Topological Bloch bands in graphene superlattices

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We outline a designer approach to endow widely available plain materials with topological properties by stacking them atop other nontopological materials. The approach is illustrated with a model system comprising graphene stacked atop hexagonal boron nitride. In this case, the Berry curvature of the electron Bloch bands is highly sensitive to the stacking configuration. As a result, electron topology can be controlled by crystal axes alignment, granting a practical route to designer topological materials. Berry curvature manifests itself in transport via the valley Hall effect and long-range chargeless valley currents. The nonlocal electrical response mediated by such currents provides diagnostics for band topology.

topological bands | graphene | van der Waals heterostructure

Electronic states in topological materials possess unique properties including a Hall effect without an applied magnetic field (1–3) and topologically protected edge states (4, 5). Accessing nontrivial electronic topology depends on identifying materials in which symmetry and interactions produce topological Bloch bands. Such bands can only arise when multiple requirements, such as a multiband structure with a Berry phase and suitable symmetry, are fulfilled. As a result, topological bands are found in only a handful of exotic materials in which good transport properties are often lacking. Formulating practical methods for transforming widely available materials with a reasonably high carrier mobility (such as silicon or graphene) into a topological phase remains a grand challenge.

Here, we lay out an approach for engineering designer topological materials out of stacks of generic materials—“Chernburgers.” Our scheme naturally produces (i) topological bands with different Chern invariant values, and (ii) tunable topological transitions. As an illustration, we analyze graphene on hexagonal boron–nitride heterostructures (G/hBN), where broken inversion symmetry is expected to generate Berry curvature (6, 7), a key ingredient of topological materials. Indeed, recently valley currents have been demonstrated in a G/hBN system (8) signaling the presence of Berry curvature (6). As we will show, Berry curvature in G/hBN can be modulated by stacking configuration, leading to a large variability in properties. Transitions between different topological states can be induced by a slight change in stacking angle.

Topological bands in G/hBN arise separately for valley K and valley K′. Graphene bandstructure reconstruction due to the coupling to hBN produces superlattice minibands (9–14), with Berry curvature Ω(k) developing near avoided crossings. The minibands for each valley possess a valley Chern number

\[ C_v = \frac{1}{2\pi} \int d^2k \Omega(k), \]

where the integral is taken over the entire superlattice Brillouin zone (SBZ) in one valley (K or K′). As discussed below, for commensurate stackings (Fig. 1A) \( C_v = \pm 1 \) for the lowest minibands. In contrast, for incommensurate moiré superlattice structures (Fig. 1B), the invariant \( C_v \) vanishes in these minibands, \( C_v = 0 \). The difference in the behavior for these configurations arises from the difference in sign of the contributions to Berry curvature from regions near SBZ center Γ (the Dirac point, hereafter denoted DP) and corners K, K′ (Fig. 2). We will see that these contributions add in the commensurate case but subtract for the incommensurate case, yielding topological and nontopological bands, respectively (Fig. 1).

Interestingly, the conditions for both topological and nontopological bands are met by currently available systems. Indeed, both commensurate and incommensurate stackings have been recently identified in G/hBN by scanning probe microscopy (15, 16). Further, the commensurate–incommensurate transition can be controlled by twist angle between G and hBN, providing a practical route in which to tailor electron topology via a tunable structural transition.

We note that time-reversal (TR) symmetry requires that \( \Omega(k) \) in K and K′ valleys have opposite signs. As a result, the total Chern invariant always vanishes, \( C_v(K) + C_v(K′) = 0 \). However, the weakness of intervalley scattering (17, 18) can enable long-range topological currents in individual valleys. As we will see, the nonlocal electrical signals mediated by such currents can provide diagnostics for valley band topology.

We also note that topological bands in graphene are sometimes presumed either impossible or impractical. Indeed, a connection between K and K′ bands at high energies, whenever present, renders valley-specific topological invariants ill-defined (19, 20). Proposals relying on large spin–orbit coupling (21, 22) are also sensitive to disorder; proposals in other systems such as optical flux lattices (23) suffer from similar implementation pitfalls. Our scheme circumvents these difficulties by exploiting Bragg scattering in the G/hBN superlattice to create energy gaps above and below the K and K′ Dirac points (Fig. 2). The Dirac points, sandwiched between these gaps, are no longer connected in a single band; the resulting minibands possess well-defined topological invariants.

Minimal Model for Superlattice Bands

Modeling the superlattice bandstructure is greatly facilitated by several aspects of the G/hBN system. First is the long-wavelength character of superlattice periodicity, which results from nearly identical periods of graphene and hBN crystal structure. For

Significance

A family of designer topological materials is introduced, comprising stacks of two-dimensional materials which by themselves are not topological, such as graphene. Previously, topological bands in graphene were presumed either impossible or impractical. The designer approach turns graphene into a robust platform with which a host of topological behavior can be realized and explored.
Fig. 1. Topology of Bloch bands for different stacking types of G/hBN, commensurate (A) and incommensurate (B). (Top) Hexagonal commensurate domains (black lines mark domain walls) and incommensurate moiré superlattice structure. (Bottom) Valley Chern number $C_V = \pm 1$ and $C_V = 0$ for the lowest reconstructed minibands labeled "**" in Fig. 2. This corresponds to the contributions to the net Berry flux, $\mathcal{F} = \mathcal{F}_{DP} + \mathcal{F}_{KK'}$, from the superlattice Brillouin zone center $\Gamma$ and corners $K, K'$ that have equal signs and opposite signs, respectively (see Eqs. 12 and 13). Red and blue shaded regions indicate the "+" and "−" signs of Berry curvature.

Fig. 2. Graphene superlattice potential transforms the massless Dirac bandstructure near points $K, K'$ of graphene Brillouin zone (A) into a family of minibands (B). Shown is the bandstructure near points $K, K'$ obtained from the Hamiltonian in Eq. 2 (parameters used: $m_3 = 60 \text{ meV}$, $e_0 = \hbar v_b = 1 \text{ eV}$, $\Delta_g = 20 \text{ meV}$). Large superlattice period translates into a small size of superlattice Brillouin zones (two hexagons positioned at points $K, K'$). Sublattice A/B dependent coupling ($\sigma_3$ term in Eq. 2) generates a Dirac mass term and opens a gap between the conduction and valence bands; it also creates avoided band crossings 1–2 and 3 above and below Dirac points.
To illustrate this important point, we present the argument in a form that does not depend on detailed knowledge of the registration within each of the domains. Of course, in practice the registration types (and hence the asymmetry signs) arise from general energetic and geometric constraints which can be easily accounted for (27). As an example, we consider three possible registrations: (i) site A in hBN aligned with site A in graphene and site B in hBN with site B in graphene; (ii) site A in hBN aligned with site B in graphene and site B in hBN with H (below) in graphene; and (iii) A in hBN aligned with site B in graphene whereas site B in hBN aligned with site A in graphene.

Configurations (i) and (iii) cost the same energy, but have a different energy than (ii). Importantly, lateral sliding of a cell with configuration (i) cannot generate configuration (iii) because it would require a lattice rotation. At the same time, whereas lateral sliding of a cell with configuration (i) can generate configuration (ii), it costs a different energy. As a result, stacking frustration between neighboring cells cannot occur, locking the registration between all hexagonal cells to yield a constant global gap, $\Delta_g$.

Next, we note that imperfect registration around the domain boundaries yields a weaker coupling between G and hBN [strained graphene sheet buckles (16) increasing the G-to-hBN distance]. Reduction in sublattice-asymmetric potential $\Delta_{g,0}$ can be modeled as

$$\Delta(r) = \Delta_{g,0} + \delta m \vec{F}(q) \cdot \vec{R}(r), \quad \text{sgn} \ \delta m = -\text{sgn} \ \Delta_{g,0},$$

where $\vec{F}(q)$ describes the unit cell of the pattern of domain walls, $\vec{R}(r)$ is the superlattice form factor, $\delta m$ is the superlattice basis vector, and $\vec{F}(q)$ indicates convolution. The relative sign $\delta m = -\text{sgn} \ \Delta_{g}$ accounts for the weaker coupling between G and hBN at the domain boundaries.

Because we are interested in bandstructure reconstruction in the lowest minibands, we expand $\Delta(r)$ into lowest harmonics yielding Eq. 2 with

$$\Delta_{g} = \Delta_{g,0} + \delta m \vec{F}_{q=0}, \quad m_{3} = 2\delta m \vec{F}_{q=b},$$

where $\vec{F}(q) = 1/A \int d^2r \vec{F}(r) \cdot e^{i \cdot r}$ is the form factor, $\vec{b}$ is the reciprocal superlattice vectors, and $A$ is the area of superlattice unit cell. Crucially, the sign of the form factor $\vec{F}$ determines the sign of $m_{3}$. Choosing a symmetric $\vec{F}(r)$ with origin at the center of a hexagonal domain (pictured in Fig. 1A, x and y are the horizontal and vertical directions) and $\delta$-functions along the hexagonal domain walls, we obtain the form factor

$$\vec{F}(q) = \frac{2w}{A} \sum_{j=0}^{3} \sin \left( \frac{d}{2} \frac{q \cdot x}{q \cdot y} \right) \cos \left( \frac{\sqrt{3}d}{2} \frac{q \cdot y}{q \cdot y} \right).$$

Here $d$ and $w$ are the domain wall length and width, and $q = R(\theta) \cdot q$, where $R(\theta)$ are the $2 \times 2$ rotation matrices with $\theta = 0$, $\theta = \pi/3$, and $\theta = 2\pi/3$. Evaluating Eq. 5 gives $\vec{F}_{q=0} = 3\zeta > 0$ and $\vec{F}_{q=b} = -9\sqrt{3}\zeta/4\pi < 0$, where $\zeta = wd/A$. Comparing with Eq. 2, we find the relation between signs of $m_{3}$ and $\Delta_{g}$:

$$\text{sgn} \ m_{3} = -\text{sgn} \ (\delta m) = \text{sgn} \ (\Delta_{g}).$$

As we will see, this leads to a nontrivial topological class $C_{g} = \pm 1$ in the lowest minibands (Fig. 3A).

The incommensurate case (moiré superlattice) differs from the commensurate case in two important ways. One is that the G-to-hBN coupling is dominated by the modulational part $\Delta(r) = m_{3} \sum_{j=0}^{3} \cos \left( \vec{b}_{j} \cdot r \right)$ arising from the moiré pattern. The other is that the global gap parameter $\Delta_{g}$ is zero in the bare Hamiltonian; however, a nonzero $\Delta_{g}$ value is generated perturbatively in $m_{3}$, with the $\Delta_{g}$ sign the opposite of the $m_{3}$ sign. The analysis is particularly simple for the long-period moiré patterns arising for rigid G and hBN stackings at small twist angles, as shown in Fig. 1B.

Of course, one $m_{3}$ harmonic cannot produce an average global gap at DP because it is sign-changing, $\langle |\Delta_{g}| \rangle = 0$. However, a combination of three different harmonics can open up a gap (14). This can be seen from a perturbation analysis of the Hamiltonian (2) which we write as $H = H_{0} + V$, where $H_{0} = \sum_{i,j} \vec{V} \cdot \vec{b}_{i} \cdot \vec{b}_{j}$, $V = \sigma_{3} \sum_{\rho} \cos \left( \vec{b}_{i,\rho} \cdot \vec{r} \right)$. Perturbation theory in $V$ yields a term describing a global gap at a third order in $V$ via

$$\delta H = V \left( \frac{1}{\epsilon - H_{0}} \right) V \left( \frac{1}{\epsilon - H_{0}} \right) V.$$ 

Choosing triplets of harmonics with $b_{i} + b_{j} + b_{k} = 0$, third-order perturbation theory in $m_{3}$ yields a gap

$$\Delta_{g} = \sum_{\rho} \frac{m_{3} \sigma_{3}}{2} \frac{1}{\sigma_{B} \cdot b_{i} \cdot b_{j}} \frac{m_{3} \sigma_{3}}{2} \frac{1}{\sigma_{B} \cdot b_{k} \cdot m_{3}} = \frac{3m_{3}^{2}}{4(\sigma_{B})^{2}},$$

where the minus sign results from the anticommutation relations $[\sigma_{1}, \sigma_{3}] = 0, [\sigma_{2}, \sigma_{3}] = 0$. Importantly, this analysis predicts a relation between signs

$$\text{sgn} \ (\Delta_{g}) = -\text{sgn} \ (m_{3}),$$

which is opposite to the relation found for the commensurate case, Eq. 6. Whereas the gap size obtained at a third order of perturbation theory in a noninteracting system is small, electron interaction effects are expected to produce an enhancement and
generate a large $\Delta_g$ (14). As we will see, the signs in Eq. 9 lead to trivial topological classes for superlattice bands, $C_v = 0$ (Fig. 3B).

In addition to the difference in signs, the commensurate and incommensurate stackings differ in the relative magnitude of the $\Delta_g$ and $m_3$ couplings. As we argued above, the global gap coupling $\Delta_g$ dominates in the commensurate case, with a relatively weaker modulational part $m_3$ arising due to registration unzipping along domain boundaries. In contrast, the modulational coupling $m_3$ is dominant in the incommensurate case, with the global gap $\Delta_g$ arising at third-order perturbation in $m_3$. The two distinct microscopic pictures result in a disparity between the $\Delta_g$ and $m_3$ scales and a sign difference, ultimately leading to different topological classes.

**Topological Classes**

We proceed to explore how stacking types impact the band topology. The topological properties of G/hBN can be analyzed through the Berry curvature in the minibands. Even though the G/hBN Hamiltonian, Eq. 2, possesses TR symmetry, its broken inversion symmetry allows for a finite Berry curvature to develop in the SBZ:

$$\Omega_n(k) = \nabla_{\kappa} \times A_n(k), \quad A_n(k) = i \langle \psi_n(k) | \nabla_{\kappa} | \psi_n(k) \rangle.$$  \[10\]

Here $n$ is the band index, $A$ is the Berry connection, and $|\psi_n(k)\rangle$ are the eigenvectors of Eq. 2. In what follows, we concentrate on a single valley and the lowest conduction miniband (labeled “1” in Fig. 2B).

Using Eq. 10, we evaluate $\Omega_n(k)$ for the bandstructure generated by Eq. 2, and obtain Berry curvature maps in SBZ reciprocal space which are shown in Fig. 3A and B. We adopted a numerical method similar to that outlined in ref. 28; see the Supporting Information for a full description. In Fig. 3A and B we plot $\Omega(k)$ corresponding to the lowest conduction band (labeled “1” in Fig. 2B); the lowest valence band exhibits the same behavior but with opposite sign. We find that $\Omega(k)$ is concentrated in the reciprocal space regions where the bandstructure exhibits gaps and avoided crossings, namely at the Dirac points and SBZ corners [Γ, K, and K’ respectively].

Integrating $\Omega(k)$ over the superlattice Brillouin zone to obtain the valley Chern number, Eq. 1, we identify two distinct cases. For the equal-sign case, Eq. 6, which corresponds to commensurate stackings, we obtain $C_v = -1$ (Fig. 3A). For the opposite-sign case, Eq. 9, which corresponds to incommensurate stackings, we obtain $C_v = 0$ (Fig. 3B). This gives topological and non-topological bands, respectively.

To gain more insight into band topology in Fig. 3, it is instructive to analyze the hot spots of $\Omega(k)$ individually. Near SBZ center Γ, the bandstructure is approximated by a constant-mass Dirac Hamiltonian $H = v_F \cdot p + \Delta_g \sigma_z$, where $\Delta_g = \Delta_g \sigma_z$ (due to TR symmetry). Berry curvature is then given by the well-known expression

$$\Omega_{\pm,K/K'}(k) = \mp \frac{\Delta_g^2 \eta_z}{2 (v_F^2 p^2 + \Delta_g^2)^{3/2}},$$  \[11\]

where $\pm$ refers to the conduction and valence bands and $\eta_z = +$ and $\eta_z = -$ for valley K and K’, respectively. This translates into the net Berry curvature flux, which is controlled by the sign of $\Delta_g$:

$$F_{DP} = \int d^2k \Omega_{\pm,K/K'}(k) = \mp \pi \eta_z \eta_n \Delta_g,$$  \[12\]

giving $\mp \pi$ for K, K’ valleys, as expected for a Dirac point.

Berry curvature also features hot spots at SBZ corners K and K’. These arise from Bragg scattering by the superlattice harmonics in Eq. 2 which mix the pseudospin textures; the energy spectrum and $\Omega(k)$ close to K, K’ can be modeled using the $k \cdot p$ method; see the Supporting Information. We find that the net Berry flux in the conduction band, $F_{K,K'} = \int d^2k \Omega(k)$ about the corners of the SBZ is controlled by $m_3$,

$$F_{K,K'} = \frac{\pi}{2} \eta_n \text{sgn}(m_3),$$  \[13\]

and are equal for both K and K’. When $m_3$ becomes small the hot spots around K, K’ contract, however the net flux $\pm \pi/2$ for each hot spot remains unchanged.

We note that the “half-Dirac” flux $\pm \pi/2$ follows from Chern number quantization. Integer $C_v = 1/2\pi \int_{SBZ} d^2k \Omega(k)$ arises from summing the Berry curvature concentrated about DP and K, K’ points in the SBZ (as shown in Fig. 3A and B). Because there are two inequivalent K points in the SBZ, $C_v = 1/2\pi (F_{DP} + 2F_{K,K'})$. Integer $C_v$ values and $F_{DP} = \pm \pi/2$ yield $\pm \pi/2$ values for $F_{K,K'}$.

**Valley Currents and Berry Curvature Spectroscopy**

Topological currents associated with each of the valleys can propagate over extended distances as long as the intervalley scattering is weak (17). Whereas TR symmetry requires no net charge Hall currents, the opposite signs of $\Omega(k)$ in K and K’ allow transverse valley currents, $J_z = J_k = J_{K'}$, to be induced by a longitudinal electric field, $E$. This valley Hall effect (VHE) is described by (6)

$$J_z = \sigma_{xy} E \times \mathbf{n}, \quad \sigma_{xy} = \frac{e \eta}{h} \int d^2k \Omega(k) f(k),$$  \[14\]

where n points perpendicular to G/hBN, $N = 4$ is valley/spin degeneracy, and $f(k) = \left( e^{i(k_{k,x} - \pi)} + 1 \right)^{-1}$. The difference between topological bands and non-topological bands is reflected in the behavior of $\sigma_{xy}$ which changes signs as a function of density varying in a single band, or maintains a constant sign, as illustrated in Fig. 3C and D. We note that sign-changing $\sigma_{xy}$ does not contradict $C_v = \pm 1$ for topological bands.
because $C_C$ tracks the total change in $\sigma_{xy}^C$ as density is swept through the band. Accounting for a total change of $\sigma_{xy}^C$ (quantized for topological bands, vanishing for nontopological bands), and using $\sigma_{xy}^C$ exactly at $\Gamma$ with magnitude $Ne^2/2h$ obtained from counting net Berry flux (19, 20, 29), we obtain the contrasting $\sigma_{xy}^C$ shown in Fig. 3 C and D.

Even though the currents $J_i$ are chargeless, they can be detected by electrical means (8). Indeed, the long propagation lengths enabled by weak intervalley scattering allow valley currents flowing in system bulk to mediate nonlocal electrical response. This is distinct from graphene edge modes that are highly susceptible to localization and gapping out on rough or imperfect edges. In contrast, recent measurements of intervalley scattering in G/hBN yield mean-free paths as large as several micrometers (17, 18). Nonlocal resistance measurements (Fig. 4) can therefore provide an all-electrical and robust way to probe the bulk valley-Hall conductivity.

Nonlocal resistance $R_{NL}$ arises in a way illustrated in Fig. 4A. Transverse valley currents $J_i$ induced by an electrical current $I$ can propagate over extended distances to induce a valley imbalance profile across the device, $\delta \rho = \delta \rho_y - \delta \rho_K$. Even far away from the current source, valley imbalance $\delta \rho$ can set up an appreciable transverse electric field via the reverse VHE:

$$E = \frac{\sigma_{xy}^C(n_2)}{\sigma_{xy}^C(n_1)} (\nabla \delta \rho) \times \mathbf{n}. \quad [15]$$

This provides a mechanism through which the chargeless long-range valley currents are converted to an electric signal at the readout contacts, producing a nonlocal transresistance $R_{NL}$. Such $R_{NL}$ was recently observed in ref. 8 for a uniform density device $n_1 = n_2$. Importantly, control over local density in the geometry of Fig. 4A, $R_{NL} = \sqrt{|I|/\delta \rho}$, is sensitive to the density and signs of $\sigma_{xy}^C$ in the two regions. Here the prefactor $A$ is positive and depends on the longitudinal conductivity $\sigma_{xx}$ of both regions, device dimensions, and intervalley scattering length, similar to that analyzed for the spin-Hall effect (30). For illustration, we set $A = (h/e^2)^2$.

Because $\sigma_{xy}^C$ for topological bands (Fig. 3C) changes sign as density is swept in a single band, we find that $R_{NL}$ displays multiple sign changes as a function of density in $n_1, n_2$ as shown in Fig. 4B. The sign-changing behavior of $R_{NL}$ can be traced back to the finite value of $C_C \approx \pm 1$ for topological bands and a $\sigma_{xy}^C$ of $Ne^2/2h$ at neutrality $n_{1,2} = 0$ (i.e., at DP) (19, 20, 30). In contrast, $R_{NL}$ maintains a constant sign for nontopological bands, $C_C = 0$, as shown in Fig. 4C. As a result, sign changes in $R_{NL}$ provide a clear diagnostic for topological bands.

In summary, graphene superlattices provide a practical route to constructing topological bands out of generic materials, as illustrated via tunable electron band topology in commensurate–incommensurate bandings. Band topology can be inferred from nonlocal transport measurements. In addition, interesting behavior is expected in superlattice systems in which large-scale inhomogeneities give rise to topological and nontopological domains. In such systems, domain boundaries are expected to support topologically protected chiral edge states. Transitions between topological and nontopological states can be induced by temperature and strain. A number of different systems can be used, including SiC where superlattice stackings have been observed (31, 32), G/BN (8, 16, 26), and twisted bilayer graphene (33–35). The ease with which stacked G/BN structures can be made (36) and the robust bulk transport signatures of their topological character open the door to accessing and probing electronic band topology in designer topological materials.

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