Supplementary Note 1

Derivation of model for electron scattering between top and bottom electrodes.

Here we consider how the misalignment of the Dirac cones of the graphene electrodes in our devices affects our measured $I(V_b)$ curves. We separate the real-space scattering potential, $V_S(\mathbf{r}, z)$, into the product of an in-plane $\mathbf{r}$ =(x, y)-part and a longitudinal z-part by expanding the (x, y)-part over the Bloch states centred on the Dirac point of the top graphene electrode (collector). We find that, to first order, the scattering potential $V_S(\mathbf{q})$ reduces to a form in which the angular misorientation of the two graphene layers appears only as a prefactor term outside the scattering integral. Hence, the misorientation reduces the overall amplitude of the tunnel current but not its dependence on V_b and / or V_g. In effect, apart from a small reduction of the tunnelling probability due to the misorientation, the effect of the scattering potential on the V_b- and V_g-dependence of the tunnel current is the same as if the two graphene layers were aligned.

The Bardeen Transfer Hamiltonian matrix element for a transition from a state Ψ_B in the bottom layer to a state Ψ_T in the top layer has the form

$$M^S_{BT} = \int_V dV \, \Psi^*_T(\mathbf{r}, z) V_S(\mathbf{r}, z) \Psi_B(\mathbf{r}, z), \tag{S1}$$

where the integral is over all space, V, $\mathbf{r}$ is the in-plane position vector, z is the co-ordinate in the tunnelling direction normal to the graphene plane and $V_S(\mathbf{r}, z)$ is the scattering potential that induces transitions between states with different in-plane wavevectors. This potential could originate, for example, from ionized impurities, mis-alignment of the graphene and barrier layers, and / or fluctuations in the electron density. The wavefunction for a state with wavevector $\mathbf{k}$ can be written in terms of basis functions,$\Phi_{j\mathbf{k}}$, on two identical atoms labelled 1 and 2 ($j = 1, 2$) per unit cell in the following way [44]

$$\Psi_{\mathbf{k}}(\mathbf{r}, z) = \chi_1(\mathbf{k})\Phi_{1\mathbf{k}}(\mathbf{r}, z) + \chi_2(\mathbf{k})\Phi_{2\mathbf{k}}(\mathbf{r}, z), \tag{S2}$$

where $\Phi_{j\mathbf{k}}(\mathbf{r}, z)$ is of the Bloch form:

$$\Phi_{j\mathbf{k}}(\mathbf{r}, z) = \frac{1}{\sqrt{A}} \exp(i\mathbf{k}.\mathbf{r}) u_{j\mathbf{k}}(\mathbf{r}) h(z). \tag{S3}$$

Here, $u_{j\mathbf{k}}(\mathbf{r})$ is the cell-periodic part of the Bloch function in the graphene layer, and $h(z)$ describes the z dependence of the electron wavefunction in the graphene and barrier.

In Eq. (S2), the χ_j factors have the well-known form for graphene within the tight binding approximation:

$$\begin{bmatrix} \chi_1 \\ \chi_1 \end{bmatrix} = \begin{bmatrix} e^{\mp i\theta_{\mathbf{k}}/2} \\ se^{\pm i\theta_{\mathbf{k}}/2} \end{bmatrix}, \tag{S4}$$

where the upper (lower) sign is used for the band around the K (K') point of Brillouin zone, $s = +1$ or -1 for electrons in the conduction or valance band respectively, and $\theta_{\mathbf{k}} = \arctan\left(k_y/k_x\right)$ specifies the orientation of the wavevector. When we take the overlap integral between states in the two different graphene layers, we have to consider all of the different combinations of the basis functions. However, for simplicity, here we only show explicitly the contribution to the matrix element due to the $j = 1$ parts of Ψ_T and Ψ_B i.e. ignoring the $j = 2$ components. These cross terms are reintroduced in Eq. (S18) and fully included in our numerical calculations.

We consider the case where the graphene lattices are rotationally misaligned by an angle ω, which produces a corresponding misalignment of equivalent Dirac points in the two graphene layers. We define the displacement of the Dirac cones in k space by the wavevector $\mathbf{Q_D}$, which satisfies $\omega = 2\sin^{-1}(3a|\mathbf{Q_D}|/8\pi)$ [15,45] Therefore, the matrix element becomes:

$$M^S_{BT} = \int_V dV \Psi^*_{\mathbf{k_T}+\mathbf{Q_D}}(\mathbf{r}, z) V_S(\mathbf{r}, z) \Psi_{\mathbf{k_B}}(\mathbf{r}, z), \tag{S5}$$

where $\mathbf{k_B}$ is the wavevector in the bottom layer relative to the Dirac point in that layer, and $\mathbf{k_T} + \mathbf{Q_D}$ is the wavevector of the electron in the top layer measured relative to the position of the same Dirac point in the LH layer. Equivalently, $\mathbf{k_T}$ is the electron wavevector in the top layer relative to the Dirac point position in that layer. In the following analysis we show that the relative rotation of the graphene layers does not affect the shape of the $I(V_b)$ curve for any V_g (although it does determine the magnitude). To do this, we first let the scattering potential be separable of the form

$$V_S(\mathbf{r}, z) = V_S(z) V_S^{||}(\mathbf{r}) \tag{S6}$$

and expand $V_S^{||}(\mathbf{r})$ over Bloch states centred on the Dirac point in the rotated (top) layer, i.e. we set

$$V_S^{||}(\mathbf{r}) = \sum_{\mathbf{k}} u^{V_S}_{\mathbf{k}}(\mathbf{r}) e^{i\mathbf{k}.\mathbf{r}} \tag{S7}$$

where the wavevector, $\mathbf{k}$, is relative to the the Dirac point in the bottom layer, i.e. expressed in terms of the unrotated co-ordinates in the bottom layer, and $u^{V_S}_{\mathbf{k}}(\mathbf{r})$ is the appropriate cell-periodic function. Writing $\mathbf{k} = \mathbf{Q_D} + \mathbf{k}'_{\mathbf{T}}$, the scattering potential becomes:

$$V_S^{||}(\mathbf{r}) = e^{i\mathbf{Q_D}.\mathbf{r}} \sum_{\mathbf{k'_T}} C(\mathbf{k'_T}) u^{V_S}_{\mathbf{k'_T}+\mathbf{Q_D}}(\mathbf{r}) e^{i\mathbf{k'_T}.\mathbf{r}}. \tag{S8}$$

Substituting this form of the potential into the matrix element, Eq. (S5), reveals that

$$M^S_{11BT} = \frac{1}{A}\int_V dV \left[\chi_1^*(\mathbf{k_T}+\mathbf{Q_D}) e^{-i\mathbf{k_T}.\mathbf{r}} u^*_{1\mathbf{k_T}+\mathbf{Q_D}}(\mathbf{r}) h_T(z) \times\right.$$

$$V_S(z) \underbrace{\left[\sum_{\mathbf{k'_T}} C(\mathbf{k'_T}) u^{V_S}_{\mathbf{k'_T}+\mathbf{Q_D}}(\mathbf{r}) e^{i\mathbf{k'_T}.\mathbf{r}}\right]}_{\overline{V_S^{||}}(\mathbf{r})} \chi_1(\mathbf{k_B}) e^{i\mathbf{k_B}.\mathbf{r}} u_{1\mathbf{k_B}}(\mathbf{r}) h_B(z). \tag{S9}$$

The function $\overline{V_S^{||}}(\mathbf{r})$ is closely related to the scattering potential $V_S^{||}(\mathbf{r})$. In particular, since these functions have the same Fourier coefficients, they have the same width in real and reciprocal space. In the barrier region, we assume that the wavefunctions have the form

$$h_B(z) \approx \frac{1}{\sqrt{D}} e^{-\kappa(z+b)} \quad \text{and} \quad h_T(z) \approx \frac{1}{\sqrt{D}} e^{\kappa z}, \tag{S10}$$

where b is the barrier width, κ is the decay constant in the barrier, and D is a normalization constant for $h(z)$, which is comparable to an inter-planar separation in graphite. Consequently, by also inserting the form of $\chi_1(\mathbf{k})$ given in Eq. (S4) for scattering between the K to K Dirac points, we find that the matrix element, Eq. (S9), becomes

$$M^S_{11BT} = \frac{e^{-\kappa b}}{AD} e^{i(\theta_T-\theta_B+\omega)/2} \Xi \times$$

$$\int_S dS e^{i(\mathbf{k_B}-\mathbf{k_T}).\mathbf{r}} u^*_{1\mathbf{k_T}+\mathbf{Q}}(\mathbf{r}) u_{1\mathbf{k_B}}(\mathbf{r}) \overline{V_S^{||}}(\mathbf{r}) \tag{S11}$$

where $\Xi = \int V_S(z) dz$ is an integral over the barrier region. Assuming that the magnitudes of $\mathbf{k_B}$ and $\mathbf{k_T}$ are much smaller than the width of the Brillouin zone, we may set $u_{1\mathbf{k}}(\mathbf{r}) = u_{1\mathbf{k=0}}(\mathbf{r})$ [46] so that the matrix element becomes

$$M^S_{11BT} = \frac{e^{-\kappa b}}{AD} e^{i(\theta_T-\theta_B+\omega)/2} \Xi \int_S dS e^{i(\mathbf{k_B}-\mathbf{k_T}).\mathbf{r}} \overline{V_S^{||}}(\mathbf{r}) u^*_{1\mathbf{k_T=0}}(\mathbf{r}) u_{1\mathbf{k_B=0}}(\mathbf{r}). \tag{S12}$$

Since the effective scattering potential, $\overline{V_S^{||}}(\mathbf{r})$ and plane wave in the integral vary slowly over a unit cell,

$$M^S_{11BT} = \frac{\gamma_{11}(\omega)e^{-\kappa b}}{AD} e^{i(\theta_T-\theta_B+\omega)/2} \Xi \sum_C e^{i(\mathbf{k_B}-\mathbf{k_T}).\mathbf{r}_C} \overline{V_S^{||}}(\mathbf{r}_C), \tag{S13}$$

where $\mathbf{r}_C$ is the position within the Cth unit cell, the sum is over all unit cells in the x, y plane, and $\gamma_{11}(\omega) = \int_S dS u^*_{1\mathbf{Q_D},\mathbf{k_T}=\mathbf{0}}(\mathbf{r}) u_{1\mathbf{k_B}=\mathbf{0}}(\mathbf{r})$ is the overlap integral over a unit cell. We can rewrite Eq. (S13) in the form

$$M^S_{11BT} = \frac{\gamma_{11}(\omega)e^{-\kappa b}}{dS_C AD} e^{i(\theta_T-\theta_B+\omega)/2} \Xi \sum_C e^{i(\mathbf{k_B}-\mathbf{k_T}).\mathbf{r}_C} \overline{V_S^{||}}(\mathbf{r}_C) dS_C, \tag{S14}$$

where dS_C is the area of a unit cell. Converting the sum to an integral gives

$$M^S_{11BT} = \frac{\alpha_{11}(\omega)e^{-\kappa b}}{AD} e^{i(\theta_T-\theta_B+\omega)/2} \Xi \int_S dS e^{i(\mathbf{k_B}-\mathbf{k_T}).\mathbf{r}} \overline{V_S^{||}}(\mathbf{r}), \tag{S15}$$

where $\alpha_{11}(\omega) = \gamma_{11}(\omega)/dS_C$. Since the Fourier transform of the effective scattering potential is $\overline{V_S^{||}}(\mathbf{q}) = \int_S dS e^{i(\mathbf{k_B}-\mathbf{k_T}).\mathbf{r}} \overline{V_S^{||}}(\mathbf{r})$, it follows that

$$M^S_{11BT} = \frac{\alpha_{11}(\omega)e^{-\kappa b}}{AD} e^{i(\theta_T-\theta_B+\omega)/2} \Xi \overline{V_S^{||}}(\mathbf{q}), \tag{S16}$$

where $\mathbf{q} = \mathbf{k_B} - \mathbf{k_T}$. The matrix element is therefore a product of two terms: the scattering potential in Fourier space, $\overline{V_S^{||}}(\mathbf{q})$, and a prefactor, which contains the barrier transmission coefficient, $e^{-\kappa b}$, and an amplitude term, $\alpha_{11}(\omega)$, which depends on the mis-orientation of the two graphene layers.

We obtain qualitatively similar $I(V)$ curves for a range of potentials with similar spatial extents. In our simulations we set

$$\overline{V_S^{||}}(\mathbf{q}) = \frac{1}{(q_c^2 + q^2)} \tag{S17}$$

where $q_c^{-1} \approx 12$ nm defines the spatial extent of the scattering potential.

If we consider the additional terms in the matrix element, and also scattering between like and unlike Dirac points we find that

$$M^S_{BT} = g(\theta_B, \theta_T) \frac{e^{-\kappa b}}{AD} \Xi \overline{V_S^{||}}(\mathbf{q}) \tag{S18}$$

where

$$g(\theta_B, \theta_T) = \alpha_{11}(\omega) \left[e^{i(\theta_B \mp \theta'_T)/2} + s_B s_T e^{-i(\theta_B \mp \theta'_T)/2} \right] +$$

$$\alpha_{12}(\omega)\left[s_T e^{i(\theta_B \pm \theta_T')/2} + s_B e^{-i(\theta_B \pm \theta_T')/2}\right], \tag{S19}$$

in which $\theta_T' = \theta_T + \omega$, $s = \pm 1$ for electrons in the conduction or valence band in the top or bottom layers and the upper sign is for transitions between like valleys (i.e. K to K or K' to K') and the lower sign is for transitions between unlike valleys (i.e. K to K' or K' to K). The current is then

$$I = \frac{4\pi e}{\hbar} g_V \sum_{BT} \left|M_{BT}^S\right|^2 \left[f_B(E_B) - f_T(E_T)\right] \delta\left(E_B - E_T\right). \tag{S20}$$

where the summation is over all states in the top and bottom layers and the Fermi functions for the top and bottom layers are given by

$$f_{B,T}(E_{B,T}) = \frac{1}{1 + e^{(E_{B,T} - \mu_{B,T})\beta}} \tag{S21}$$

where μ_B and μ_T are the chemical potential in the bottom and top electrodes respectively, and β^{-1} is the thermal energy.

Supplementary References

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