

Ultra-sharp lateral p - n junctions in modulation-doped graphene

Jesse Balgley,[†] Jackson Butler,[†] S. Biswas,[‡] Z. Ge,[¶] S. Lagasse,[§] T. Taniguchi,^{||}
K. Watanabe,[⊥] M. Cothrine,[#] D. G. Mandrus,[#] J. Velasco Jr.,[¶] R. Valentí,[‡]
and E. A. Henriksen^{*,†}

[†]*Department of Physics, Washington University in St. Louis, 1 Brookings Dr., St. Louis
MO 63130, USA*

[‡]*Institut für Theoretische Physik, Goethe-Universität Frankfurt, 60438 Frankfurt am Main,
Germany*

[¶]*Physics Department, UC Santa Cruz, 1156 High Street, Santa Cruz, CA 95064, USA*

[§]*Electronics Science and Technology Division, United States Naval Research Laboratory,
Washington, DC 20375, United States*

^{||}*International Center for Materials Nanoarchitectonics, National Institute for Materials
Science, 1-1 Namiki, Tsukuba, 305-0044, Japan*

[⊥]*Research Center for Functional Materials, National Institute for Materials Science, 1-1
Namiki, Tsukuba, 305-0044, Japan*

[#]*Material Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge,
Tennessee 37831, USA*

[@]*Department of Material Science and Engineering, University of Tennessee, Knoxville,
Tennessee 37996, USA*

E-mail: henriksen@wustl.edu

March 15, 2022

Abstract

We demonstrate ultra-sharp ($\lesssim 10$ nm) lateral p - n junctions in graphene using a combination of electronic transport, scanning tunneling microscopy, and first principles calculations. The p - n junction lies at the boundary between two differentially-doped regions of a monolayer graphene sheet, where one side is intrinsic and the other is charge-doped by close proximity to a flake of α -RuCl₃ across a thin insulating barrier. We extract the p - n junction contribution to the device resistance, and use it to place bounds on the junction width. We achieve an ultra-sharp junction when the boundary between the intrinsic and doped regions is defined by a cleaved crystalline edge of a α -RuCl₃ flake located 2 nm from the graphene. Scanning tunneling spectroscopy in heterostructures of graphene, hexagonal boron nitride, and α -RuCl₃ shows potential variations on a sub-10 nm length scale. Density functional theory calculations reveal the charge-doping of graphene decays sharply over just nanometers away from the edge of the α -RuCl₃ flake.

Introduction

Ideal p - n junctions in graphene with a step-function change in carrier density underlie the physics of Klein tunneling,¹⁻⁴ negative refraction required for Veselago lensing,^{5,6} guiding of plasmons⁷ and snake states,⁸ and may enable controlled anisotropy of the band velocity,^{9,10} novel electron-optical devices based on transformation optics,¹¹ or the ability to focus electron beams.¹²⁻¹⁵ In practice, p - n junctions defined by electrostatic gating are far from this ideal, with the change in carrier density taking place over ≈ 40 -100 nm due to fringe electric fields from the edges of the metallic gates,^{6,16,17} precluding observation of effects such as Veselago lensing that rely on sub-10-nm junctions.⁶ Here we employ modulation-doping of graphene by close ($\lesssim 2$ nm) proximity to α -RuCl₃,¹⁸ and a well-defined boundary to the doped region via a cleaved edge of the α -RuCl₃ flake, to create ultra-sharp junctions, demonstrated with evidence from electronic transport, scanning tunneling probes, and first

principles calculations.

When the layered Mott insulator alpha-ruthenium(III) chloride (α -RuCl₃) is placed in direct contact with graphene, it accepts approximately $4 \times 10^{13} \text{ cm}^{-2}$ electrons, leaving the graphene strongly hole-doped.¹⁹⁻²³ If an insulating spacer is introduced between the two materials, the charge transfer is weakened and the mobility increases commensurate with the setback of α -RuCl₃ from graphene, analogous to modulation doping of conventional two-dimensional electron gases.^{18,24} The spatial distribution of the hole-doping is determined by where the α -RuCl₃ overlaps the graphene which can in principle have an atomically-abrupt boundary. Thus charge-doping by α -RuCl₃ appears to be a viable route toward ultra-sharp p - n junctions in graphene.

Here we fabricate p - n junctions in graphene by a combination of modulation-doping to differentially charge-dope two regions, and electrostatic gating to independently tune the densities in each. We use the resistance measured across the junctions to extract the junction width and find an ultra-sharp, $\lesssim 10$ nm junction when a cleaved crystalline edge of the dopant α -RuCl₃ flake placed 2 nm away from the graphene defines the boundary between the regions. We further use low-temperature scanning tunneling microscopy and spectroscopy (STM/STS) to explore devices where a graphene sheet is either directly in contact with α -RuCl₃ or separated from it by thin flakes of hexagonal boron nitride (hBN). We observe a sharp change in the charge doping of the graphene over a sub-10 nm length scale across step edges in the insulating hBN spacer. Finally, we perform density functional theory (DFT) calculations that reveal the hole-doping of graphene due to electron transfer to α -RuCl₃ falls off rapidly over just a few graphene lattice constants away from the α -RuCl₃ edge.

Results

Electronic Transport

We present electronic transport in two graphene devices containing lateral p - n junctions. In both, half the graphene sheet is intrinsic while the other half is modulation-doped by an α -RuCl₃ flake. Device D1 has a ≈ 1.5 -nm-thick AlO_{*x*} film between the graphene and α -RuCl₃, while device D2 has a 2-nm-thick flake of hBN as a spacer. Figure 1a shows an optical microscope image of D1, which consists of a 16.5-nm-thick flake of α -RuCl₃ coated by the AlO_{*x*} film, topped by a graphene Hall bar that lies partly above the α -RuCl₃ and partly on the bare substrate. The Hall bar is capped by a flake of hBN (≈ 30 nm) supporting a global Cr/Au top gate, and is contacted by Cr/Au leads. The entire device rests on 300 nm of SiO₂ on p -Si; the latter also serves as a global back gate. Further fabrication details are given in the Supporting Information.²⁵ In Fig. 1a, the device regions labeled “g” and “mod” correspond to the intrinsic and α -RuCl₃-doped graphene, respectively, and a profile of the device stack is shown schematically in Fig. 1b.

Four-terminal resistance measurements at $T=4$ K of the g and mod side of device D1 are shown in Fig. 1d, as a function of the top gate voltage. On either side of the junction we see resistance maxima at the graphene charge neutrality point (CNP or Dirac point), shifted by a few volts relative to each other due to the p -type modulation-doping.^{19,20,26} Measurements of the low-field Hall coefficient, R_H , shown in Fig. 1e directly show the charge-doping difference to be 3.2×10^{12} cm⁻². Similarly, in the hBN-spaced device D2, the charge transfer is 1.5×10^{12} cm⁻².²⁵ Typical g-side (mod-side) mobilities and mean free paths in D1 range from 8,000-12,000 cm²/Vs and 100-250 nm (6,000-10,000 cm²/Vs and 50-200 nm).²⁵ Intriguingly, although the top and back gates are global, the carrier densities on either side can be independently tuned. Thus the back-gate electric field on the modulation-doped side must be screened by the α -RuCl₃.

The width of a graphene p - n junction can be determined by its contribution to the

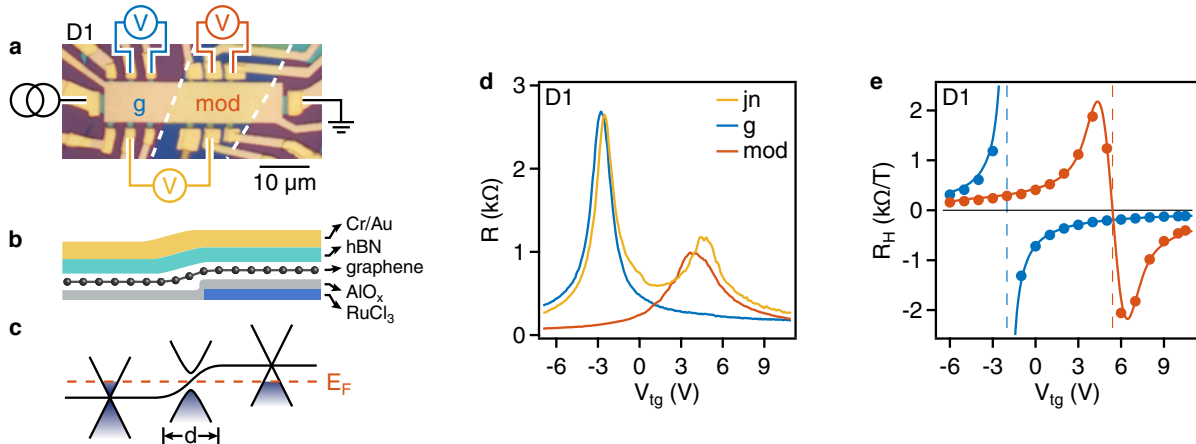


Figure 1: **Spatial control of modulation doping in graphene.** **a** Optical micrograph of device. The white dashed lines indicate the α - RuCl_3 flake boundary separating regions of intrinsic and α - RuCl_3 -modulation-doped graphene, labeled “g” and “mod”, respectively. **b** Schematic of device layer profile. **c** Schematic of the graphene band structure crossing from n -type to p -type across a junction of width d , showing an effective gap opening in the junction at the Fermi energy E_F . **d** Four-terminal resistance measured simultaneously in g (blue) and mod (orange) regions, and also across their interface (yellow), color-coded to the voltage measurement schematics of part **a**. **e** Low-field Hall coefficient of g and mod regions. Solid lines shows $R_H = -1/ne$ for the intrinsic (blue) and α - RuCl_3 -doped (orange) regions; the latter is convolved with a Gaussian of width $\sigma = 3.5 \times 10^{11} \text{ cm}^{-2}$ representing a spread of densities from electron-hole puddling.²⁷

total device resistance. Charge carriers incident on a p - n junction in graphene obey an electronic analog of Snell’s law at an interface of right- and left-handed optical materials: the momentum along the junction, $k_y = k_F \sin \theta$, is conserved, but the momentum k_x normal to the junction changes sign, the end result being a negative refraction.^{28,29} Here k_F is the Fermi momentum and θ is the carrier angle of incidence on the junction. Carriers are transmitted across an abrupt junction with probability $T(\theta) = \cos^2 \theta$ due to pseudospin conservation. In real devices there is always a density gradient from p - to n -type over some characteristic width d , analogous to the depletion region of a classical doped-Si p - n junction. Although there is no band gap in graphene, an effective gap to transmission arises when $k_x(x) = \sqrt{(E(x)/\hbar v_F)^2 - k_y^2}$ becomes imaginary, where $E(x) = \hbar v_F k_F$ is the position-dependent energy of the graphene Dirac point across the junction, and $v_F \approx 10^6 \text{ m/s}$ is the Fermi velocity. This is depicted schematically in Fig. 1c. Tunneling across this gap reduces

the transmission probability as a function of impact angle and junction width which, for a balanced junction ($|p|=|n|$), is given by²⁸

$$T(\theta, d) = \cos^2 \theta e^{-\pi k_F d \sin^2 \theta} . \quad (1)$$

The reduced transmission leads to a finite resistance that has both ballistic and diffusive contributions, $R_{p-n}=R_{bal} + R_{dif}$, whose relative magnitudes depend on the carrier mean free path and also many-body effects.^{25,30,31} Experimental values of R_{p-n} range from a few hundred ohms in graphene-on-oxide junctions to 100 Ω in hBN-encapsulated junctions.^{2,6,32}

We extract the width of lateral p - n junctions in two devices as follows, illustrating the procedure by analyzing the transport in device D1. First, in Fig. 2a we show the top- and back-gate dependence of the total resistance across the junction, R^{jn} , using the contacts marked in yellow in Fig. 1a for D1. This quantity includes the sheet resistances from both sides of the junction, and R_{p-n} due to the junction itself. The density of the intrinsic portion of the graphene depends on both gates as $n_g = \alpha_{tg}V_{tg} - \alpha_{bg}V_{bg} + n_{g,0}$, while the α -RuCl₃-doped portion depends only on the top gate via $n_{mod} = \alpha_{tg}V_{tg} + n_{mod,0}$. Here the top and back gating efficiencies are $\alpha_{tg} = 4.4 \times 10^{11} \text{ cm}^{-2}/\text{V}$ and $\alpha_{bg} = 6.0 \times 10^{10} \text{ cm}^{-2}/\text{V}$, respectively, and the densities for zero applied gate bias are $n_{g,0} = 1.1 \times 10^{12} \text{ cm}^{-2}$ and $n_{mod,0} = -1.3 \times 10^{12} \text{ cm}^{-2}$. Using these relations, in Fig. 2b we re-plot R^{jn} vs the carrier densities of the intrinsic graphene, n_g , and the modulation-doped graphene, n_{mod} . The CNPs of these two differentially-doped regions appear as vertical and horizontal bands separating the regimes of same-sign (p - p' or n - n') carrier transport across the interface from those having bipolar transport.

Next we isolate the resistance, R_{p-n} , of the p - n junction itself. We start with line cuts of R^{jn} along lines of equal carrier density and same sign ($n_g=n_{mod} > 0$, yielding R_{n-n}^{jn}) or opposite sign ($n_g = -n_{mod} > 0$, R_{n-p}^{jn}). These are plotted together in Fig. 2c. To the extent that sheet resistances on either side of the interface are symmetric with respect to charge neutrality, then the difference of these curves, $R_{odd}^{jn}=R_{n-p}^{jn} - R_{n-n}^{jn}$, will be due *only*

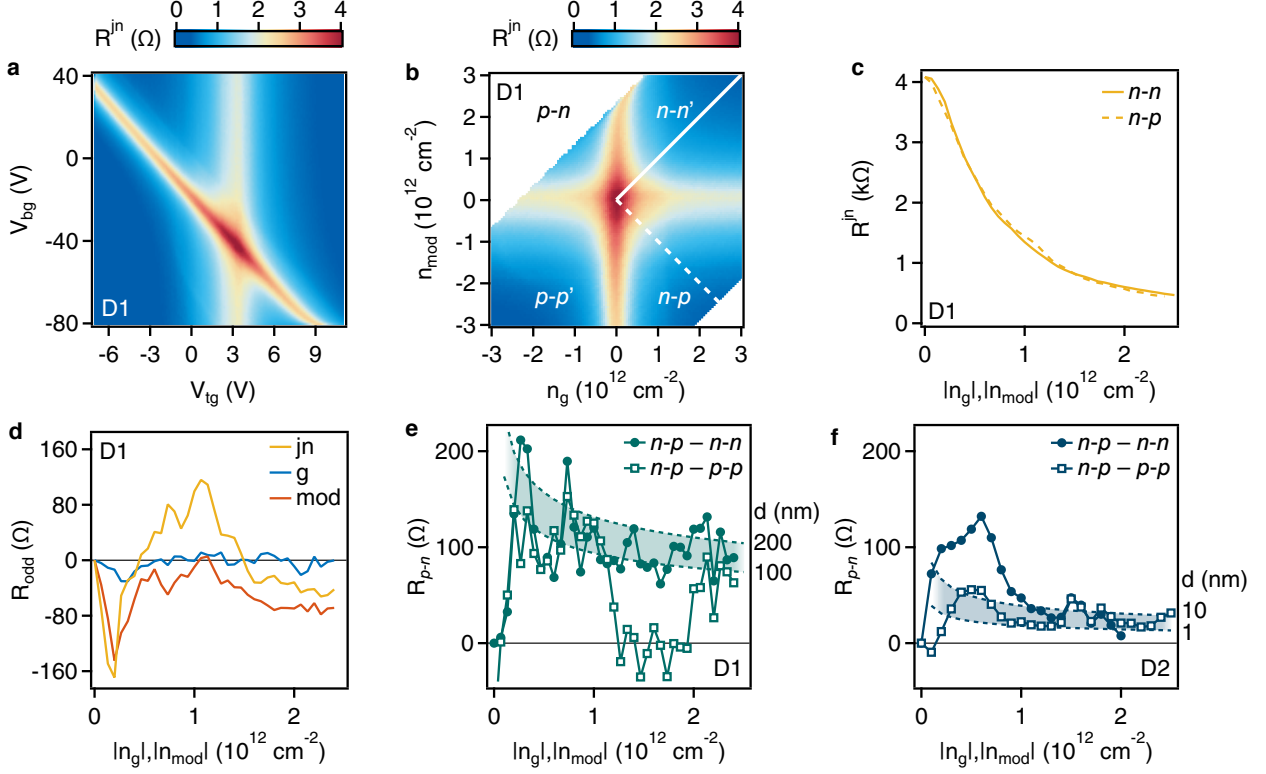


Figure 2: **Resistance across a modulation-doping-defined p - n junction.** **a** Four-terminal resistance across the interface of intrinsic (g) and modulation-doped (mod) graphene, as a function of top and back gate voltages. **b** Same as **a**, now re-plotted as a function of the g - and mod -side carrier densities. The labels show the polarity of the four quadrants defined by the charge neutrality peaks, either monopolar (e.g. n - n') or bipolar (n - p). White solid and dashed lines mark where the carrier density on either side of the interface is equal (n - n), or of equal magnitude but opposite sign (p - n). **c** Comparison of resistances at the white solid and dashed lines in **b**. **d** Difference of R^{jn} (yellow) for the two curves in **b**; and of the g -side resistance R^g (blue) and mod -side resistance R^{mod} (orange) for line cuts at the same carrier densities (or gate voltages). **e** p - n junction resistance for device D1. Shaded region marks the theoretical resistance for a ballistic device with junction width ranging between 100 and 200 nm. **f** p - n junction resistance for device D2. Shaded region marks the theoretical resistance for a ballistic device with junction width ranging between 1 and 10 nm.

to the resistance of the p - n junction: $R_{odd}^{jn} = R_{p-n}$. In fact, for the line cuts in Fig. 2c the contribution from the g -side sheet resistance ought to be identical because it does not change sign. In contrast, the two line cuts include either n - or p -type doping of the mod side, so any asymmetry about the CNP in this region will add a trivial resistance to R_{odd}^{jn} that must be subtracted off. To determine the presence of this trivial contribution, we make resistance

maps analogous to Fig. 2b for both the g and mod side.²⁵ From equivalent line cuts along the n - n and n - p directions, we calculate R_{odd}^g and R_{odd}^{mod} . These, along with R_{odd}^{jn} , are shown in Fig. 2d. As expected, R_{odd}^g lies close to zero; but R_{odd}^{jn} and R_{odd}^{mod} are finite and share a similar lineshape.

It remains to subtract this asymmetric part of the sheet resistance to finally obtain the p - n junction resistance: $R_{p-n} = R_{odd}^{jn} - (c_g \times R_{odd}^g + c_{mod} \times R_{odd}^{mod})$, where c_i are scaling factors appropriate to the device geometry.²⁵ Figure 2e shows the resulting R_{p-n} values, along with the analysis for line cuts along the (n - p) and (p - p) directions which should in principle yield the same junction resistance. Indeed, both show values of $\sim 100 \Omega$ (but for a brief excursion by the (p - p)-derived trace which can be attributed to a dip in R^{jn} ²⁵). Figure 2f shows the results of similar analyses carried out in device D2. Here, both curves show peaks at low density that rapidly converge to values between 20 and 30 Ω over much of the carrier density range. We compare these results to theoretical predictions for the resistance of p - n junctions in disordered graphene,³¹ which we plot as shaded bands calculated for junction widths d that span 100 to 200 nm and 1 to 10 nm in Fig. 2e and f, respectively. Device D2 is thus found to have an ultra-sharp, sub-10-nm junction, while D1 has a much wider ~ 100 nm junction.

At first, this result is surprising: why are the two junction widths so different? Both have insulating spacers of approximately the same thickness, with modulation-doping levels only a factor of two apart. The differing mobilities are unlikely to be the culprit, as transport across the junction is firmly in the ballistic regime.²⁵ The interface in device D1 is angled at 22° so the junction appears wider, but only by a factor of $1/\cos(22^\circ) \approx 1.08$.³³ Ultimately, inspection of the α -RuCl₃ flakes used in the devices offers a clear resolution: in D1, the edge of the α -RuCl₃ flake at the boundary between the intrinsic and modulation-doped regions is slightly curved, with no obvious relation to its crystalline axes. In contrast, for D2 the edge defining the boundary of charge transfer is straight and makes an angle of $\approx 119^\circ$ with another portion of the flake just outside where it contacts the graphene.²⁵ This implies the

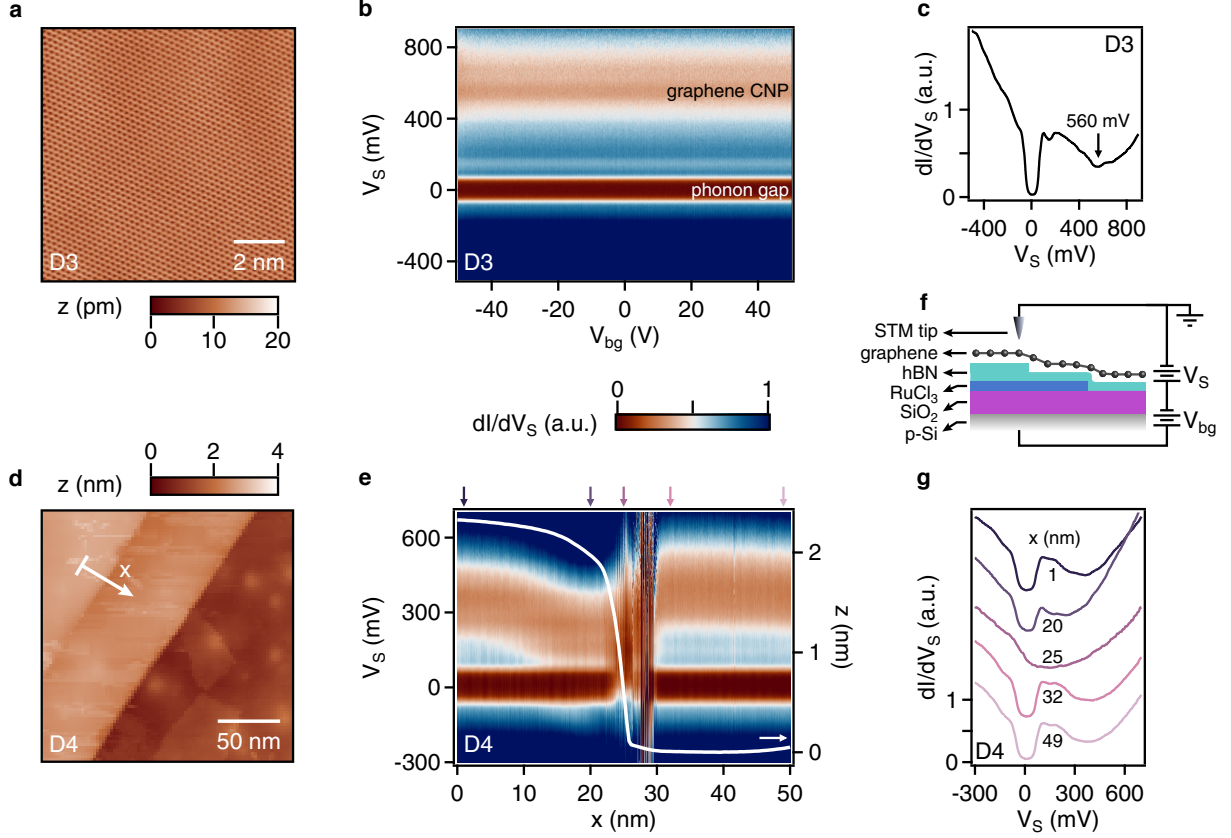


Figure 3: STM and STS across step edges in graphene/hBN/ α -RuCl₃ heterostructures. **a** Atomically resolved STM topography of a $10 \times 10 \text{ nm}^2$ region taken on graphene/ α -RuCl₃ in Device D3 at $T=4.8 \text{ K}$. **b** Color map of the measured dI/dV_S (V_S, d) from region in **a**, as a function of gate voltage applied to substrate. The strong feature near $V_S=0 \text{ mV}$ corresponds to phonon-assisted inelastic tunneling while the additional suppression in intensity near $V_S=560 \text{ mV}$ corresponds to the graphene charge neutrality point. **c** Average of spectra across all V_{bg} shows a clear phonon gap and minimum at CNP. **d** STM topography of a $200 \times 200 \text{ nm}^2$ window in a graphene/hBN/ α -RuCl₃ region in Device D4. **e** Color map of the measured dI/dV_S along the white line over a step edge in **d**. The phonon gap and graphene CNP are readily visible, with the latter showing a non-monotonic dispersion as the tip travels over the step edge. White line tracks the change in tip height in crossing the step. **f** Schematic of measurement over step edges in graphene/hBN/ α -RuCl₃/SiO₂/Si heterostructure. **g** Measured dI/dV_S spectra at various x positions (labeled in figure) along white line in **d**, highlighting the non-monotonic shift of the CNP feature.

boundary in D2 is a cleaved crystalline edge, and in D1 is likely to be rough with various facets along the edge.

Scanning Tunneling Microscopy

We use scanning tunneling microscopy and spectroscopy at $T=4.8$ K to study the spatial variation of the Dirac point across differentially-doped regions in two other devices, D3 and D4, both composed of overlapping flakes of graphene, hBN, and α -RuCl₃ on a SiO₂/*p*-Si substrate. Figure 3a shows an atomically resolved topographic map of a region in D3, consisting of graphene in direct contact with α -RuCl₃. The differential tunneling current, dI/dV_S , proportional to the local density of states (LDOS), is acquired as a function of the tip-sample bias, V_S , and a back gate voltage applied to the substrate, V_{bg} , with results plotted in Fig. 3b where a dark blue (brown) color corresponds to high (low) LDOS. A strong dark brown band centered about $V_S=0$ mV appears along with several fainter features. We show the averaged spectra from $V_{bg}=-50$ V to +50 V in Fig. 3c which shows a 120-mV-wide U-shaped suppression of dI/dV_S centered about $V_S=0$ mV, with a less pronounced minimum at $V_S=560$ mV. The former is a familiar phonon-assisted inelastic tunneling gap,³⁴ while the latter corresponds to the graphene CNP.^{34,35} We estimate the graphene carrier density using $n_g = (E_{DP} - \hbar\omega)^2/(\pi\hbar^2v_F^2)$, with $\hbar\omega$ the phonon energy and E_{DP} the energy of the Dirac point in Fig. 3c, and find a large *p*-type doping of $n=-1.8\times 10^{13}$ cm⁻², on the low side of prior observations of the graphene/ α -RuCl₃ charge transfer.^{18,19,23} The surprising lack of response to the back gate corroborates the screening effect in noted above in transport for α -RuCl₃-doped graphene.

In Fig. 3d, we show a topographic map of a region in device D4 that shows terraces due to separation of graphene and α -RuCl₃ by an hBN spacer of varying thickness, shown schematically in Fig. 3f. In Fig. 3e, we show dI/dV_S spectra acquired over one such edge (along the arrow in Fig. 3d) as a function of both V_S and position x to map the change in charge transfer. The white curve shows the height profile (right axis). As above, the phonon gap appears at $V_S=0$ mV, but the graphene CNP feature disperses non-monotonically with x , briefly decreasing as the step edge is approached and then sharply increasing to a final plateau once the step edge is crossed. Point spectroscopy taken at different x values (shown

in Fig. 3f) illustrates the non-monotonic variation of the CNP across the step edge. Far from the edge, the charge density is found to be $5.8 \times 10^{12} \text{ cm}^{-2}$ ($8.3 \times 10^{12} \text{ cm}^{-2}$) for the higher (lower) step, confirming that larger charge transfers are associated with thinner hBN spacer layers. The positive shift of the CNP takes place rapidly over $\approx 7 \text{ nm}$. An instability in STS is observed right at the step edge, where we suspect the graphene is suspended over the step and thus briefly decouples from the underlying hBN and $\alpha\text{-RuCl}_3$.

Density Functional Theory Calculations

To understand both the lateral and vertical spatial distribution of the charge transfer due to the modulation-doping of graphene by $\alpha\text{-RuCl}_3$, we perform first principles calculations of a monolayer-thick $\alpha\text{-RuCl}_3$ ribbon on graphene as shown in Fig. 4a and b. By using DFT calculations as implemented in VASP,²⁵ we first calculate the properties of the interface when no spacer layers are present in a large supercell in the ribbon-on-sheet geometry (see Fig. 4a), with supercell lattice parameters $a=34.16 \text{ \AA}$, $b=9.84 \text{ \AA}$ and $c=20 \text{ \AA}$ (distance between periodic images along c direction is $\approx 16.5 \text{ \AA}$), and the graphene C-C bond-lengths fixed at 1.42 \AA . Geometrical optimization of the internal atomic degrees of freedom leads to a mildly distorted Ru hexagon with shorter Ru-Ru bonds ($l_s=3.17 \text{ \AA}$) arranged in a periodic-step-function like pattern along the direction \hat{b} , compared with the other Ru-Ru bonds of the ribbon ($l_s=3.48\text{-}3.52 \text{ \AA}$).

With the two materials in close proximity, a new charge density distribution develops which we illustrate by subtracting of the charge densities of intrinsic graphene and $\alpha\text{-RuCl}_3$ from that of the graphene/ $\alpha\text{-RuCl}_3$ heterostructure, $\Delta\rho = \rho_{\alpha\text{R/g}} - \rho_{\alpha\text{R}} - \rho_{\text{g}}$. We find that charge accumulates in the $\alpha\text{-RuCl}_3$ ribbon with a concomitant depletion in the graphene, as shown in Fig. 4b where we plot the charge isosurface at $|\Delta\rho| = 5 \times 10^{-4} e/\text{a.u.}^3$, and in Fig. 4c by directly plotting the variations in the planar average of $\Delta\rho$ (over $\hat{b} \times \hat{c}$) along the \hat{a} direction for the graphene and $\alpha\text{-RuCl}_3$ layers. These results are in accord with findings for the graphene/ $\alpha\text{-RuCl}_3$ commensurate bilayer case.²² Figure 4c shows the excess electronic

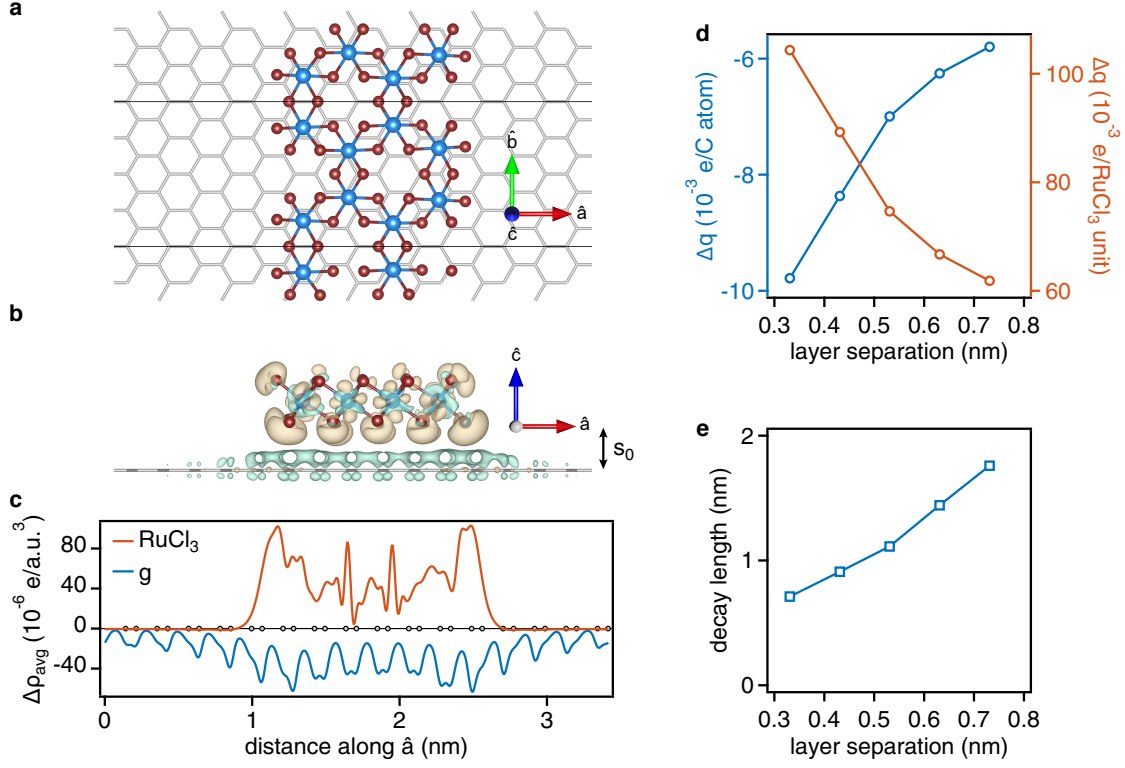


Figure 4: **Calculated charge densities of α -RuCl₃ ribbon on graphene.** **a** Top view of the supercell and the corresponding orthogonal directions of the lattice vectors (\hat{a} , \hat{b} , and \hat{c}). The blue (red) spheres represent the Ru (Cl) atoms. **b** Side view of supercell with illustration of the change in charge density, $\Delta\rho$, showing accumulation and depletion in beige and teal, respectively. The isosurface value of $\Delta\rho$ is chosen to be $5 \times 10^{-4} e/a.u.^3$, and $s_0 = 3.31 \text{ \AA}$ is the equilibrium separation of the relaxed geometry. **c** Planar average of the change in charge density (in the \hat{b} , \hat{c} plane) along the \hat{a} direction, $\Delta\rho_{avg}$, separated by color for charge on α -RuCl₃ (orange) and graphene (blue). **d** Total integrated charge on α -RuCl₃ (orange, right axis) and graphene (blue, left axis) as a function of separation between the layers. **e** Decay length of charge distribution in graphene as a function of lateral distance away from the edge of the α -RuCl₃ ribbon.

charge in α -RuCl₃ tends to lie largely on the Cl atoms facing the graphene. The majority of the charge depletion in graphene is concentrated at the C atom locations underneath the α -RuCl₃, reaches maxima near the boundaries of the α -RuCl₃ ribbon, and proceeds to decrease rapidly beyond the edge. Adding a second α -RuCl₃ layer does not qualitatively alter this result.²⁵

The equilibrium height of the α -RuCl₃ above the graphene, $s_0=3.31 \text{ \AA}$, is defined as the average distance between the C atoms in graphene and the graphene-facing Cl atoms in α -

RuCl₃, shown in Fig. 4b. To mimic the presence of a dielectric spacer layer, we calculate how the charge transfer changes if the separation is increased up to an additional 4 Å (without further relaxing the geometry, but with the supercell lattice parameter c also increased up to 24 Å). The results, in Fig. 4d, show a clear decrease of the charge exchange between the two layers, in qualitative agreement with the experimentally observed modulation doping effect. Performing the calculation with a dielectric present would alter the absolute magnitude of charge transfer but is expected to keep the relative changes similar to what we have found here.¹⁸

Finally, we can estimate the characteristic length scale over which the charge transfer decays away outside the ribbon by fitting the decrease of the charge density peaks around the C atoms, visible in Fig. 4c, as a function of distance. We find the best fit to the data is made using an equation of the form $Ae^{-(x-x_0)/B}$, with x_0 measured along \hat{a} from the average position of the zigzag C atoms just outside the α -RuCl₃ ribbon. The decay length, B , plotted in Fig. 4e is an average of the fits made on either side of the α -RuCl₃ ribbon, and is found to be roughly 2.5 times greater than the graphene/ α -RuCl₃ separation.

Discussion

We have demonstrated ultra-sharp p - n junctions in modulation-doped graphene devices. This innovation relies upon several advantages conferred by using α -RuCl₃ to charge dope graphene.

First, we use a cleaved crystalline edge to define an atomically-sharp and straight interface along the several-micron-length of the p - n junction. Prior work has determined that roughness of this interface can be a significant hurdle to achieving ultra-sharp junctions.³⁶ This advantage is not unique to α -RuCl₃; however, even in cleaved graphite-gate-defined p - n devices with sub-nm lateral roughness along the interface, 40-nm-wide junctions still are observed.¹⁷ Thus a sharp interface may be necessary but is apparently not sufficient to

obtain ultra-sharp junctions.

Therefore we note additional advantages unique to the α -RuCl₃ approach. Our devices are composed of two monolayer charge distributions (only the layer of α -RuCl₃ closest to graphene is appreciably charged^{18,25}). These form a nearly ideal parallel-plate capacitor geometry with arbitrarily small separation between the plates. Indeed, the charge distribution in the α -RuCl₃ shows accumulation near the edge of the ribbon, visible in Fig. 4c, just as expected for a classical charged sheet over a metallic plane. This suggests the extent of the potential variation in graphene beyond the edge of α -RuCl₃ (e.g. the junction width) is, apart from a possible role for nonlinear screening,³⁰ essentially a matter of electrostatics and thus due to the usual fringing electric fields which have a lateral extent on the order of the plate separation. Given this, it should be possible to achieve a similar result by implementing a graphite gate just as close to the graphene; however, this presents numerous practical difficulties including dielectric breakdown, unwanted leakage currents to the conducting gate, and the onset of tunneling for dielectric thicknesses below 2 nm.³⁷ These are *not* limitations for α -RuCl₃-doped graphene, for which the charge transfer is fixed, requires no external bias, and crucially has no leakage current due to the insulating nature of α -RuCl₃.

Thus the narrowest *p-n* junctions can be achieved by placing a flake of α -RuCl₃ with a cleaved edge as close as possible to the graphene. Junctions defined in this manner should be narrow enough to enable observation of electron-optical effects such as Veselago lensing and other useful devices based on electron refraction or reflection.⁶

Acknowledgments

We acknowledge enjoyable and informative discussions with K. Burch, D. Basov, Y. Wang, E. Gerber, and D. Rizzo. Support for device fabrication and measurement by the Institute of Materials Science and Engineering at Washington University in St. Louis is gratefully acknowledged. E.A.H., J.B., and J.B. received partial support from the National Science Foundation under DMR-1810305, and E.A.H. was additionally supported by NSF CAREER DMR-1945278. S.L. was supported by Office of Naval Research 6.1 Base Funding

and the Jerome and Isabella Distinguished Scholar Fellowship. K.W. and T.T. acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan (Grant Number JPMXP0112101001) and JSPS KAKENHI (Grant Numbers 19H05790, 20H00354 and 21H05233). J.V.J. and Z.G. acknowledge support from the National Science Foundation under award DMR-1753367. J.V.J. acknowledges support from the Army Research Office under contract W911NF-17-1-0473. S.B. and R.V. thank the Deutsche Forschungsgemeinschaft (DFG, German Research Foundation)-TRR 288-422213477 (Project A05). M.C. and D.M. were supported by the National Science Foundation, Grant No. DMR-1808964.

References

- (1) Katsnelson, M. I.; Novoselov, K. S.; Geim, A. Chiral tunnelling and the Klein paradox in graphene. *Nature Physics* **2006**, *2*, 620–625.
- (2) Stander, N.; Huard, B.; Goldhaber-Gordon, D. Evidence for Klein Tunneling in Graphene p-n Junctions. *Physical Review Letters* **2009**, *102*, 026807.
- (3) Young, A. F.; Kim, P. Quantum interference and Klein tunnelling in graphene heterojunctions. *Nature Physics* **2009**, *5*, 222–226.
- (4) Allain, P. E.; Fuchs, J. N. Klein tunneling in graphene: optics with massless electrons. *The European Physical Journal B* **2011**, *83*, 301–317.
- (5) Cheianov, V. V.; Fal'ko, V.; Altshuler, B. L. The focusing of electron flow and a Veselago lens in graphene p-n junctions. *Science* **2007**, *315*, 1252.
- (6) Chen, S.; Han, Z.; Elahi, M. M.; Habib, K. M. M.; Wang, L.; Wen, B.; Gao, Y.; Taniguchi, T.; Watanabe, K.; Hone, J.; Ghosh, A. W.; Dean, C. R. Electron optics with p-n junctions in ballistic graphene. *Science* **2016**, *353*, 1522–1525.

- (7) Mishchenko, E. G.; Shytov, A. V.; Silvestrov, P. G. Guided Plasmons in Graphene p-n Junctions. *Physical Review Letters* **2010**, *104*, 156806.
- (8) Williams, J. R.; Marcus, C. M. Snake States along Graphene p-n Junctions. *Physical Review Letters* **2011**, *107*, 046602.
- (9) Park, C.; Yang, L.; Son, Y.; Cohen, M.; Louie, S. Anisotropic behaviours of massless Dirac fermions in graphene under periodic potentials. *Nature Physics* **2008**, *4*, 213–217.
- (10) Li, Y.; Dietrich, S.; Forsythe, C.; Taniguchi, T.; Watanabe, K.; Moon, P.; Dean, C. R. Anisotropic band flattening in graphene with one-dimensional superlattices. *Nature Nanotechnology* **2021**, *16*, 525–530.
- (11) Vakil, A.; Engheta, N. Transformation Optics Using Graphene. *Science* **2011**, *332*, 1291–1294.
- (12) Park, C.-H.; Son, Y.-W.; Yang, L.; Cohen, M. L.; Louie, S. G. Electron Beam Supercollimation in Graphene Superlattices. *Nano Letters* **2008**, *8*, 2920–2924.
- (13) Sajjad, R. N.; Ghosh, A. W. High efficiency switching using graphene based electron “optics”. *Applied Physics Letters* **2011**, *99*, 123101.
- (14) Péterfalvi, C. G.; Oroszlány, L.; Lambert, C. J.; Cserti, J. Intraband electron focusing in bilayer graphene. *New Journal of Physics* **2012**, *14*, 063028.
- (15) Jang, M. S.; Kim, H.; Son, Y.-W.; Atwater, H. A.; Goddard, W. A. Graphene field effect transistor without an energy gap. *Proceedings of the National Academy of Sciences* **2013**, *110*, 8786–8789.
- (16) Lee, G.-H.; Park, G.-H.; Lee, H.-J. Observation of negative refraction of Dirac fermions in graphene. *Nature Physics* **2015**, *11*, 925–929.
- (17) Zhou, X.; Kerelsky, A.; Elahi, M. M.; Wang, D.; Habib, K. M. M.; Sajjad, R. N.; Agnihotri, P.; Lee, J. U.; Ghosh, A. W.; Ross, F. M.; Pasupathy, A. N. Atomic-Scale

- Characterization of Graphene p–n Junctions for Electron-Optical Applications. *ACS Nano* **2019**, *13*, 2558.
- (18) Wang, Y. et al. Modulation Doping via a Two-Dimensional Atomic Crystalline Acceptor. *Nano Letters* **2020**, *20*, 8446.
- (19) Zhou, B.; Balgley, J.; Lampen-Kelley, P.; Yan, J.-Q.; Mandrus, D. G.; Henriksen, E. A. Evidence for charge transfer and proximate magnetism in graphene– α -RuCl₃ heterostructures. *Physical Review B* **2019**, *100*, 165426.
- (20) Mashhadi, S.; Kim, Y.; Kim, J.; Weber, D.; Taniguchi, T.; Watanabe, K.; Park, N.; Lotsch, B.; Smet, J. H.; Burghard, M.; Kern, K. Spin-Split Band Hybridization in Graphene Proximitized with α -RuCl₃ Nanosheets. *Nano Letters* **2019**, *19*, 4659–4665.
- (21) Gerber, E.; Yao, Y.; Arias, T. A.; Kim, E.-A. Ab Initio Mismatched Interface Theory of Graphene on α -RuCl₃: Doping and Magnetism. *Physical Review Letters* **2020**, *124*, 106804.
- (22) Biswas, S.; Li, Y.; Winter, S. M.; Knolle, J.; Valenti, R. Electronic Properties of α -RuCl₃ in Proximity to Graphene. *Physical Review Letters* **2019**, *123*, 237201.
- (23) Rizzo, D. J. et al. Charge-Transfer Plasmon Polaritons at Graphene/ α -RuCl₃ Interfaces. *Nano Letters* **2020**, *20*, 8438–8445.
- (24) Dingle, R.; Störmer, H. L.; Gossard, A. C.; Wiegmann, W. Electron mobilities in modulation-doped semiconductor heterojunction superlattices. *Applied Physics Letters* **1978**, *33*, 665–667.
- (25) *See Supporting Information*
- (26) Wang, K.; Elahi, M. M.; Wang, L.; Habib, K. M. M.; Taniguchi, T.; Watanabe, K.; Hone, J.; Ghosh, A. W.; Lee, G.-H.; Kim, P. Graphene transistor based on tunable Dirac fermion optics. *Proceedings of the National Academy of Sciences* **2019**, *116*, 6575–6579.

- (27) Chandni, U.; Henriksen, E. A.; Eisenstein, J. P. Transport in indium-decorated graphene. *Physical Review B* **2015**, *91*, 245402.
- (28) Cheianov, V.; Fal'ko, V. Selective transmission of Dirac electrons and ballistic magnetoresistance of n - p junctions in graphene. *Physical Review B* **2006**, *74*, 041403.
- (29) Low, T.; Hong, S.; Appenzeller, J.; Datta, S.; Lundstrom, M. S. Conductance Asymmetry of Graphene p - n Junction. *IEEE Transactions on Electron Devices* **2009**, *56*, 1292–1299.
- (30) Zhang, L.; Fogler, M. Nonlinear Screening and Ballistic Transport in a Graphene p - n Junction. *Physical Review Letters* **2008**, *100*, 116804.
- (31) Fogler, M. M.; Novikov, D. S.; Glazman, L. I.; Shklovskii, B. I. Effect of disorder on a graphene p - n junction. *Physical Review B* **2008**, *77*, 075420.
- (32) Huard, B.; Sulpizio, J.; Stander, N.; Todd, K.; Yang, B.; Goldhaber-Gordon, D. Transport Measurements Across a Tunable Potential Barrier in Graphene. *Physical Review Letters* **2007**, *98*, 236803.
- (33) Sajjad, R. N.; Ghosh, A. W. Manipulating Chiral Transmission by Gate Geometry: Switching in Graphene with Transmission Gaps. *ACS Nano* **2013**, *7*, 9808–9813.
- (34) Zhang, Y.; Brar, V. W.; Wang, F.; Girit, c. O.; Yayan, Y.; Panlasigui, M.; Zettl, A.; Crommie, M. F. Giant phonon-induced conductance in scanning tunnelling spectroscopy of gate-tunable graphene. *Nature Physics* **2008**, *4*, 627–630.
- (35) Zhang, Y.; Brar, V. W.; Girit, c. O.; Zettl, A.; Crommie, M. F. Origin of spatial charge inhomogeneity in graphene. *Nature Physics* **2009**, *5*, 722–726.
- (36) Elahi, M. M.; Masum Habib, K. M.; Wang, K.; Lee, G.-H.; Kim, P.; Ghosh, A. W. Impact of geometry and non-idealities on electron “optics” based graphene p - n junction devices. *Applied Physics Letters* **2019**, *114*, 013507.

- (37) Britnell, L.; Gorbachev, R. V.; Jalil, R.; Belle, B. D.; Schedin, F.; Katsnelson, M. I.; Eaves, L.; Morozov, S. V.; Mayorov, A. S.; Peres, N. M. R.; Castro Neto, A. H.; Leist, J.; Geim, A. K.; Ponomarenko, L. A.; Novoselov, K. S. Electron Tunneling through Ultrathin Boron Nitride Crystalline Barriers. *Nano Letters* **2012**, *12*, 1707–1710.