Resonant tunnelling between the chiral Landau states of twisted graphene lattices

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A class of multilayered functional materials has recently emerged in which the component atomic layers are held together by weak van der Waals forces that preserve the structural integrity and physical properties of each layer. An example of such a structure is a transistor device in which relativistic Dirac fermions can resonantly tunnel through a boron nitride barrier, a few atomic layers thick, sandwiched between two graphene electrodes. An applied magnetic field quantizes graphene’s gapless conduction and valence band states into discrete Landau levels, allowing us to resolve individual inter-Landau-level transitions and thereby demonstrate that the energy, momentum and chiral properties of the electrons are conserved in the tunnelling process. We also demonstrate that the change in the semiclasic cyclotron trajectories, following an inter-layer tunnelling event, is analogous to the case of intra-layer Klein tunnelling.

A n electron moving through the hexagonal crystal structure of graphene is not only quasi-relativistic but also exhibits chirality. The chirality originates from the diatomic unit cell of the graphene crystal lattice and means that the amplitude of the electron wavefunction is intrinsically coupled to the direction of motion. This gives rise to the phenomenon of Klein tunnelling, whereby an electron can pass with unity transmission through a potential barrier formed in the graphene layer. In principle, chirality should also affect the properties of devices in which electrons tunnel between graphene layers. To investigate this effect, we focus on a van der Waals heterostructure in which Dirac fermions can resonantly tunnel between two graphene electrodes separated by a hexagonal boron nitride tunnel barrier.

Recent work has demonstrated that even a small misalignment of the crystalline lattices of the two graphene electrodes lowers the translation symmetry in the plane of the tunnel barrier and gives rise to an impulse which modifies the dynamics of the tunnelling electron. By applying a magnetic field perpendicular to the layers we show that electron tunnelling is governed by the laws of conservation of energy and in-plane momentum, as was demonstrated for the case of inter-Landau-level (inter-LL) tunnelling between two quantum wells in III–V semiconductor heterostructures. Here, our measurements on electron tunnelling between two graphene layers reveal an additional phenomenon, namely, chirality, and its enhancement by the quantizing magnetic field.

Our device, with bias, $V_b$, and gate, $V_g$, voltages applied, is shown schematically in Fig. 1a. It consists of a four-layer thick (1.4 nm) hexagonal boron nitride (hBN) tunnel barrier sandwiched between two high-purity crystalline graphene electrodes. The active area of the device, in which tunnelling occurs, is 100 $\mu$m$^2$. The lower graphene electrode sits on an atomically flat hBN layer (lower yellow region in Fig. 1a), directly above a SiO$_2$/n-Si substrate, in which the doped Si layer acts as the gate electrode and the oxide thickness is 300 nm. Our device is thus a graphene–boron nitride analogue of an independently contacted GaAs/AlGaAs double quantum well structure with back gate.

The device fabrication is described in the methods section of ref. 6. The two graphene layers are intentionally aligned to within an angle of 1° (see ref. 6 for details). However, even this slight misalignment, or twist angle, $\theta$, leads to a significant $k$-space displacement of magnitude $\Delta K = |\Delta K_b| = |K_b^c - K_b^w| = 2\sin(\theta/2)|K_b^w|$ of the Dirac cones at the Brillouin zone corners (see Figs 1b and 2a, insets). This displacement induces an impulse on a tunnelling electron and has a large effect on the measured current–voltage characteristics and their magnetic field dependence.

Figure 2a (black dashed curve) shows the measured current–voltage curve, $I(V_b)$, at $V_g = 0$ in the absence of a magnetic field. The current increases at a threshold bias voltage $V_t$ and reaches a resonant peak when $V_b = V_t = 0.58$ V, beyond which there is a region of negative differential conductance (NDC). When $V_b = V_t$, see inset i of Fig. 2a, the Fermi circle in one cone partially overlaps with empty states in the other, so that electrons can tunnel with $\alpha$ a cone displacement induces an impulse on a tunnelling electron and has a large effective change in the semiclassical cyclotron trajectories, following an inter-layer tunnelling event, is analogous to the case of intra-layer Klein tunnelling.

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Effect of magnetic field: experiment and theory
Landau level quantization induces weak features in $I(V_b)$ when $V_b = 0$ for 0.08 V < $V_b$ < 0.35 V (see region of the red curve in Fig. 2a indicated by the red horizontal bar) and sharp, large amplitude, resonant features in the differential conductance, $G(V_b) = dI/dV_b$, as shown in Fig. 2b for gate voltages $V_g = 0$ and ±40 V. By combining similar plots at intermediate gate voltages, we generate the colour maps of $G(V_b, V_g)$ shown in Fig. 3a and c, for $B = 2$ and 4 T, respectively. The regions of high conductance are patterned by small ‘islands’ that originate from resonant tunnelling of electrons when LLs in the two graphene layers become aligned in energy (shown schematically in Fig. 4a,b). We observe no evidence of lifting of spin–valley degeneracies in the $G(V_b, V_g)$ plots at these magnetic fields. These islands are sharply defined for $|V_b| < 0.2$ V, but become broadened at high $|V_b|$, which could arise from carrier heating due to high current levels and/or increased lifetime broadening. In Fig. 3, there are regions of high $G$ along the curves, marked by black dots, where the chemical potential in one graphene electrode coincides with the high density of states at the $n_b = 0$ LL (and Dirac point energy) in that electrode. By contrast, when $B = 0$, the density of states vanishes at the Dirac point; so $G = 0$ when the chemical potential lies there.

We model our data (see Fig. 3b,d) using a Bardeen transfer-Hamiltonian approach, taking the full two-component form of the LL eigenstates and the following device parameters: the doping densities in the bottom and top graphene layers are $2.0 \times 10^{11}$ cm$^{-2}$ (p-type) and $3.6 \times 10^{11}$ cm$^{-2}$ (n-type) respectively, and the twist angle $\theta = 1\degree$. We also include a Gaussian scattering potential with a spatial width of $\sigma = 9$ nm, which arises from disorder in the graphene electrodes and allows small $k$ changes ($< \Delta K$) that broaden the linewidth of the current peak (see Supplementary Section 5). A fit to the $I(V_b, V_g)$ curves at $B = 0$ (ref. 6) provides accurate values of these parameters (see Supplementary Information).

Our model gives a good fit to the magneto-tunnelling data, in particular the shape and relative strength of the islands of high conductance. It enables a detailed analysis of the pattern of conductance peaks, which is dominated by the lateral overlap of the Landau states. The height of the boron nitride barrier affects the tunnel current magnitude, but has little effect on the relative heights of the conductance peaks. We now focus on the underlying physics that controls the overall pattern of peak amplitudes, in particular the effect of twist angle and chirality on the tunnelling process.

Transition rates between chiral LL eigenstates
The displacement, $\Delta K$, of the Dirac cones due to the twist angle is shown schematically in Fig. 4a,b. It can be represented by, and is equivalent to, the effect of a strong pseudo-magnetic field applied parallel to the graphene layers. We describe the combined effects of the misalignment and the Landau-quantizing applied magnetic field by a vector potential given by

$$A_{b,t} = \left( l\hbar \Delta K^+ , -eBx + l\hbar \Delta K^+ , 0 \right) / c$$

(1)

where $l = 0, 1$ for the b, t layers. In a perpendicular magnetic field, the electron wavefunctions at the $K^+$ point have the analytic forms given by

$$\psi_{m_b,k_{x,b},y}^{K^+}(r) \propto \exp\left( ik_{y,b} \right) \begin{pmatrix} \phi_{m_b} \left( \text{sgn}(2m_b) \right) \\ -\phi_{m_b-1} \end{pmatrix}$$

These two-component chiral states comprise plane waves along $y$, with wavenumber $k_{y,b}$ in the b and t layers, and simple harmonic
oscillator (SHO) waves, \( \phi \), along \( x \) with indices that differ by 1. The centres of the SHO wavefunctions in the top and bottom layers are shifted by \( l_0^2 \Delta K^\pm \) and there is an additional plane wave factor for the top layer whose argument is \( \Delta K^\pm (x - X_t) \), where \( X_t = l_0^2 (k_y + \Delta K^\pm) \). The Bloch states near the \( K^- \) point have a similar form and make an equivalent contribution to the tunnelling matrix element (see Supplementary Information). The tunnel rates between LLs, \( W(n_i, n_f) \), depend on the overlap integrals of the initial and final wavefunctions summed over the \( k \)-states in the two layers (see Supplementary Information), and therefore permit tunnelling between SHO states with a range of different \( n \) indices. Figure 4a,b shows the energies and semicircular trajectories (orbits) of the quantized Landau states.

Figure 4c is a colour map of the inter-LL transition rate \( W(n_0, n_1) \) at \( B = 4 \) T (see equation (25) of the Supplementary Information). It reveals narrow yellow regions where \( W(n_0, n_1) \) is high. In other areas (black), tunnelling is suppressed. The regions of high \( W(n_0, n_1) \) originate from the spatial form and relative positions of the wavefunctions in the bottom and top electrodes. Within the upper right and lower left quadrants of the colour map, transitions between equivalent bands (conduction–conduction, \( c-c \), and valence–valence, \( v-v \)) are strongly enhanced compared to tunnelling between different bands (\( c-v \) and \( v-c \)). This asymmetry, found for all values of \( B \), is a consequence of the chiral nature of the electrons in graphene and arises from the interference of the electron wavefunction on the two sublattices of the bottom and top graphene layers. The figure shows that when the chirality of the electron is conserved—that is, for transitions between equivalent bands—tunnelling is enhanced. In contrast, when we remove the effect of chirality from our model by using pure (single component) LL wavefunctions, the tunnelling matrix elements are the same for transitions between equivalent and different bands (see Supplementary Information).

**Effect of chirality on tunnel current**

The asymmetry in the transition rate colour map in Fig. 4c manifests itself in the observed pattern of conductance peak amplitudes. In certain regions of the \((V_b, V_g)\) plot, tunnelling is exclusively between equivalent bands, as shown in Fig. 3. Here, the black and yellow dashed curves bound the regions of \( V_g - V_b \) space, where tunnelling is either only \( c-c \) (upper region, \( V_g > 0 \)) or \( v-v \) (lower region, \( V_g < 0 \)), respectively. Within these regions the amplitudes of most resonant peaks are high (shown as dark red). Increasing \( V_b \) beyond the lower region induces a changeover from tunnelling between

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Figure 3 | Differential magnetoconductance maps: experiment and theory. Colour maps showing \( G(V_b, V_g) \) at \( T = 4 \) K measured (a) and calculated (b) when \( B = 2 \) T and measured (c) and calculated (d) when \( B = 4 \) T. Colour scales for (a) and (c) are in microseconds, whereas for (b) and (d) they are normalized to the maximum conductance in the maps. Black and yellow dashed curves enclose regions around \( V_g = 0 \) within which only conduction–conduction band (region labelled \( c-c \) with \( V_g > 0 \)), or only valence-valence band (region labelled \( v-v \) with \( V_g < 0 \)) tunnelling occurs. White right angles in (a) and (b) mark upper corners of the colour map regions enlarged in Fig. 5a,b, respectively. Filled black circles running top left to bottom right (bottom left to top right) show loci along which the chemical potential the bottom (top) layer intersects with the Dirac point in that layer.
equivalent bands to a mixture of tunnelling between equivalent and different bands, and is therefore accompanied by a suppression of the conductance peaks. This is a direct manifestation of electron chirality. A similar changeover occurs as $V_b$ decreases across the left-hand edge of the region bounded by the upper dashed curve in Fig. 5a–d.

The effect of chirality on the peak amplitudes in these regions is seen more clearly in the enlarged lower region of the $G(V_b, V_g)$ maps at $B = 2$ T shown in Fig. 5a–c. In both our experiment (a), and calculations (b), the conductance peak amplitudes are larger in the bounded region in the lower left-hand side of the plot (labelled L in Fig. 5d), where $v-v$ tunnelling dominates, and smaller in the bounded region in the lower right-hand side of the plot (labelled R in Fig. 5d), where tunnelling is a mixture of $v-v$ and $v-c$ transitions. For comparison, in Fig. 5c we show $G(V_b, V_g)$ calculated when chirality is ‘switched off’—that is, with each eigenstate represented by a single SHO wavefunction with no pseudospin component (see Supplementary Information). In contrast to the chiral theory and experimental data, the conductance peaks for the non-chiral calculations have similar amplitudes in regions L ($v-v$) and R ($v-v$ and $v-c$).

To quantify the effect of chirality on the tunnel current, we calculate the ratio of the mean conductance in region L to that in region R, $(G)_L/(G)_R$ (see Fig. 5d). In the bar chart in Fig. 5e we show $(G)_L/(G)_R$ when $B = 0$, 2 and 4 T. For each field value, $(G)_L/(G)_R$ for the measured data (red) and the chiral calculations (yellow) are similar to each other. In contrast $(G)_L/(G)_R$ is significantly smaller for the non-chiral calculations (blue). In addition, with increasing $B$ the difference between the chiral and non-chiral results becomes larger: at higher $B$ there are fewer LL transitions within regions L and R and, for those transitions that do occur, the difference between the chiral and non-chiral conductance is more pronounced. Hence, the measured dependence of the conductance peak amplitudes on $V_b$, $V_g$ and $B$ reveals and demonstrates the chiral nature of the electrons and the associated asymmetry in the tunnelling rates (see Fig. 4c).

**Nested and figure-of-8 cyclotron orbits**

A semiclassical picture, in which electrons undergo cyclotron motion in both real- and $k$-space, provides further insights into the physics of tunnelling in these devices. In $k$-space, the orbital radii in the two graphene layers are $\kappa_{k_1} = \sqrt{2|\mu_{k_1}|/\hbar}$ and the orbit centres are separated by $\Delta K^2$. The solid and dotted curves in Fig. 4c are loci of initial and final states along which the corresponding semiclassical orbits just touch, so that the tunnelling electrons can make a continuous classical trajectory in the $(k_x, k_y)$ plane. These loci are defined by

$$k_i = \Delta K(\theta) \pm \kappa_{k_i}$$

Here, the $-$ and $+$ signs specify, respectively, cyclotron orbits that describe a ‘figure-of-8’ ($F-8$) and nested (N) form. Examples are shown by the projected circles in the lower parts of Fig. 6a,b. The spatial variation of the real (dark) and imaginary (light) components of the corresponding two-component LL wavefunctions are also shown ($x$ axis re-scaled by $1/\mu_{k_1}$ to enable comparison between the $k$-space trajectories and the spatial form of the SHO wavefunctions). The maxima in the wavefunction amplitude are located at the turning points of the semiclassical orbit so that, when equation (2) is satisfied (that is, along the solid (dotted) locus in Fig. 4c for $N$ ($F-8$) orbits), the wavefunction overlap integral is large. This large spatial overlap produces the bright yellow regions of high tunnelling rates in Fig. 4c.

The twist angle of the graphene electrodes determines the value of $\Delta K(\theta)$ in equation (2) and thus shapes the boundary loci of the four distinct regions ($c-c$, $v-v$, $v-c$, $c-v$). The twist angle also manifests itself through the suppression of the $0-0$ transition at the centre of the plot, because the spatial displacement of the two states exceeds their spatial width. The scattering potential broadens the variation of the tunnelling rates with $n_h$ and $n_i$, but preserves the effects of chirality and of the spatial wavefunction overlap on the overall form of the plot (see Supplementary Information).

The N and F-8 semiclassical orbits determine the dependence of $G$ on $B$, $V_b$ and $V_g$. At the onset of current (see red curve and arrow labelled $V_1$ in Fig. 2a) the energetically aligned LLs correspond to semiclassical orbits with the F-8 form (see black rings in Fig. 4a). Consequently the matrix elements are large, allowing tunnel current to flow. At the resonant current peak (see red curve and arrow labelled $V_1$ in Fig. 2a) the Dirac cones just touch and their intersection is a straight line. As a result, all energetically aligned LLs have high matrix elements because all the corresponding
1.0
0
−0.1
0.0 0.1 0.2 0.3
Vb (V)
Vg (V)
80 120 0.0 0.4 0.8 0.0 0.4 0.8
d
exp
bc

Figure 5 | Effect of chirality on the differential magnetoconductance: experiment and theory. a–c. Colour maps showing $G(V_b, V_g)$ for $B=2$ T. a, b are enlargements of the lower parts of the colour maps in Fig. 3a,b respectively (defined by white right angles). a. Experimental data ($T=4$ K). b. Calculated using the full model with chiral electrons. c. Calculated using non-chiral wavefunctions—that is, each comprising a single simple harmonic oscillator state. Colour bars in a are in microseconds and colour bars in b,c are in normalized units. d. The solid curves in a–c enclose regions of the colour map where tunnelling is only v–v (labelled L) or a mixture of v–v and v–c (labelled R). e. Bar charts showing the ratio, $(G_L/G_R)$, of the mean conductance in regions L and R (see d) for the measured data (red), and calculated for chiral (yellow) and non-chiral (blue) electrons.

Consequently, in our semiclassical model, for $N$ orbits in both valleys $\langle \sigma \rangle$ is unchanged for equivalent band transitions but is rotated by 180° for transitions between different bands. In contrast, for F-8 orbits $\langle \sigma \rangle$ is reversed for transitions between equivalent bands and unchanged for transitions between different bands.

Cyclotron orbits and Klein tunnelling

In graphene, the chiral nature of an electron in the absence of a magnetic field can be expressed by the expectation value of the pseudospin operator with respect to the eigenstate. For the $K^\pm$ valley this expectation value is $\langle \sigma \rangle=s(\pm \cos\phi, \sin\phi)$, where $\phi$ is the polar direction of the wavevector. When $B \neq 0$, $\langle \sigma \rangle$ can be defined semiclassically in a similar way (see Supplementary Section 5).

Discussion

Our results show that the chirality can play an important role in determining the electronic characteristics of graphene-based
Figure 6 | Electron wavefunctions and semiclassical cyclotron orbits in the two graphene layers for figure-of-8 and nested tunnelling transitions. a,b: Upper: vertical (horizontal) curves show the real (imaginary) parts of the real space electron wavefunction in the bottom (red curves) and top (blue curves) graphene electrodes, respectively, with \( B = 4 \) T for \( n_y = 1 \) (red) and \( n_y = 3 \) (blue) (a) and \( n_y = 2 \) (red) and \( n_x = 16 \) (blue) (b). The x axis is scaled by \( \hbar / e B \) for comparison with lower plots: circles show corresponding figure-of-8 and nested cyclotron orbits in \( k\)-space (\( k_x, k_y \) axes inset and direction of motion marked by arrows) with orbit centres separated by \( \Delta k \). The vertical black lines connecting upper and lower parts of the figure show the classical turning points.

vertical devices. This property represents a degree of freedom additional to those which arise from electron spin and valley degeneracies and, therefore, should be taken into account when assessing possible performance of electronic components based on materials with Dirac-like energy spectra.

Received 19 May 2015; accepted 11 September 2015; published online 19 October 2015

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Acknowledgements
This work was supported by the EU Graphene Flagship Programme and ERC Synergy Grant, Hetero2D. M.T.G. acknowledges The Leverhulme Trust for support of an Early Career Fellowship. V.I.F. acknowledges support of a Royal Society Wolfson Research Merit Award. E.E.V. and S.V.M. were supported by NUST “MISiS” (grant K1-2015-046) and RFBR (15-02-01221 and 14-02-00792). Gratitude to E. Y. Andrei and A. V. Balatsky for useful discussions.

Additional information
Supplementary information is available in the online version of the paper. Correspondence and requests for materials should be addressed to L.E.

Competing financial interests
The authors declare no competing financial interests.