Topological Valley Currents in Graphene with Local Sublattice Asymmetry

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A broken sublattice symmetry opens a band gap in graphene. Due to Berry curvature effects, it has also been predicted to induce equal and opposite anomalous velocities for electrons in the K and K′ valleys. The resulting valley Hall effect (VHE), driven by currents in the Fermi sea, has been used to interpret experimental non-local resistance (RNL) measurements. However, this is not supported by quantum transport simulations, where such features are suppressed by the band gap opening. Here, we demonstrate that electron scattering from regions with broken sublattice symmetry in otherwise pristine graphene sheets, also leads to a splitting of electrons according to their valley index. This effect is robust against fluctuations in the size and composition of the regions. Furthermore, no global band gap is required and a finite Fermi surface contribution to the valley Hall effect is observed. Our findings suggest both an alternative bulk mechanism to induce VHE in graphene and a route towards valley-dependent electron optics devices.

Valleytronics [1] exploits the relative occupation of inequivalent band extrema to encode, transport and process information. Two-dimensional materials, and graphene in particular, are promising valleytronic candidates due to the K and K′ valleys at the Dirac points. A key obstacle is the absence of external controls, analogous to magnetic fields and ferromagnetic contacts in spintronics, to manipulate and detect valley-polarized currents [2]. While circularly-polarized light allows optoelectronic access to the valley degree of freedom in certain materials [3–6], an all-electronic control is highly desirable for device applications [7]. Towards this end, particular geometries have been proposed to filter or split electrons according to their valley index. Many of these are based on atomically-precise edges or defects [8–14], strain-induced pseudomagnetic fields which act oppositely in the K and K′ valleys [15–30]. However, experimental approaches are hampered by the relatively small parameter-space regions that give significant valley effects. The most promising signatures of valley-dependent phenomena have instead emerged from RNL measurements in commensurately stacked graphene/hexagonal boron nitride (hBN) systems [31–33]. Large RNL signals have been interpreted in terms of a VHE driven by a bulk Berry curvature [34–36]. This in turn emerges from a sublattice-asymmetric potential, or mass term, which acts differently of the A and B sublattices. Similar behaviour has also been observed in bilayer graphene [37, 38], where the VHE is related to the application of an interlayer bias.

The interpretation of experimental RNL in terms of a bulk-driven VHE has been questioned by quantum transport simulations [2, 39, 40]. These studies found that bulk-driven RNL signals in uniformly-gapped systems are suppressed beyond evanescent contributions, rendering them negligible at experimental scales. Experimental mapping of current flow further suggests that edge currents may play a role [41], but recent theoretical studies question the topological origin of such currents [40, 42, 43]. Finally, the lattice mismatch between graphene and hBN, which leads to a Moiré pattern for commensurate structures [31, 44], also suggests that electrons may not experience a uniform mass term [45], making the interpretation of current experiments in terms of VHE a true conundrum.

In this Letter, we demonstrate the emergence of a valley-splitting bulk transport mechanism in the absence of a global band gap. This phenomenon requires instead the presence of local spatial regions of finite mass in otherwise pristine graphene – a situation analogous to graphene-hBN heterostructures with commensurate layer alignment. We first derive the exact solution to the scattering problem for a circular dot, which establishes a strong valley dependence in the scattered wavefunction at low energies. This leads to a valley-splitting of incoming electrons and pure valley current flow around the dot. Tight-binding simulations confirm that this behaviour is robust for a wide range of dot profiles and mass distributions, in contrast to other mechanisms which require precise edges or strain profiles. The valley Hall conductivity for a periodic array of dots is then calculated using the Kubo-Bastin formalism, and confirms the formation of valley Hall signals and neutral charge currents. Unlike the uniform mass distributions considered elsewhere, this system shows both Fermi sea and Fermi surface contributions to the valley Hall conductivity at the charge neutrality point (where the Fermi surface contribution is expected to be zero), and is not dependent on a global
where we define scaled variables \( \tilde{\varphi} \). To solve the scattering problem analytically, we use the function [56] and Kubo-Bastin [57, 58] approaches, but used for atomistic simulations using the patched Green’s function patterned hBN or other substrates [44–47], or doping [48–55] to induce spatially-varying mass in graphene devices.

The electronic structure of graphene is described using a nearest-neighbour tight-binding (TB) Hamiltonian

\[
H = \sum_{\langle i,j \rangle} t (\hat{a}_{i}^{\dagger} \hat{b}_{j} + c.c.) + \sum_{i} V_{A}(r_{i}) \hat{a}_{i}^{\dagger} \hat{a}_{i} + \sum_{j} V_{B}(r_{j}) \hat{b}_{j}^{\dagger} \hat{b}_{j}
\]

where \( \hat{a}_{i}^{\dagger}, \hat{b}_{j}^{\dagger} \) are creation states and annihilation operators and \( V_{A}(r), V_{B}(r) \) represent spatially-dependent potentials on the A, B sublattices and \( t \approx -2.7 \text{eV} \) is the hopping integral. We consider a scatterer of radius \( R \), with mass term \( \Delta \), such that \( V_{A/B}(r < R) = \pm \Delta \) where the \( +/− \) sign choice corresponds to the A (B) sublattice. Such a term leads to band-gap opening in the range \( \frac{\Delta}{2} < E < \frac{\Delta}{2} \) where it is applied. This Hamiltonian is used for atomistic simulations using the patched Green’s function [56] and Kubo-Bastin [57, 58] approaches, but to solve the scattering problem analytically, we use the Dirac equation near the \( K \) and \( K' \) \((\tau = ±1)\) points

\[
H_{\tau}(r) = \hbar v_{F} \begin{pmatrix}
\tilde{V}_{A}(r) & -i\tau \partial_{x} - \partial_{y} \\
\partial_{x} + \partial_{y} & \tilde{V}_{B}(r)
\end{pmatrix}
\]

where we define scaled variables \( \tilde{X} \equiv \frac{X}{\hbar v_{F}} \).

Following Refs. [59–61], we consider an incoming electron plane wave along the \( x \)-axis \( (\psi_{\text{inc}}) \), the wave scattered from the dot \( (\psi_{\text{sc}}) \) and the wave transmitted into the dot \( (\psi_{\text{tr}}) \), as in Fig. 1. \( \psi_{\text{inc}} \) and \( \psi_{\text{tr}} \) are each expanded in terms of angular momentum basis states \( m \) using Bessel and Hankel functions respectively, identically to the potential dot case [60, 61]. The wavefunction inside the dot can similarly be expanded in terms of Bessel functions, but the mass term introduces energy and sublattice-dependent coefficients [62]. A closed form expression for the wavefunction is found by enforcing continuity at the dot boundary and solving for the scattering and transmission coefficients, \( c_{n} \) and \( c'_{n} \), for each mode. The Hamiltonian in Eq. (2) is invariant under \( V_{A} \leftrightarrow V_{B}, \psi_{\text{inc}}^{x} \leftrightarrow \psi_{\text{inc}}^{y}, \psi_{\text{sc}}^{x} \leftrightarrow -\psi_{\text{sc}}^{y} \), so that the \( K \) valley result can be used to calculate the \( K' \) case. The \( m \) and \( -m - 1 \) modes are closely connected and rely on the same Hankel/Bessel functions but with opposite signs [62]. These terms contribute similarly, but with different valley, sublattice and/or band indices, so we define \( m = -m - 1 \), and note that \( c_{m}(K') = c_{m}(K) \).

The local electron density \( n = \psi^{\dagger} \psi \) and particle current \( j = \psi^{\dagger} \sigma \psi \) are then calculated for each valley separately, with the total electronic (valley) quantity given by the sum (difference) of the individual valleys. We define the local valley polarisation of scattered current \( \xi(r, \phi) = j_{\text{val}}^{\pm}(r, \phi)/j_{\text{tot}}^{\pm}(r, \phi) \), which takes values \(-1, 0, 1\) for fully \( K' \) polarized, valley neutral, and fully \( K \) polarized currents respectively. A figure-of-merit for the valley-splitting efficiency of a dot is given by

\[
\xi_{\text{avg}} = \lim_{r \to \infty} \frac{1}{\pi} \int_{0}^{\pi} \frac{d\phi}{\pi} \xi(r, \phi),
\]

i.e. the far-field limit of \( \xi(r, \phi) \) averaged over the upper-half plane. This measures the valley polarisation of transverse currents, and non-zero \( \xi_{\text{avg}} \) indicates VHE-type behaviour.

In Fig 2, we consider a dot with \( \bar{\Delta} = 2, \bar{V}_{0} = 0 \) and \( R = 4.5 \), corresponding to tight-binding parameters \( V_{A/B}(r < R) = ±0.1 |t| \) for \( R = 10 \text{nm} \). The band edge inside the dot occurs at \( E = ±1 \) for the analytic results. We focus here on valley-dependent effects – for general scattering properties see [62]. \( \xi_{\text{avg}} \) (Fig 2a) quickly takes large negative values as \( E \) increases from zero, before it reaches a peak and decays to about half its maximum value, before vanishing at energies above the band edge. Electrons from the \( K' \) valley are therefore preferentially scattered in the \(+y\)-direction for all energies \( 0 < E < \frac{\Delta}{2} \), with the strongest effect near the band gap centre.

Fig. 2(b–d) examine the angular dependence of this effect by plotting the far-field limit of \( j_{\text{val}}^{\pm}(\phi) \) for the total current (solid line) and the individual valley contributions, shaded and unshaded areas. The three panels correspond to the three energies marked with symbols in Fig 2a). In all cases, we note equal \( K \) and \( K' \) contributions at \( \phi = 0, \pi \), corresponding to a valley-neutral situation for perfect forward- and back-scattering from the dot. At all other angles the two valleys scatter antisymmetrically with respect to the \( x \)-axis. This is most clear at the lowest energy (Fig. 2b), where scattering is largest in the transverse directions and electrons from different valleys scatter almost entirely to opposite sides of the dot. This corresponds to energies near the peak in Fig 2a). As \( E \) increases, the angular separation between the principal lobes of each valley decreases, leading to an overall preference for forward-scattering. Secondary pref-
FIG. 2. a) Valley polarization of scattered currents, averaged over the upper half plane. A strong $K'$ polarisation is seen at low energies. b)-d) Total (bold) and individual valley angular scattering profiles in the far-field limit, i.e. the radial component of $\mathbf{j}^\phi (r \to \infty)$, for the three energies ($E = 0.12, 0.7, 1.12$) highlighted in a). The three panels have the same normalization.

The spatial maps are complicated by bound states inside the dot, and beating patterns outside it due to contributions from a greater number of outgoing wave modes $H_m$. At very low energies, only the $m, \bar{m} = 0$ modes contribute, but a gradual onset of contributions from the $m, \bar{m} > 0$ modes occurs as $E$ increases. Although individual modes are valley-antisymmetric, phase differences lead to a decrease in $\xi_{\text{avg}}$ as valley-splitting effects from different modes partially cancel. However, the presence of higher-order modes maintains the overall sign of $\xi_{\text{avg}}$ within the gap [62], resulting in consistent deflection directions for a wide range of energies and dot sizes.

FIG. 3. Electronic and valley current flow, calculated within the Dirac model, for the lowest (a-c) and highest (d-f) energies marked in Fig. 2. a,d) Local current flow (arrows) and valley polarisation (colour). b,e) Electronic current flow (arrows) and electron density (colour) for the $K$ valley only. c,f) Valley current (arrows and colour).

We next consider local behaviour near the dot for low and high energy cases (purple and black symbols in Fig. 2a). The columns in Fig. 3 show, from left to right, the electron current (black arrows) and its valley polarisation (color), the current (arrows) and electron density (color) for $K$ valley electrons only and the effective valley current around the dot. At first glance, the valley polarisation at low energy (Fig. 3a) seems to agree with the angular scattering results from Fig. 2b, i.e. electrons from different valleys flow at opposite sides of the dot. Closer comparison of Figs. 3a and 2b reveal that the local and far-field scattering currents have opposite valley polarisations. To reconcile these two pictures, we observe that the valley-polarised currents in the immediate vicinity of the dot in Fig. 3a recombine shortly beyond the dot to give a largely valley-neutral forward current, and so do not contribute significantly to far-field effects. This is also evident from Fig. 3b, where the high $K$-valley current around the top of the dot diffuses quickly beyond it. More important contributions come from currents slightly further from the edge of the dot, which flow in valley-alternating streams that are antisymmetric upon reflection through the $x$-axis. It is the $K$ current flowing to the bottom left of the dot in Fig. 3b which dictates the behaviour which survives to the far-field limit. The valley-alternating streams are more obvious at larger energies (Fig. 3d,e) where their frequency is higher. Valley current flows around the dot in a series of counter-propagating channels (Fig. 3c,f), the nearest of which to the dot diffuses quickly beyond it, whereas the next channel maintains a more concentrated flow, and is almost transverse to the incoming electron wave at low energies (Fig. 3c).

To consider larger dots or stronger mass terms, we note that $R$ appears only multiplied by a wavevector $k$ or $q$. $\xi_{\text{avg}}$ therefore has a master diagram, shown in Fig. 4a), with axes proportional to the energy and dot mass [63]. The dotted line denotes the band edge, and we observe different behaviour inside (top-left) and outside the gap (bottom-right). A uniform preference for enhanced $K'$ scattering is observed inside the gap, with a weaker oscillatory pattern noted outside the gap where resonances with bound states inside the dot are observed. The strongest polarization is at the gap centre for all dot sizes and mass strengths.

To test the robustness of the valley-splitting behaviour seen in Fig. 3, we perform tight-binding simulations using the patched Green’s function approach [21, 56]. Here a finite dot is embedded into an infinite sheet...
of graphene, and we perform non-perturbative transport calculations using point-like source probe very far from the dot (250 nm) so that the incoming electrons closely resemble a plane wave, as in Fig. 1a). The system Green’s functions [64], $G(E)$, and lead broadening term ($\Gamma$) give the spectral density of injected states $A(E) = G(E)\Gamma G^{\dagger}(E)$, and local current flow in real-space. Projecting $A$ onto the pristine graphene basis $|\psi_k\rangle$ gives a measure of the local distribution of scattered states in $k$-space, $\rho(k) = \langle \psi_k | A | \psi_k \rangle$, from which the valley polarisation in real space can be calculated $\xi_{TB} = (\sum_{k\in K} \rho(k) - \sum_{k\notin K} \rho(k)) / \sum_{k\in K,K'} \rho(k)$. The analytically-predicted valley polarisation is reproduced numerically. At low energies in particular, this holds not only for a uniform dot, but also for non-uniform mass distributions. This is shown for a disordered mass term in Fig. 4b, where onsite potentials are placed only on 1% of sites within the dot, but with increased magnitudes to preserve the average mass term. A Gaussian distribution $\Delta \rho(r)$ with $\sigma = R/2$ and cut-off $R_c = 2R$ is also considered in Fig. 4c, with an amplitude such that $\langle \Delta \rho \rangle R_c \approx \Delta R$. In both these cases, and for other dot shapes, the valley polarisation is almost identical to the analytic model. In particular, the strong valley-splitting features at low energies do not crucially depend on the exact composition of the mass dot. A relative insensitivity to the strength, size, composition or shape of the mass regions suggests that experimental realisation of valleytronic devices may be possible through substrate [44-46] or defect engineering [48-55] and with less constraints than for strain-based proposals.

The equal and opposite deflections of $K$ and $K'$ electrons in the $y$ direction give rise to a pure valley current in the transverse direction. This is reminiscent of Berry curvature-related deflections predicted for a uniform mass term. In general, intrinsic topological Hall currents emerge due to Fermi sea contributions, e.g. primarily from Berry curvature hotspots at the band edge below the Fermi energy, and give rise to a quantised $\sigma_{xy} = \frac{2e^2}{\hbar}$ plateau within the band gap [2, 31, 36, 65]. Experimental $R_{NL}$ are often explained by such a mechanism [66]: a VHE in graphene, due to a hBN-induced mass term, creates a long-rangevalley current which enhances $R_{NL}$ beyond standard Ohmic contributions. However, within the relevant linear response regime, only Fermi surface contributions should play a role in device measurements [39, 40]. $R_{NL}$ peaks in the band gap can also arise due to non-topological edge channels at the Fermi surface [40]. Extrinsic Hall currents, appearing only at the Fermi surface, can occur due to scattering [67], such that non-zero Fermi surface contributions to $\sigma_{xy}$ act as a smoking gun for extrinsic Hall effects.

To clarify the role of scattering from non-uniform mass distributions, we consider a periodic array of mass dots. We employ the single-particle Kubo-Bastin formula [23, 57, 58, 68-72] to calculate both the complete and Fermi surface contributions to $\sigma_{xy}$ [62]. The density dependence is investigated by placing a $R = 10$ nm mass dot into three different square supercells with side lengths $W_C = 5R$, $10R$, $20R$. Fig. 5a) shows that $\sigma_{xy}$ has a peak around $E = 0$ in each case. Aside from valley-splitting due to scattering, such a peak could arise due to an effective, uniform mass $\approx \Delta$ due to the finite concentration $c$ of sites with a mass term $\Delta$. However, under this mechanism the peak height should remain constant, with wider peaks expected for larger $\Delta_{eff}$ (smaller $W_C$). Instead, the peak height decreases quickly with dot density, while the peak width is independent of $W_C$ so that the three curves coincide when normalised by their amplitude (Fig. 5b). This suggests a valley-splitting mechanism similar to an extrinsic spin Hall effect induced by skew-scattering from impurities [73]. In this case, the magnitude of $\sigma_{xy}$ should vary with the dot density, but with an energy dependence following the scattering profile of a single dot, as observed. To further test this hypothesis, we examine the Fermi surface (FS) contributions to $\sigma_{xy}$, and find it remains finite throughout the band gap for the per-
otic mass dot system in Fig. 5c), with $\sigma_{xy}^{\text{FS}} \approx 0.4 \sigma_{xy}$ at $E = 0$. For a uniform mass, this contribution vanishes (aside from broadening effects [62]) inside the gap, as shown in Fig. 5d). Similar behaviour is found for each dot density [62], demonstrating that a robust Fermi surface contribution to $\sigma_{xy}$ emerges in the presence of non-uniform mass distributions.

**Conclusions.** We have determined the valley dependence of electrons scattered from finite mass dots in graphene, and illustrated a clear splitting of electrons according to their valley index. This effect is robust over a wide range of dot sizes, mass distributions and electron energies. We also demonstrated that a non-uniform mass distribution in graphene, consisting of a periodic array of such dots, gives rise to a valley Hall conductivity whose behaviour contrasts sharply with that of the uniform mass case. In particular, a non-quantised mass dot system in Fig. 5c), with $\sigma_{xy}$ vanishes (aside from broadening effects [62]) inside the gap, as shown in Fig. 5d). Similar behaviour is found for each dot density [62], demonstrating that a robust Fermi surface contribution to $\sigma_{xy}$ emerges in the presence of non-uniform mass distributions.

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