\) are \(2 \times 2\) Pauli matrices acting on the sublattice amplitudes. A small angle relative rotation of the crystallographic axes of the two layers offsets the crystal momenta of their closest Dirac nodes by \(\Delta K = 2K \sin(\theta/2)\).

This can be described by a pair of layer-polarized Dirac Hamiltonians parametrized by the momentum offset \(\Delta \vec{K} = \vec{K}(\theta) - \vec{K}\) in the manner
\[ H_K = h v_F \sigma \cdot \left[-i \nabla - \frac{\Delta \vec{K}}{2}\right] \]
\[ H_{K(\theta)} = h v_F \sigma^\theta \cdot \left[-i \nabla + \frac{\Delta \vec{K}}{2}\right] \]  
(17)
where \(\sigma^\theta = \exp(i\sigma, \theta/2)\sigma_L \exp(-i\sigma, \theta/2)\) because of the relative rotation of the two layers.

Electrons in neighbouring layers are coupled by a \(\theta\)-dependent interlayer coupling amplitude projected into the
pseudospin basis. For small angle rotations these interlayer amplitudes vary smoothly in real space and one can focus on their lowest Fourier components. In the theory of Lopes dos Santos et al [17] the offset momentum $\Delta K$ is not in the reciprocal lattice of the commensuration cell and these authors focus on the three momenta $\vec{G}_i$ that leave the offset $[\vec{K}(\theta) - \vec{K}]$ invariant. These momenta can be expressed in terms of the offset $\Delta K$: in complex notation they are $G_i = (0, \bar{G}_i = \sqrt{3}e^{i\pi/6} \Delta K, \vec{G}_i + \vec{G}_2 = \sqrt{3}e^{-i\pi/6} \Delta K)$. In the pseudospin basis, the interlayer coupling for each of these momentum transfers is characterized by a $2 \times 2$ matrix-valued tunnelling coefficient $T(\vec{G})$ whose elements have been estimated numerically using a tight binding model. This yields in the small angle limit
\[ T(\vec{G} = 0) = \tilde{t}_\perp \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix}; \quad T(\vec{G} = -\vec{G}_1) = \tilde{t}_\perp \begin{pmatrix} \tilde{z} & 1 \\ \bar{z} & \tilde{z} \end{pmatrix}, \]
\[ T(\vec{G} = -\vec{G}_1 + \vec{G}_2) = \tilde{t}_\perp \begin{pmatrix} \bar{z} & 1 \\ \tilde{z} & \bar{z} \end{pmatrix}, \]
where $z = e^{2\pi i/3}, \bar{z} = e^{-2\pi i/3}$ and $\tilde{t}_\perp \sim 0.11 \text{eV}$, approximately independent of the supercell period.

The asymmetry in the set of selected $\vec{G}$‘s appearing in equation (18) occurs because of the choice of the reference valley for the long wavelength expansion. Nevertheless, this approach explicitly preserves the threefold rotational symmetry of the commensuration cell. This is seen most clearly by observing that the $T$ matrices are off-diagonal operators in the layer degree of freedom and one may therefore arbitrarily ‘shift’ the interlayer coupling momenta by a layer-dependent $U(1)$ gauge transformation. In particular the gauge shift $e^{-i\Delta K \vec{p}}$ in the rotated layer brings the two Dirac nodes into coincidence and shifts the three momentum transfers so that they form the three arms of a star generated by $Q = -\Delta K$ and its $\pm 2\pi/3$-rotated partners. The three processes with momentum transfers $-Q$ represent three complementary layer-reversed interlayer transfer processes. Thus the expansion about a single zone corner point preserves the full three fold symmetry of the commensuration cell, as required.

The essential features of this theory are (1) the existence of a crystal momentum offset due to the rotational fault, (2) the coupling of plane wave states in one layer to a triad of plane wave states in its neighbour and (3) the existence of a $G = 0$ term in the effective interlayer tunnelling Hamiltonian. Feature (1) suggests that at sufficiently low energy the effect of the interlayer coupling can be treated perturbatively in the dimensionless coupling parameter $\Gamma = \tilde{t}_\perp/\hbar v_F \Delta K$. Feature (2) implies the perturbative effects of this coupling vanish by symmetry precisely at $E = 0$ so that the coupled system preserves the Dirac nodes of its two (decoupled) layers. Perturbative effects of the coupling arise at linear order in the momentum differences $\delta \pm \Delta K/2$ and can be interpreted as a twist-dependent renormalization of the Fermi velocity
\[ \frac{v_F^2}{v_F} = 1 - 9 \left( \frac{\tilde{t}_\perp}{\hbar v_F \Delta K} \right)^2. \] 
Equation (19) needs to be applied with care since it breaks down both in the limit of small rotation angles due to a failure of the perturbation theory when $\Delta K \rightarrow 0$ and at large rotation angles when commensuration effects, neglected in this treatment, can intervene. Finally, feature (3) indicates that electron states in the two layers that have the same crystal momentum modulo $G$ are coupled through the interlayer Hamiltonian. In the low energy theory the layer-polarized Dirac cones degenerate in the planes that bisect lines connecting their nodes (above the crossover energy $\hbar v_F \Delta K/2$) and one expects the strongest interlayer mixing to occur in these planes. There are three such planes that bisect the lines $\Delta K$ and its $\pm 2\pi/3$ rotated counterparts. The onset of this mixing is associated with a change of topology of the bilayer bands, connecting a low energy sector with layer-decoupled Dirac cones to higher energy layer-coherent hyperbolic bands. In the lowest band this transition is associated with a saddle point in the electronic spectrum and a logarithmic van Hove singularity in the two-dimensional density of states [18].
4. Atomistic calculations

The novel physics of rotationally induced layer decoupling has stimulated theoretical work by several groups to explore this effect using various atomistic models. Ab initio calculations have been carried out for rotationally misaligned bilayer supercells containing up to \( \sim500 \) atoms (\( \theta \sim 5^\circ \)) while tight binding methods have allowed workers to access larger systems of up to 15,000 atoms [10, 21–24]. These studies have examined the Fermi velocity renormalization, the form of the low energy electronic spectrum near the \( K \) points and the spatial modulation of the electronic charge density.

Much of the \textit{ab initio} work has understandably focused on the shortest period twisted structures, e.g. \( \sqrt{7} \times \sqrt{7} \) and \( \sqrt{13} \times \sqrt{13} \) commensurations [10, 23]. Calculations on these systems generally confirm a suppression of the interlayer coupling scale and a Fermi velocity near the \( K \) point which is essentially indistinguishable from that of single-layer graphene. The most thorough investigation of the Fermi velocity renormalization has been given by de Laissardière \textit{et al} [22] who suggest that the rotational faults are characterized by three different velocity renormalization regimes, determined by the fault angle: (a) \( 15^\circ < \theta < 30^\circ \) where the Fermi velocity is essentially the same as for single-layer graphene, (b) \( 3^\circ < \theta < 15^\circ \) where a downward renormalization is found, well described by the perturbation theory of [17], and (c) a low angle regime \( \theta < 3^\circ \) where the low energy bands are flattened and not described by the perturbative treatment. The small renormalization in the large fault angle regime (a) is at least qualitatively consistent with the continuum theory since the renormalization occurs via a virtual mixing of low energy states with states separated by an energy barrier \( \hbar v_F \Delta K \). The breakdown of the perturbation theory for sufficiently small angle faults is similarly understandable since it involves an expansion of the perturbation theory for sufficiently small angle faults.

5. Second generation continuum theories

There has been progress in the development of new long wavelength models that extend the physics identified in the original continuum formulation [17]. These theories examine the effects of lattice commensuration [19] and of multiband mixing [25] on the low energy electronic structure. The former turns out to be most important for special large angle faults while the latter is critical to the physics at small rotation angles. The new models are also formulated as continuum theories in order to circumvent the technical difficulty posed by fully microscopic atomistic treatments of large commensuration cells. Concurrently there has been an effort to distill the original continuum model to a simpler effective two band model [27, 28] in an effort to explore the effects of the novel band topology on the orbital quantization of its electronic states in a perpendicular magnetic field. We refer to all these new models as ‘second generation’ continuum theories.

5.1. Interlayer matrix elements

A microscopic theory of the interlayer coupling can be formulated in the basis of two Bloch orbitals

\[
\langle \psi_a(\mathbf{k}) | H | \psi_b(\mathbf{k}) \rangle = \frac{1}{N} \sum_{\mathbf{T}} e^{i \mathbf{k} \cdot \mathbf{T}} \phi_a(\mathbf{T}) \phi_b(\mathbf{T})
\]

where \( \phi_{a\alpha(\mathbf{A}, \mathbf{B})} \) are orbitals centred at positions \( \mathbf{T} + \mathbf{a}_\alpha \) and \( \mathbf{T} \) is a lattice translation. In this basis the interlayer Hamiltonian is

\[
\langle \psi_{\beta}(\mathbf{\bar{K}}') | H | \psi_a(\mathbf{\bar{K}}) \rangle = \frac{1}{N} \sum_{\mathbf{\bar{T}}, \mathbf{T}} e^{-iK' \cdot (\mathbf{\bar{T}} + \mathbf{\bar{T}})} \langle \phi_{\beta}(\mathbf{\bar{T}}') | \mathcal{H} | \phi_a(\mathbf{\bar{T}}) \rangle e^{iK \cdot (\mathbf{\bar{T}} + \mathbf{\bar{T}})}
\]

Assuming that the inter-site tunnelling amplitude depends on the layer-projected difference coordinate, the matrix element can be expressed

\[
\langle \phi_{\beta}(\mathbf{\bar{T}}') | \mathcal{H} | \phi_a(\mathbf{\bar{T}}) \rangle = \frac{1}{(2\pi)^2} \int d^2 q \cdot f(q) e^{iq \cdot (\mathbf{\bar{T}} + \mathbf{\bar{T}} - \mathbf{\bar{T}})}
\]

Carrying out the lattice sums in equation (21) and expressing the momenta in terms of their differences from the respective zone corners, \( \mathbf{k} = \mathbf{K} + \mathbf{\bar{q}} \), one obtains an expression for the interlayer tunnelling amplitude in terms of sums over the range of structures they studied this mass scale appears to a rapidly decreasing function of the commensuration cell period. But the existence of this mass matrix in the low energy theory presents a significant challenge to the interpretation of the electronic states even at energies above the mass scale. Notably, in order to match smoothly to these low energy eigenstates the bilayer eigenstates at higher energy must be (near) equal weight states coherently mixed between the two layers instead of the layer-polarized eigenstates that one would infer from the momentum space structure of the continuum theory.

6
reciprocal lattices of the reference ($\vec{G}$) and rotated ($\vec{G}'$) layers

$$T_{\mu\nu}(\vec{q}', \vec{q}) = \frac{1}{A} \sum_{\vec{G}, \vec{G}'} f(\vec{q} + \vec{G} + \vec{G}') e^{i\vec{G}' \cdot \vec{r}_\nu} e^{-i\vec{G} \cdot \vec{r}_\mu}$$

$$\delta(\vec{q}' - \vec{q} + \Delta K + \vec{G}' - \vec{G})$$

(23)

where $A$ is the area of the unit cell.

When $q \ll G$ equation (23) describes two distinct types of interlayer tunnelling processes: (1) direct interlayer terms conserve the crystal momentum $k$ and occur when $\Delta K = (K(\theta) - \vec{K} + \vec{G}' - \vec{G}).$ (Note that this occurs for $\vec{G} = \vec{G}' = 0$ and for all boosts by the reciprocal lattice vectors $\vec{G} = \vec{G}' - \vec{G}$ that symmetrically shift the initial and final states to nearly aligned valleys of the two layers.) (2) Indirect interlayer terms conserve the Dirac momentum $\vec{q}$ and occur when $\vec{K} + \vec{G} = \vec{K}' + \vec{G}$. The matrix element for this latter process is dominated by the Fourier amplitude of the tunnelling potential $K(\theta)$ and provides a microscopic basis for the continuum formulation of Lopes dos Santos et al [17]. By contrast process (2) allows (indeed requires) $\vec{q} = \vec{q}'$ coupling and in particular it provides a mechanism for coupling between the tips of the Dirac cones in neighbouring layers. It can be understood as an interlayer umklapp process whereby the scattering by a reciprocal lattice vector of the commensuration cell provides precisely the right momentum to bridge the momentum offset $\Delta K$. The ratio of the amplitudes for the indirect and direct couplings is approximately $f((K + G))/f(|K|)$ so that the indirect term is generally weaker than the direct term.

5.2. Superlattice commensuration effects

As discussed in section 2.2 either $K \rightarrow K(\theta)$ or $K' \rightarrow K'(\theta)$ couplings are in the reciprocal lattice of the commensuration cell for a faulted bilayer, depending on the sublattice symmetry, and using equation (23) they are allowed interlayer tunnelling processes. The low energy theory is fundamentally changed by these terms since they introduce an interlayer mass operator in the long wavelength Hamiltonian. Interestingly, the analytic structure of this mass matrix is determined solely by the sublattice symmetry of the commensuration. Thus one can define two complementary families of commensurate faults where all members of a common family have a common form for their low energy Hamiltonians. The energy scale of this mass operator depends on the period of the commensuration, and it is largest for low order commensurate rotations. The primitive stacked structures with $AB$ and $AA$ stacking are parent structures for this family behavior which give the two prototypical examples of the possible interlayer mixing in a generic commensurate bilayers.

The commensuration physics for these systems can be understood most easily by explicitly writing the layer Bloch states in real space in the ‘first star’ approximation that retains only the three reciprocal lattice vectors that keep the combination $K + G$ to the first star of $K$ points

$$\Psi(\vec{r}) = \sum_a \Phi_\alpha(\vec{r}) n_\mu(\vec{r}); \Phi_\alpha(\vec{r}) = \frac{1}{\sqrt{3}} \sum_{m=1}^3 e^{iK_m(\vec{r} - \vec{r}_\mu)}.$$

(24)

The coupling between layers is a functional of the Bloch fields $\Psi(\vec{r})$. This can be modelled using a local functional

$$U = \frac{1}{2} \int d^2 r |T_\ell(\vec{r})| \Psi_1(\vec{r}) - \Psi_2(\vec{r})|^2$$

(25)

where $T_\ell$ is a real modulated supercell-periodic function arising from the lattice structure of the commensuration cell. The bilinear terms in this functional describe contributions to the total energy from single particle interlayer hopping processes that coherently couple the two layers. This coupling function represents a symmetry-allowed discretized $|\nabla \Psi|^2$ term in the Hamiltonian which acts to correlate the amplitudes and phases of the $\Psi$’s in the neighbouring layers. The purely local coupling between layers in equation (25) can be readily generalized to describe interlayer coupling with a finite range without substantially changing the physics. Equation (25) describes a coupled mode theory where the full Bloch waves $\Psi$ of the two layers (importantly these are not the Dirac envelope functions $u_\alpha(\vec{r})$) are coupled through by a local spatially modulated potential.

The coupling function $T_\ell$ describes the interlayer hopping potential. Although its exact form is unknown its important properties are constrained by symmetry: it is a real supercell-periodic function with local maximum near aligned sites of the two layers and with minima for regions where atoms in neighbouring layers are out of registry. These properties can be represented by an analytic model constructed from the primitive density waves in each of the layers

$$n_{\mu=1,2}(\vec{r}) = \sum_{m \in [1]} \sum_{a=\alpha,B} e^{i\vec{G}_{a,m}(\vec{r} - \vec{r}_{a,m})}$$

(26)

summed over the first star of reciprocal lattice vectors in the $\mu$th layer. Then, a nonlinear functional of the density fields that satisfies all the symmetry constraints is

$$T_\ell(\vec{r}) = C_0 e^{C_1(n_1(\vec{r}) + n_2(\vec{r}))}.$$ 

(27)

The argument of the exponential is a sum of primitive layer density waves: it is a real function with the translational symmetry of the commensuration cell and no shorter. Equation (27) is maximized at special positions where the two density functions in each layer are separately maximized which correspond to aligned atomic sites, and it exhibits exponential suppression in regions where the density waves are out of registry. This ansatz for the coupling function has three important features. (1) It is a nonlinear function of the primitive reciprocal lattice vectors of each of the layers so that all the reciprocal lattice vectors of the commensuration cell are represented in the expansion of the exponential in powers of its argument. (2) It is a separable function, constructed from a product of functions each of which is spanned by the individual reciprocal lattices of the two layers. (3) It is parametrized by two constants $C_0$ and $C_1$ which, respectively, describe the strength and range of the microscopic interlayer tunnelling amplitudes. (Thus for example, very long range hopping is described by a small value of $C_1$.) The values of the two constants $C_0$ and $C_1$ can be estimated from microscopic theory.
The left pattern has odd sublattice exchange parity, the right structure is even. The patterns contain combinations of fivefold resonance rings that are combined in clusters to form a periodic two-dimensional pattern. Adapted from [19].

Figure 4 gives a density plot of the local coupling function \( T_\ell(\vec{r}) \) calculated for partner commensurations at \( \theta = 21.787^\circ \) and \( \theta = 38.213^\circ \). The former corresponds to a sublattice exchange ‘odd’ structure and has a threefold rotational symmetry. Its partner is a sublattice exchange ‘even’ structure and retains the full sixfold symmetry of the graphene layer, though on an inflated commensuration supercell. This illustrates a general property of all ‘odd’ and ‘even’ commensurate faults. The patterns shown in this density plot provide a real space image of the interlayer resonance pattern for a twisted bilayer. Interestingly, for large angle faults, one finds that the appearance of fivefold resonance rings (due to rotated misaligned hexagons) is a robust motif in the coupling function. For small angle faults the coupling function is described instead by the familiar Moire pattern that evolves smoothly between zones locally defined by \( AB \), \( BA \) and \( AA \) stacking.

The couplings between the Dirac fields \( u_\alpha \) in neighbouring layers are obtained from the cross terms in equation (25) after integrating out the lattice scale oscillations and are given by the Fourier transform of \( T_\ell \) on the reciprocal lattice of the commensuration cell \( \Gamma(\vec{G}) \). The \( \vec{G} = 0 \) term describes the crystal momentum-conserving interlayer couplings discussed in the theory of Lopes dos Santos et al [17]. In addition there are umklapp terms involving \( \vec{G} \neq 0 \) terms that express the symmetry-allowed couplings between Dirac nodes \( K_m \rightarrow K_m(\theta) \). The geometrical considerations of section 2.2 require that for any given commensuration there are couplings within two distinct pairs of Dirac nodes at the corners of their respective Brillouin zones. In the Bloch basis these matrix elements are spanned by a \( 3 \times 3 \) matrix of finite momentum scattering amplitudes \( \tilde{V}_\text{ps} \) which, using the threefold rotational symmetry, takes the form

\[
\tilde{V}_\text{ps} = \begin{pmatrix} V_0 & V_1 & V_2 \\ V_2 & V_0 & V_1 \\ V_1 & V_2 & V_0 \end{pmatrix},
\]

(28)

Here \( V_0 \) describes the scattering amplitude for a momentum transfer \( \vec{G} = |\Delta \vec{K}| \) coupling the two layers while \( V_1 \) and \( V_2 \) describe scattering amplitudes with larger momentum transfers \( \sim \vec{G} \). By projecting equation (28) onto the sublattice (pseudospin) basis, one obtains the \( 2 \times 2 \) interlayer mass matrices \( \hat{H}_\text{int}^\pm \) that couple the Dirac fermions of the two layers. The low energy Hamiltonian for an even bilayer is thus expressed as a \( 4 \times 4 \) matrix operator (acting on the two sublattice and two layer degrees of freedom). The two lowest energy bands are obtained by retaining the \( q = 0 \) part of the interlayer matrix

\[
\hat{H}_\text{even} = \begin{pmatrix} -i\hbar \tilde{v}_F \sigma_1 \cdot \nabla & \hat{H}_\text{int}^+ \\ \hat{H}_\text{int}^- & -i\hbar \tilde{v}_F \sigma_2 \cdot \nabla \end{pmatrix}
\]

(29)

while for the odd bilayer

\[
\hat{H}_\text{odd} = \begin{pmatrix} -i\hbar \tilde{v}_F \sigma_1 \cdot \nabla & \hat{H}_\text{int}^- \\ \hat{H}_\text{int}^+ & i\hbar \tilde{v}_F \sigma_2 \cdot \nabla \end{pmatrix}
\]

(30)

where \( \sigma_n \) are Pauli matrices acting in the sublattice pseudospin basis of the \( n \)th layer and \( \tilde{v}_F \) is the renormalized Fermi velocity. Note that for even parity faults the bilayer Hamiltonian couples nodes of the same chirality, whereas the odd parity faults introduce coupling between nodes of compensating chirality. In either case the spectrum for coupled system retains a two-valley character due to the two ways of matching nodes in either family of structures. The interlayer mass matrices \( \hat{H}_\text{int}^\pm \) are

\[
\hat{H}_\text{int}^+ = \tilde{V} e^{i\varphi} \begin{pmatrix} \cos(\varphi/2) & 0 \\ 0 & e^{-i\varphi/2} \end{pmatrix}, \quad \hat{H}_\text{int}^- = \tilde{V} e^{i\varphi} \begin{pmatrix} 0 & 1 \\ 0 & 0 \end{pmatrix}.
\]

(31)

Equations (29) and (31) show that for sublattice ‘even’ faults the mass term involves an \( xy \) rotation of its pseudospin through angle \( \varphi \). This angle cannot be identified with the rotation angle \( \theta \) but it results instead from the interference of the three complex scattering amplitudes \( \tilde{V}_i \). By contrast in equations (30) and (31) one finds that interlayer motion across an ‘odd’ fault is mediated by the amplitude on its dominant eclipsed sublattice. In both mass matrices the overall phase of the operator \( \varphi \) can be removed by a gauge transformation. Note that equations (29)–(31) project the full Hamiltonian...
into the subspace spanned by just its lowest energy bands. This projection thus selects the Fourier component of the interlayer potential responsible for mixing the Dirac nodes and it correctly describes the low energy dispersion of the twisted bilayer. Other Fourier components of the coupling that are relevant for example to the backfolding of its multiple higher energy bands are not contained in this model. A complete treatment of the latter presents a complex though interesting problem due to the existence of two competing periodic potentials.

Equation (31) describes a coupling between Dirac waves in the neighbouring layers that persists in the long wavelength \( q \to 0 \) limit, qualitatively changing the structure of the low energy spectra. Their effects are illustrated in figure 5. For sublattice odd parity faults one pair of coupled bands is gapped on the interaction scale \( \mathcal{V} \) leaving an \( E = 0 \) contact point between a second pair of quadratically dispersing bands. For the even parity structures, shown on the right, feature a bonding/antibonding splitting, with a fully developed gap near the charge neutrality point which degenerates to a gapless state for the AA stacked structure. Adapted from [19].

5.3. Nonlocal potential scattering

For small rotation angle the offset momentum \( \Delta K \to 0 \) and perturbation theory in the dimensionless parameter \( I_1 / \hbar v_F \Delta K \) fail. The breakdown of the perturbation theory occurs because of an incomplete treatment of multiple scattering processes involving the interlayer coupling operator. Recognizing this, Bistritzer and MacDonald (BM) [25] developed a theory that treats multiple scattering through the three fundamental interlayer amplitudes that describe a spatially modulated interlayer coupling with the period of the commensuration supercell. Their results show that the Fermi velocity renormalization of the perturbation theory presages more dramatic physics at small rotation angle which can be described as ‘velocity reversal’, i.e. the Fermi velocity changes sign as a function of (small) rotation angle crossing through zero at a series of discrete magic angles. As a consequence the small angle regime is predicted to feature a manifold of nearly flat bands at low energy.

The BM model is formulated as a two layer scattering theory: states with momentum \( \mathbf{k} \) in one layer are scattered into states at momentum \( \mathbf{k} + \mathbf{Q} \) in its neighbour. In the pseudospin basis the amplitudes for these processes are the \( 2 \times 2 \) matrices given in equation (18). The gauge transformation \( e^{i \Delta \mathbf{k} \cdot \mathbf{r}} \) on the rotated layer brings two Dirac nodes of neighbouring layers into coincidence, and in this momentum shifted basis the three momentum transfers \( \mathbf{Q}_{\pm 0, \pm 1} \) are \( \mathbf{Q}_0 = -\Delta \mathbf{K} \) and two \( \pm 2\pi / 3 \)-rotated partners \( \mathbf{Q}_{\pm 1} \) which form a threefold symmetric triad. Thus this construction considers a twofold layer-degenerate Dirac cone whose states are coupled through an off-diagonal nonlocal operator containing three possible momentum transfers \( \mathbf{Q}_i \) in the interlayer hopping. In the long distance theory the single-layer Hamiltonians are isotropic, so for arbitrary rotation angle \( \theta \) the theory is specified by its unrenormalized Fermi velocity, the rotation angle and the coupling strength labelled \( \alpha \) in BM [25], which combine to form a single dimensionless scaling parameter \( \alpha = \omega / 2 \hbar v_F \sin(\theta / 2) \).

Repeated action of the nonlocal interlayer operator generates a lattice of coupled momenta as shown in the top
Figure 6. Top panel: a lattice of momenta is generated by repeated action of a nonlocal interlayer coupling operator on a Bloch state in a single layer. The nonlocal operator transports an electron between layers and boosts the momentum by a threefold symmetric triad of momentum transfers $Q_i$. An even number of applications of the operator generates a triangular lattice of momenta in the original layer (red), an odd number generates a triangular lattice offset by momentum transfers $Q_i$. The combination forms a honeycomb lattice of coupled momenta. Bottom panel: band structures obtained by numerical diagonalization of the continuum Hamiltonian in a truncated plane wave basis retaining kinetic energies of order the reduced velocity parameter $v_F$. The bands are plotted along the momentum space trajectory $ABCDA$ in the top figure. For small rotation angles the bands flatten and the Fermi velocity of the zero energy states is strongly suppressed. Adapted from [25].

Figure 7. The magnitude of the renormalized Fermi velocity for the zero energy states is plotted as a function of the coupling parameter $\alpha^2 = (\hbar v_F \sin(\theta/2))^2$ for small rotation angles. The oscillations result from sign changes of the Fermi velocity in the small rotation angle regime. For large rotation angles the renormalization factor $v^*/v \approx 1 - 9\alpha^2 \rightarrow 1$. Adapted from [25].

BM studied this model by numerically diagonalizing a truncated Hamiltonian expanded in a plane wave basis and retaining plane waves with kinetic energies below the coherence scale $\sim w$. The effects of multiple scattering through the interlayer coupling terms are then encoded in the structure of the bilayer eigenstates which are coherent superpositions of the single-layer Dirac modes. For large rotation angles ($\theta > 3^\circ$) the model reproduces the perturbation theory of Lopes dos Santos et al. By contrast, in the very small angle regime the bandwidth $\hbar v_F Q_0$ collapses, the number of elements in the low energy basis grows correspondingly and the electronic structure becomes spectrally congested as illustrated in figure 6. Thus the small angle regime is described by a strong coupled multiband theory that introduces physics inaccessible to a low order perturbation theory. BM find that the low energy spectra in this regime show a very substantial reduction in the Fermi velocity (typically $\lesssim 0.1$ of its single-layer value) due to level repulsion among the coupled bands. Remarkably, the reduced velocity parameter oscillates as a function of the fault angle as shown in figure 7 and crosses zero at a series of magic rotation angles. They suggest that this oscillation likely results from a $\theta$ dependence of the superpositions of single-layer modes that contain velocities of opposite sign, though a complete theory of the velocity oscillations has yet to be developed. Thus the velocity renormalization found in the weak coupling limit represents just the first step towards a complete twist-induced reconstruction of the low energy spectrum!

5.4. Two band models

There has been interest in distilling the continuum theory to a simpler effective two band model that captures the topological structure of its low energy spectrum. The approach is similar in spirit to the theory of Bernal stacked bilayers [3, 26, 29] where one can integrate out its high energy degree of freedom to arrive at an effective theory for its low energy states. For the Bernal bilayer this procedure identifies a new class of layer-coherent chiral fermions that have quadratic low energy dispersion and
a Berry’s phase of $2\pi$. For the twisted bilayer, neglecting commensuration effects, the low energy spectrum contains two layer-polarized linear Dirac cones that are recoupled at an energy scale $\hbar v_F|\Delta K|/2$ where they merge. The two band model attempts to provide a compact description of the topological transition of the band dispersion that connects the low energy ‘doubled cone’ sector to its high energy layer-hybridized sector.

For twisted bilayers the two band construction can be understood as a variant of the low energy theory for a Bernal bilayer that allows for a finite momentum offset $|\Delta K|$ between its Dirac nodes. Thus the low energy theory for Bernal stacking is modified in the manner

$$H_K = -\frac{\hbar^2}{m} \begin{pmatrix} 0 & \frac{\partial^2}{\partial \vec{q}^2} \frac{\Delta K}{2} \\ \frac{\partial^2}{\partial \vec{q}^2} \frac{\Delta K}{2} & 0 \end{pmatrix}$$

$$\rightarrow -\frac{\hbar^2}{m(\theta)} \left( \frac{\partial^2}{\partial \vec{q}^2} \frac{\Delta K}{2} - \left( \Delta K \right)^2 \right)$$

valid for very small fault angles where $\hbar v_F|\Delta K| \ll \vec{t}_\perp$ and $m = \vec{t}_\perp / v_F^2$. This expression can be introduced by replacing the interlayer operators of equation (18) by a simpler expression $\vec{t}_\perp \sigma_z$ which physically describes an interlayer tunnelling amplitude across a single sublattice in each layer. The spectrum of this Hamiltonian features a pair of Dirac cones, split by the momentum offset $|\Delta K|$, that merge at a two-dimensional saddle point at $q = 0$ representing the topological transition of the band structure. Importantly, the single-layer Dirac cones in this model have the same chirality so that annihilation of the Dirac points when they are coupled is topologically forbidden. Generically, this model does allow for an energy offset between the Dirac points of the coupled bilayer but this is believed to be small for physically reasonable coupling strengths.

The Hamiltonian in equation (32) has been used to study the orbital quantization of a twisted bilayer in the presence of a perpendicular magnetic field. By construction the limit $\Delta K = 0$ describes the Landau quantization of Bernal bilayer graphene: a Berry’s phase of $2\pi$ and quantized energy levels $\propto \sqrt{nB}$. By contrast for finite rotation angles the low energy spectrum of the offset model is a ‘doubled’ theory of single-layer graphene: the fourfold degeneracy due to the spin and valley degrees of freedom is doubled by an approximate layer decoupling of its low energy eigenstates. An asymptotic analysis of the eigenvalues within this model demonstrates that splittings of the Landau level degeneracies due to the interlayer coupling are exponentially suppressed as a function of the rotation angle in the low energy regime [28]. The spectrum thus features a zero mode and Landau levels that disperse $\propto \sqrt{nB}$ [27, 28]. This twofold layer degeneracy is quickly eliminated as one passes through the crossover energy $\hbar v_F|\Delta K|/2$ where the Dirac cones merge and hybridize. Above this crossover the spectrum has a different character: layer degeneracies are removed and the quantized energies are $\propto (n+1/2)B$ as expected for a massive interlayer coherent band.

6. Discussion

Rotational faults commonly occur in several different forms of graphene and their electronic properties are actively studied experimentally. The rapidly growing experimental literature on this subject has not yet provided a unified picture of the effects of faults on the electronic behaviour, possibly due to differences in the electronic properties of samples produced by different experimental methods.

A significant point of agreement among the various experimental works is that the interlayer coherence scale is small in these systems [9, 10, 14, 15]. This can be deduced clearly from their Landau level spectra which have been measured by scanning tunnelling spectroscopy (STS). These spectra show a scaling of the Landau level energies $E_n \propto \sqrt{nB}$ [14, 15] the signature of the Landau quantization of a massless Dirac band, as observed for single-layer graphene and quite distinct from the level sequence observed for Bernal stacked bilayers [3]. Perhaps the strongest evidence for a reduction of the interlayer coupling scale comes from angle resolved photoemission experiments which directly measure the quasiparticle dispersion relation [16, 30] and find spectra that follow the expected form for an isolated Dirac cone. These measurements have been interpreted as providing the first direct measurement of the Dirac dispersion relation in graphene, uncontaminated by substrate or other interlayer effects [16].

Since the effects of the interlayer coupling in twisted multilayers are intrinsically weak, their study is posing a significant experimental challenge. It is here where different experiments carried out on different samples disagree. For example, the Fermi velocity can be deduced from the slope of the $\sqrt{nB}$ scaling relation for the Landau quantized energies. The strongest evidence for a twist-induced renormalization of $v_F$ comes from the Landau level spectra measured by scanning tunnelling spectroscopy of CVD graphenes grown on Ni substrates [20]. This work reports that $v_F$ is not constant as a function of scanned position across a macroscopic sample, but instead it is found to vary in a range $0.87 \times 10^6$ m s$^{-1} < v_F < 1.1 \times 10^6$ m s$^{-1}$. Simultaneous measurement of the topography of the Moire superlattice period of these samples correlates the velocity reduction with the period and hence the rotation angle. The larger value, found for large angle rotations, agrees well with the $v_F$ for single-layer graphene and the 20% reduced value is correlated with a small angle rotation $\sim 3^\circ$ as suggested by a perturbative analysis of the continuum theory [17, 25]. This contrasts with analogous STS measurements carried out for multilayer graphenes grown epitaxially on SiC (0 0 0 1). These also show the $\sqrt{nB}$ scaling of the Landau quantized energies. However, for these materials the slope of the scaling relation yields a Fermi velocity $1.1 \times 10^6$ m s$^{-1}$ for all samples studied down to a rotation angle of $1.4^\circ$ [15] completely spanning the range of rotation angles where a velocity renormalization is expected.

A similar discrepancy arises in the spectroscopy of the van Hove singularity presumed to occur in the region where the momentum-offset Dirac cones of a twisted bilayer merge. Low energy STS on Ni/CVD grown graphene reveals low energy peaks whose energies disperse with their topographically measured rotation angles in the low angle regime $1.2 < \theta < 3.5^\circ$ [18] roughly consistent with the van Hove scenario. Yet these features are not seen at all in spectroscopy of
the SiC epitaxial twisted graphenes regardless of the fault angle. Perhaps the strongest challenge to the idea of a twist-induced spectral reconstruction comes from angle resolved photoemission. These measurements directly measure the quasiparticle dispersion and clearly resolve the Dirac cone with a Fermi velocity that is indistinguishable from that of single-layer graphene. Despite a careful search, no evidence is found in these measurements for any hybridization between Dirac cones in the spectral regions where they cross [30]. The simplest interpretation of the ARPES data is that the first few graphene layers accessible to this spectroscopy are electronically floating, i.e. extremely weakly coupled to each other and to deeper layers in the film.

An important goal for theory in this area is therefore to identify situations where the effects of the interlayer coupling across a rotational fault are manifest in their electronic behavior. There has been progress in this direction. Bistritzer and MacDonald have studied the effect of the rotation angle of a bilayer on its interlayer tunnelling conductance, predicting dramatic enhancements of the vertical conductance at special rotation angles that can be identified with low order commensurate superlattices [31]. More recent work has pointed to nontrivial effects of a $\theta$-dependent interlayer coupling on the equilibrium charge redistribution across the bilayer in a perpendicular electric field [32]. Kindermann and I studied the Landau level spectra for weakly coupled bilayers in a perpendicular magnetic field and find that even a weak coherence splitting of bilayer bands at energies well above the mass scale produces a striking new effect, the Dirac comb [33]. Here small differences in the orbitally quantized states in two slightly coherence-split bands produce an amplitude modulation of the Landau level spectrum with a period that significantly exceeds the coherence scale, and should be observable by magnetotransport in the weak field regime. Small rotation angles can introduce long spatial Moire periods for twisted bilayers that can be made commensurate with the magnetic length $\sqrt{\hbar/eB}$ on accessible field scales, accessing Hofstadter commensuration physics in a new family of materials [34]. The band flattening theoretically predicted for in the small twist angle regime will surely focus attention on many body effects in the low energy physics. Further studies along all of these lines provide a very open area for further work.

One might be discouraged by the lack of a definitive theory of the electronic structure of rotationally faulted graphenes. In contrast this is an exciting situation. These systems are challenging to the most familiar tools of electronic structure theory and their understanding is likely to involve creative new approaches.

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