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Transport Gap Opening and High On-Off Current Ratio in Trilayer Graphene with Self-Aligned Nanodomain Boundaries

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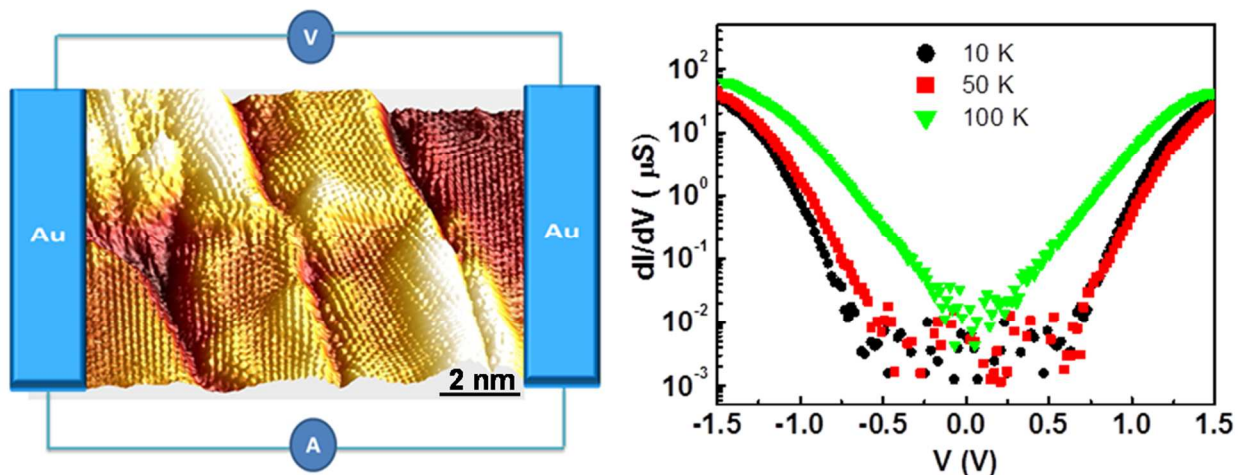
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Abstract



Trilayer graphene exhibits exceptional electronic properties that are of interest both for fundamental science and for technological applications. The ability to achieve a high on-off current ratio is the central question in this field. Here, we propose a simple method to achieve a current on-off ratio of 10^4 by opening a transport gap in Bernal stacked trilayer graphene. We synthesized Bernal stacked trilayer graphene with self-aligned periodic nanodomain boundaries (NBs) on the technologically relevant vicinal cubic-SiC(001) substrate and performed electrical measurements. Our low temperature transport measurements clearly demonstrate that the self-aligned periodic NBs can induce a charge transport gap greater than 1.3 eV. More remarkably, the transport gap of ~ 0.4 eV persists even at 100 K. Our results show the feasibility of creating new electronic nanostructures with high on-off current ratios using graphene on cubic-SiC.

KEYWORDS: trilayer graphene; nanodomain boundary; transport gap; scanning tunneling microscopy; ARPES

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3 Graphene is a single-atom-thick carbon sheet with extraordinary properties unrivalled by any
4 other known material,¹⁻⁷ which will likely lead to a revolution in many areas of technology.⁸ It
5 displays linear band dispersion,⁹⁻¹⁰ massless Dirac fermions,¹¹ and extremely high mobility.¹²
6
7 Potentially, graphene-based electronics could consist of just one or a few layers of graphene;
8 however, the absence of a band gap presents a conundrum for the implementation of
9 conventional device architectures, similar to those based on semiconducting materials.¹³⁻¹⁸
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11 Several methods have been proposed for opening band or transport gaps in graphene, such as
12 patterning single-layer graphene into narrow ribbons,¹⁹ introducing nanoholes into the graphene
13 sheets,²⁰ applying a perpendicular electric field,^{13-18,21-24} or applying mechanical strain.^{25,26}
14
15 Unlike bilayer graphene, gap-opening in trilayer graphene depends on the stacking order of the
16 layers, and notably for ABA (Bernal) stacking it remains metallic even in the presence of a
17 perpendicular electric field.^{2,21-24} Generally, the fabrication, and thus the stacking order, of
18 trilayer graphene devices relies on the mechanical exfoliation of graphite crystals.¹ Although
19 mechanical exfoliation of graphene from graphite is an effective and successful sample
20 preparation method for fundamental research, it is found that roughly 60% of trilayer samples
21 prepared this way have a pure ABA stacking order, while the remainder exhibit mixed ABA-
22 ABC stacking orders. Alternatively, chemical vapor deposition,^{27,28} and vacuum synthesis on
23 silicon carbide surfaces²⁹⁻³² are excellent ways to fabricate large-area few-layer and monolayer
24 graphene. It is known that the graphene produced by these techniques typically contains
25 domain boundaries,³⁰⁻³⁵ which can considerably modify the electronic transport in the
26 graphene.^{35,36} Recent theoretical investigations show that for graphene with self-aligned
27 nanodomain boundaries (NBs), depending on the structure of the NBs, there are two distinct
28 transport behaviors: either high transparency or perfect reflection of charge carriers over large
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3 energy ranges.³⁶ Although this would provide a new way to control the charge carriers without
4 the need to introduce bulk band gaps, there has been no direct experimental evidence so far. The
5 main challenge is to produce graphene with self-aligned periodic NBs.
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11 In this paper, we propose a simple method to synthesize graphene with self-aligned periodic NBs
12 on a semiconducting substrate compatible with silicon technologies and give a clear
13 demonstration of the existence of a transport gap. Our ABA-stacked trilayer graphene with self-
14 aligned periodic NBs was synthesized on SiC(001) thin films grown on vicinal Si(001) wafers.
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16 Our transport measurements clearly demonstrate that the self-aligned periodic NBs induce a
17 charge transport gap which can reflect charge carriers over a remarkably wide range of energies
18 (0.4– 1.3 eV). Moreover, a high on-off current ratio of 10^4 was achieved with a voltage of 0.7 V
19 below 50 K and with a voltage of 0.25 V at 100 K. Our studies pave a way to tailor the transport
20 properties of trilayer graphene.
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32 33 **RESULTS AND DISCUSSION**

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35 For this study, uniform trilayer graphene with a preferential NB direction was fabricated on
36 vicinal SiC(001)/Si(001) wafers using Si-atom sublimation followed by high temperature surface
37 graphitization in ultra-high vacuum (UHV).³⁰⁻³² The step direction on the substrate was close to
38 [110], as shown in Figure 1. Raman spectroscopy, scanning tunneling microscopy (STM), low
39 energy electron microscopy (LEEM), low energy electron diffraction (LEED), and angle
40 resolved photoelectron spectroscopy (ARPES) were used to characterize the synthesized trilayer
41 graphene. Figure S1 shows the typical Raman spectrum of graphene on the vicinal SiC substrate
42 measured using a wavelength of 532 nm. The G' or 2D band can be fitted well with 6
43 Lorentzians with full widths at half maximum (FWHM) ranging from 21–24.5 cm^{-1} , indicating
44 that the graphene grown is trilayer graphene with NBs present, which is also consistent with the
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3 electron reflectivity curves measured in different areas of the graphene/SiC(001) sample (Figure
4 S2b). More details about the Raman characterization can be found in the Supporting Information.
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8 Figures 2a and 2b show large area STM images of trilayer graphene on the vicinal SiC(001)
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10 substrate. Interestingly, our synthesized graphene contains nanometer-scale domains with
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12 boundaries elongated in one direction, which is very close to the step direction of the vicinal
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14 SiC(001) sample before graphene synthesis (Figure 1). More remarkably, those nanometer-scale
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16 domain structures are continuous even at the intrinsic surface defects (micrometer-sized domain
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18 boundaries) as observed in Figure 2a. Therefore, we present a simple method to synthesize
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20 graphene with self-aligned periodic NBs. However, LEED characterization suggests that the
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22 periodic structures at the nanodomain boundaries between domains are not uniform and straight
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24 over the entire sample. The reason for this is the great difficulty in preparing uniform and straight
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26 steps for the entire SiC substrate.
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33 Figure 2c shows an atomically resolved STM image containing several nanometer-scale domains
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35 connected to each other through the NBs. Detailed analysis of the high resolution STM images
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37 measured near the NBs shows that, in most cases, NBs on the vicinal sample (Figures 2c and S3)
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39 are rotated by 3.5° relative to the one of the $\langle 110 \rangle$ directions as depicted in Figures 2e and S4.
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42 The mechanism responsible this arrangement can be understood as follows. There is a zigzag
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44 structure on the left hand side and an armchair structure on right hand side of the NBs (Figure
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46 S4a) and the angle on the left side between Gr_L and the substrate SiC [110] and the angle on the
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48 right side between Gr_R and SiC [110] are both 13.5° . This implies that carbon atoms deposited on
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50 top of the SiC have equal probability of developing zigzag or armchair structures. Considering
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52 the complicated structure near NBs, there are three different graphene structures present:
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54 pentagonal, hexagonal, and heptagonal. The internal angle of a pentagon is 108° , a hexagon is
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120°, and a heptagon is 128.57°. When a pentagon grows next to a hexagon the angle difference is 12° smaller; while for a heptagon grows next to a hexagon the angle is 8.57° more. Therefore NBs on top of the SiC deviate from the symmetry of the line by 3.43°, which is consistent with the experimental result of 3.5°. For the left hand side of the NB, a zigzag structure is developed from the NB. This means that two hexagons transform into one pentagon and one heptagon, so the angle difference is $(8.57^\circ+12^\circ)/2=10.28^\circ$. For the right hand side of the NB, the armchair structure developed means hexagons must merge with two sides of a pentagon, so the total angle change is $8.57^\circ\times 2=17.14^\circ$. As Figure 2e illustrates, this asymmetry near the NBs leads to the formation of a periodic structure along the boundaries, with a period of 1.37 nm. The periodic structure consists of distorted heptagons and pentagons (Figure 2e), which produce the modulations in the atomically resolved STM image measured at the NB (Figure 2d).

In order to extract information about the electronic structure and the stacking order of the nanostructured trilayer graphene synthesized on cubic-SiC(001), we performed ARPES measurements (Figures 3 and S5) and analysed the fine structure of the multiple bands seen near the \overline{K}_A and \overline{K}_B points in Figure 3c. Additionally to gain a better understanding of the experimental data, we performed simulations of photoemission from samples hosting two rotational domains of the trilayer, accounting for the observed momentum splitting $\Delta k=0.12 \text{ \AA}^{-1}$. Simulations were performed for the band structure of graphene corresponding to Bernal or Rhombohedral stacking. The band structure of the ABA- and ABC-stacked trilayer graphene acquired within the tight-binding (TB) model was taken from the literature.³⁷ The section of trilayer band structure along the direction perpendicular to the $\overline{\Gamma}-\overline{K}$ line of the surface Brillouin zone, which corresponds to the acquisition direction of the experimental dispersion shown in Figure 3c, was simulated by an array of energy distribution curve (EDC) slices composed of the

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3 Lorentzian peaks residing at the energy positions of the trilayer bands. As ARPES is a non-local
4 method, it sums up photoemission signals from all the various structural phases of graphene at
5 the sample surface. Therefore, the rotational variants of the graphene domains and their
6 rotational displacements were considered in a framework of rotated Brillouin zones and
7 approximated in simulation set by corresponding multiplication and displacement of the model
8 band structure along the momentum axis K_{\parallel} . This approach is illustrated in Figure S6. In
9 particular, photoemission from two principal rotational variants of the trilayer graphene on
10 SiC(001) were constructed through the superposition of two identical TB band structures of the
11 trilayer displaced by $\pm 0.06 \text{ \AA}^{-1}$ relative to the \bar{K} -point. We assume that intensity of
12 photoemission from the trilayer's bands is proportional to the density of states (DOS) of
13 graphene. Therefore, it was approximated for each trilayer band by a linear function of the
14 binding energy. In fact, there is no noticeable impact of this factor on the results of the
15 simulation (*e.g.* qualitatively similar results were obtained also for a constant DOS). Conversely,
16 the crucially important factor for agreement or disagreement between the experiment and
17 simulations is the width of the Lorentzian peaks composing EDC slices. In our simulation we
18 varied the FWHM of the Lorentzians between 100 meV and 800 meV to test the correlation
19 between the measured (Figure 3c) and simulated dispersions over energy. Very good correlation
20 between measured ARPES dispersions (Figure 3c) and simulations (Figure 3d) was achieved for
21 a FWHM of the π -bands as large as 600 meV and for an initial band structure corresponding to
22 the ABA-trilayer. Thus it can be concluded that the stacking order of the trilayer graphene on the
23 cubic-SiC(001) is indeed of ABA-type. Moreover, the FWHM=600 meV is three times larger
24 than FWHM measured by ARPES for high-quality quasi-freestanding single layer graphene on
25 hexagonal SiC, Ir or Au.³⁸⁻⁴⁰ Such significantly enhanced broadening can be ascribed either to
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3 geometric contributions from a minority of rotational variants distinct from the nanodomains
4 rotated by $\pm 13.5^\circ$ from the $\langle 110 \rangle$ directions or to quantum scattering of quasiparticles on
5 structural imperfections. Both origins of the broadening are associated with the large number of
6 rotational NBs. The ARPES measurements conducted on the graphene/SiC(001) samples also
7 reveal that the Dirac points are very close to the Fermi level (Figures 3c and S5). This is in full
8 agreement with theoretical simulations for ideal trilayer graphene (Figures 3d and S6) which
9 assume a negligibly small doping level is present in our trilayer graphene.

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11 Figure 4a shows a schematic drawing of a typical graphene nano-gap device. Devices with sub-
12 30 nm nano-gap contacts were fabricated using standard electron beam lithography techniques.
13 Ti/Au (5/45 nm) electrodes were deposited by electron-beam evaporation. The bias voltage was
14 applied perpendicular to the NBs to measure the local transport properties due to the NB defects.

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16 Figure 4b shows the temperature dependent resistance (R-T) measured with a bias voltage of 0.5
17 V. The resistance is around 1000 Ω at room temperature and increases significantly with
18 decreasing temperature. Moreover, below 100 K, we cannot detect an appreciable current signal
19 for a bias voltage of 0.5 V, which clearly demonstrates a transport gap in the synthesized trilayer
20 graphene. We fitted the R-T curve with $R(T) \approx R_0 \exp\left(\frac{E_a}{K_B T}\right)$, where R_0 is the fitting parameter,
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43 K_B is the Boltzmann constant and E_a is the activation energy. The fitting gives an activation
44 energy of 130 meV. To further demonstrate the existence of the transport gap, we measured I-V
45 at 10 K (Figure 4c) and plotted the corresponding dI/dV in Figure 4d. Interestingly, for a small
46 bias voltage, we cannot detect any reasonable current signal and the corresponding dI/dV is
47 around 0.01 μS , indicating the existence of a transport gap. The transport gap derived from the
48 dI/dV plot is approximately 1.3 eV at 10 K. To understand the effect of the NBs on the electron
49 transport properties, we simulated current across the NBs as a function of gate voltage. In our
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3 calculations, we used the unit cell as shown in Figure 5a and the gate voltage is adjusted through
4 the on-site energy. A Nonequilibrium Green Function (NEGF) is used and the current is
5 calculated with Landauer-Keldysh formalism.^{41,42} Details about the simulation can be found in
6 the Methods section. Our simulations are summarized in Figure 5b. There is a plateau region
7 where electrons cannot travel, indicating that a gap does open with current driven across the self-
8 aligned NBs and can be tuned with the gate voltage.
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Figures 6a and 6b show temperature dependent I-V curves. The transport gap can be clearly observed below 100 K but disappears at temperatures above 150 K. To obtain the exact value of the transport gap, we plotted the corresponding dI/dV curves in Figure 5c for temperatures below 150 K. Remarkably, the transport gap is approximately the same at 50 K and 10 K but substantially lower (0.4 eV) at 100 K. Applying a bias voltage smaller than the transport gap the conductivity of the device is only 10⁻² μS, but this increases to 10² μS when the bias voltage is larger than the transport gap, which gives a high on-off current ratio of 10⁴. Moreover, in our nano-gap contact devices, the NBs are uniform and directed along the step direction on the vicinal substrate (Figure 4a), which gives this system the potential for high-density memory applications. We would like to point out that it was suggested by Huang *et al* that the scattering rate for a periodic boundary can be written as

$$\langle W(k) \rangle = \frac{S^2 m_x}{2\pi^2 \hbar^3 |k_x| d} \times \left[\frac{1 - \exp(-4k_x^2 \sigma^2)}{1 + \exp(-4k_x^2 \sigma^2) - 2 \exp(-2k_x^2 \sigma^2) \cos(2k_x d)} \right],$$

where d is the periodicity along the boundary. Thus, attaining a larger on-off current ratio may result in a reduction in the mobility.

The observed temperature dependence of transport gap could be attributed to two possible mechanisms.^{25,26,36} First, it was suggested by Yazyev and Louie³⁶ that a charge transport gap of

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4 $E_g = \hbar v_F \frac{2\pi}{3d} \approx \frac{1.38}{d(\text{nm})}$ (eV) can be formed by a non-symmetric NB associated with a lattice

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7 mismatch at the boundary line, where \hbar is the reduced Planck's constant, v_F is the Fermi velocity.

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10 As indicated in Figures 2d and 2e, the asymmetric rotation of the graphene lattices in the
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12 neighbouring domains relative to the NB leads to a 1.37 nm periodicity along the NB. According
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14 to the theory,³⁶ this periodicity should produce a transport gap of approximately 1.0 eV, which is
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16 consistent with our transport measurements. Therefore, our electrical and STM characterizations
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18 support the explanation of the transport gap being based on the Yazyev and Louie theory of
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20 asymmetrically rotated domains³⁶ for the Bernal-stacked trilayer graphene with the NBs oriented
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22 close to the [110] direction (Figure 2e). The disappearance of the transport gap at temperatures
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24 above 150 K in this case can be related to the presence of defects in the graphene trilayer. It is
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26 known that defects can remarkably modify the transport properties of graphite and graphene.⁴³
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28 There are two kinds of defects in our trilayer graphene: defects at the NBs and interstitial defects
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30 in subsurface layers. In STM experiments (Figures 2c and 2d) and Raman characterization
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32 (Figure S1), we found that defects at the NBs are stable even at room temperature. Thus they
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34 should not have a pronounced impact on the temperature dependence of the trilayer graphene's
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36 conductivity (Figures 4 and 6). However, the stability of interstitial defects in subsurface layers
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38 (Figure 6d) is temperature dependent. In graphite, the migration activation energy of interstitials,
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40 which is usually below 0.03 eV,⁴⁴⁻⁴⁷ depends on the presence of domain boundaries and the
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42 stress in the layer. In our trilayer graphene on vicinal SiC, defects and interstitials may exist
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44 which may be frozen under 100 K and migrate at higher temperatures (100-300 K).⁴⁴ The
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46 mobility of such interstitial defects could be responsible for the disappearance of the transport
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48 gap at temperatures above 100 K (Figure 6).
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3 Another possible origin of the transport gap observed in trilayer graphene on vicinal SiC(001) at
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5 temperatures below 150 K could be related to mechanical strain applied to the graphene
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7 nanoribbons during the sample cooling. It was shown that applying a uniaxial mechanical strain
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9 to bilayer graphene²⁶ or graphene nanoribbon²⁵ can provide a band gap of the order of 0.2-0.4 eV
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11 which is also in agreement with the electrical measurements (Figure 6). For our graphene grown
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13 on vicinal SiC, to relieve the in-plane compressive strain, ripples can be formed at asymmetric
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15 NB.³⁶ Indeed, Figures 2c, 2d and S3 show that the boundaries of all the individual domains on
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17 the SiC(001) do possess such a rippled morphology. As shown in our previous work³⁰, the
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19 graphene is quasi-freestanding on the SiC, with only some parts of the layers (usually, NBs)
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21 supported by the substrate. The thermal expansion coefficients and their temperature dependence
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23 are remarkably different for SiC⁴⁸ and both micrometer-sized and nanostructured graphene,⁴⁹⁻⁵¹
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25 which can induce in-plane strain and lead to the ripple formation during sample cooling after
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27 high temperature graphene synthesis.
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34 To clarify the origin of the charge transport gap, we measured I-V at 10 K with the current
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36 applied along the boundaries (Figure 7a). No transport gap is observed and the I-V curve
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38 displays nonlinear behavior. This indicates that the observed charge transport gap for current
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40 across the NBs is mainly due to the self-aligned periodic NBs which can reflect charge carriers
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42 over a range of energies. It also rules out the effect of the contact resistance between graphene
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44 and electrode. In addition to this, we also conducted photoemission experiments on the vicinal
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46 SiC(001) samples at temperatures between 100 K and 300 K (Figure 7b). Our experimental data
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48 undoubtedly show that the valence band edge position does not change with decreasing
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50 temperature proving that the observed transport gap is not related to a band gap opening during
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52 the sample cooling. At the same time, the secondary electron cutoff (SEC) measurements reveal
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3 a change of the work function between 150 K and 100 K of ~ 0.36 eV, *i.e.*, the values observed in
4 the transport measurements (Figure 6c). Thus, both I-V characterization and low temperature
5 photoemission measurements lead us to conclude that the observed charge transport gap most
6 probably should be attributed to the self-aligned periodic NBs.
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12 13 **CONCLUSION**

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16 In summary, we have presented transport studies on trilayer graphene synthesized on vicinal
17 cubic-SiC(001) substrates. Our electrical measurements clearly demonstrate the opening of a
18 transport gap in the nanostructured graphene synthesized on this stepped surface. Remarkably,
19 the effect can be observed even at 100 K and produces a current on-off ratio of 10^4 . This
20 transport gap was notably observed in the Bernal stacked trilayer graphene. This behavior shows
21 that it is possible to create new tunable electronic nanostructures with graphene on cubic SiC,
22 thus creating opportunities for a wide range of new electronic applications.
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36 **Methods**

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38 Uniform trilayer graphene was fabricated on SiC(001) thin films, grown on vicinal (2° miscut)
39 Si(001) wafers, using Si-atom sublimation followed by high temperature surface graphitization
40 in the UHV preparation chamber of a room temperature (RT) GPI-300 scanning tunneling
41 microscope.^{30,31} The base pressure in the analytical chamber was in the range of $4\text{--}6 \times 10^{-11}$ mbar.
42 It did not exceed 2×10^{-10} mbar during the direct current heating of the $3 \times 8 \times 0.5$ mm³
43 SiC(001)/Si(001) samples at temperatures of 1300—1350°C and rapidly recovered after the
44 thermal cycles. The graphene synthesis included an UHV deposition of several monolayers of
45 silicon atoms onto the clean, carbon-rich SiC(001)- 1×1 surface and annealing at gradually
46 increasing substrate temperatures in the range of 700—1300°C until the carbon-rich $c(2 \times 2)$
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3 reconstruction was achieved. The typical duration of each annealing step leading to the
4 consecutively assembly of the (3×2), (5×2), c(4×2), (2×1), and c(2×2) reconstructions was in the
5 range of 10-30 minutes depending on the temperature applied and the pressure in the UHV
6 chamber. Then, flash heating (10-20 seconds) at 1350°C with post-annealing at 600—700°C
7 were used to convert the c(2×2) surface reconstruction into honeycomb lattice. Full details of the
8 graphene synthesis and step-by-step STM studies of the SiC(001) surface atomic structure during
9 synthesis have been described elsewhere.³¹ The principal difference between the current
10 graphene fabrication procedure and previously reported works^{30,31} is the utilization of the vicinal
11 (stepped) SiC(001)/Si(001) substrate.
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24 The growth of few-layer graphene on cubic-SiC(001) in UHV at pressure below 2×10^{-10} mbar is
25 self-limiting at three graphene layers.^{30,31} The graphitization occurs in a multi-stage process, and
26 is extremely dependent on sample cleanliness, pressure in the vacuum chamber *etc.* For these
27 reasons, it is very difficult to obtain a uniform graphene coverage of less than 3 monolayers on a
28 millimeter-scale SiC(001) sample. Therefore, for the transport measurements we used vicinal
29 SiC(001)/Si(001) samples with uniform trilayer graphene as proved by LEEM and ARPES data
30 (Figures S2 and S5).
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41 All STM experiments were conducted at RT with a single crystalline tungsten tips cleaned and
42 sharpened in the UHV chamber.⁵² STM topographic images were processed using WSxM
43 software.⁵³ ARPES experiments were performed with an ARPES 1²endstation equipped with a
44 Scienta R8000 hemispherical electron analyzer operated at the UE112-PGM2a beamline at
45 BESSY-II. All ARPES measurements were performed at a photon energy of 62 eV with a linear
46 (mixed *s+p*) light polarization. The beam spot size on the sample was about 0.4 mm. Before
47 ARPES characterization, the trilayer graphene/SiC(001) samples were annealed in the
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3 corresponding UHV preparation chambers at a variety of temperatures ranging from 200°C to
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5 1000°C to remove possible contamination. SEC and valence band spectra of graphene
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7 synthesized on vicinal SiC(001) as a function of temperature in the range of 104—300 K were
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9 taken using the endstation of the Russian German Laboratory at BESSY-II. The LEEM and
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11 micro-LEED measurements were done using a SPELEEM microscope (Elmitec GmbH) installed
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13 on beamline I311 at the MAX-laboratory in Sweden.
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17 The device with nano-gap contacts was fabricated by electron beam lithography (EBL) using
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19 single layer positive tone resist PMMA supplied by MicroChem Corp. After development, thick
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21 metal contacts consisting of Ti (5 nm)/Au (45 nm) were deposited by e-beam evaporation.
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24 **Electron transport calculations**

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26 We used the tight-binding approximation with a nearest neighbor hopping energy t ($t=2.7\text{eV}$) to
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28 describe the graphene system,⁵⁴
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$$32$$
$$33 H = -t \sum_{i,j} C_i^\dagger C_j + \sum_i U_i C_i^\dagger C_i$$
$$34$$
$$35$$
$$36$$

37 where C_i^\dagger (C_i) creates (annihilates) an electron at site i and U_i is the on-site energy at site i .

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39 Since a defect causes very small deviations in the bond length,^{55,56} we therefore propose that all
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41 of the hopping energies are the same since all the carbon atoms are assumed to be equidistant.
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45 In the electron transport simulation, we use nonequilibrium Green function (NEGF) theory and
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47 the Landauer-Keldysh method is used to calculate the transmission and I-V behaviors for the NBs
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49 in our system.^{41,42} The transmission function between the leads is evaluated by
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$$52$$
$$53 T_{pq} = \text{Tr}[\Gamma_p G^R \Gamma_q G^A]$$
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3 where G^R (G^A) is the regular(advance) Green's function of the system. The lead broadening
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5 function $\Gamma_{p(q)} = i(\Sigma_{p(q)} - \Sigma_{p(q)}^\dagger)$, where $\Sigma_{p(q)}$ is the self-energy of the lead. The conductance
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7
8 $G(E)$ can be obtained from the transmission function by $G(E) = G_0 T(E)$, where $G_0 = \frac{2e^2}{h}$ is the
9
10 conductance quantum. Finally, the current can be obtained by $I = \frac{2e}{h} \int T(E) [f_p(E) - f_q(E)] dE$
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12
13 where $f_{p(q)}$ is Fermi distribution of each lead.
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19

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37
38 measurements, and A. Zakharov for help with LEEM/micro-LEED characterization.
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45 **ASSOCIATED CONTENT**

46
47 *Supporting Information Available:* Raman, LEEM, micro-LEED, STM, and ARPES
48
49 characterization of the graphene/SiC(001) samples. This material is available free of charge *via*
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51 the Internet at <http://pubs.acs.org>.
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Figures and captions

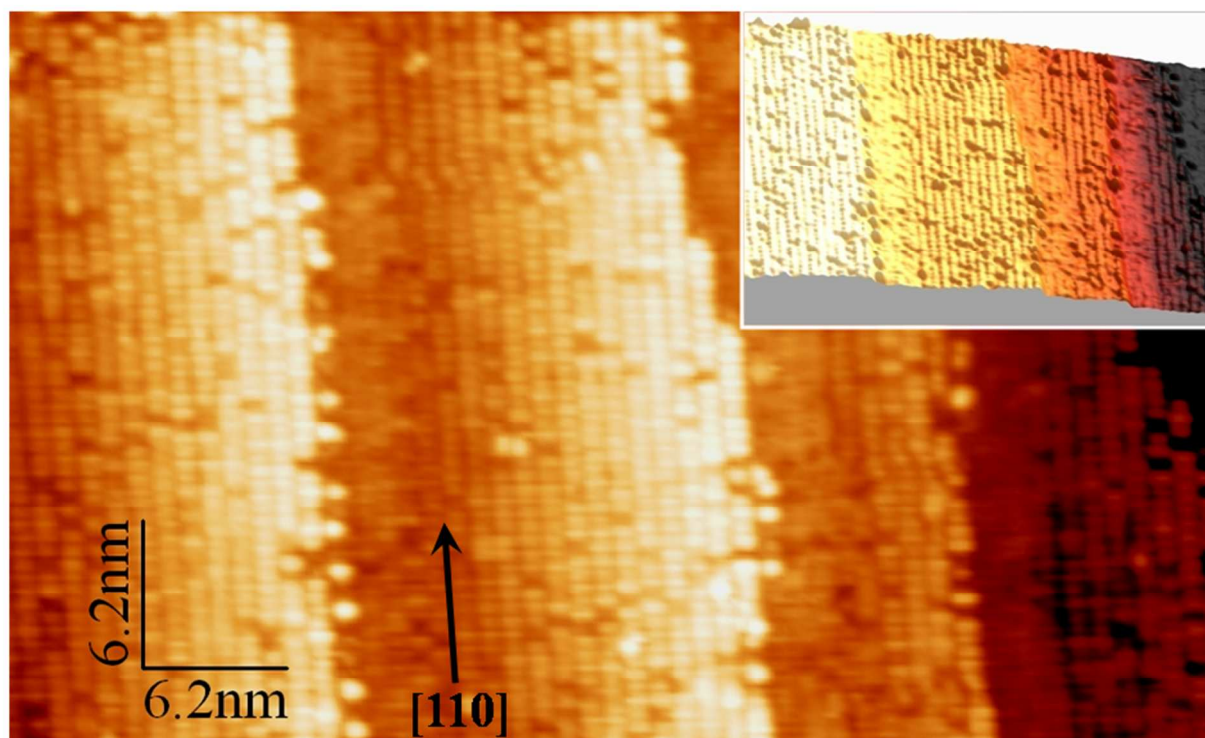


Figure 1 | STM characterization of the vicinal SiC(001). 2D- and 3D-representations (inset) of a $51 \times 31 \text{ nm}^2$ atomically resolved STM image of the SiC(001) 3×2 reconstruction. The image demonstrates that the step direction is close to the [110] direction of the SiC crystal lattice. The image was measured at $U = -2.3 \text{ V}$ and $I = 80 \text{ pA}$.

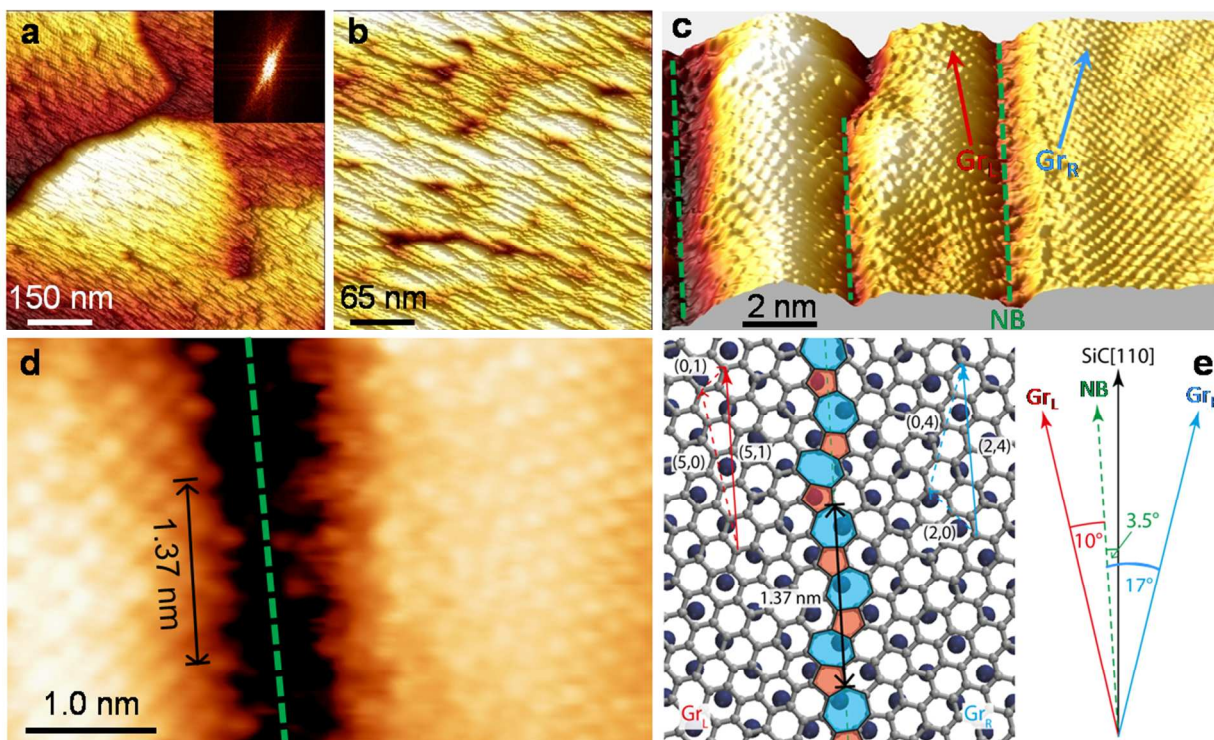


Figure 2 | STM characterization of the graphene grown on vicinal SiC(001). (a) and (b) Large area STM images of graphene nanoribbons synthesized on the vicinal SiC(001). Inset in panel (a) shows the Fast Fourier Transform of the STM image proving one preferential direction of the NBs on the vicinal sample. (c) and (d) Atomically resolved STM images of graphene nanoribbons showing the system of domains rotated 17° clockwise (Gr_R) and 10° anticlockwise (Gr_L) relative to the NB which is rotated 3.5° anticlockwise from the [110] direction (c) and the atomic structure of the NB (d). The images were measured at $U=-100$ mV, $I=68$ pA. (e) Schematic model of the NB for the asymmetrically rotated nanodomains in panels (c) and (d). For the angles shown a periodic structure of distorted pentagons and heptagons is formed.

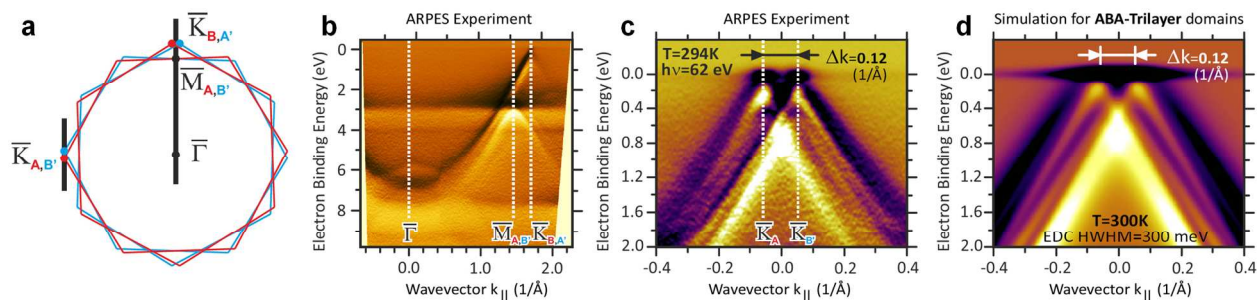


Figure 3| ARPES characterization determining the electronic structure and the Bernal stacking nature of the trilayer graphene on the vicinal SiC(001). (a) Effective surface Brillouin zone corresponding to four rotated domain variants. (b) and (c) Dispersion of the π -band in the graphene along the directions indicated in panel (a). (d) Simulations for ABA-stacked trilayer graphene with a high density of nanodomain boundaries.

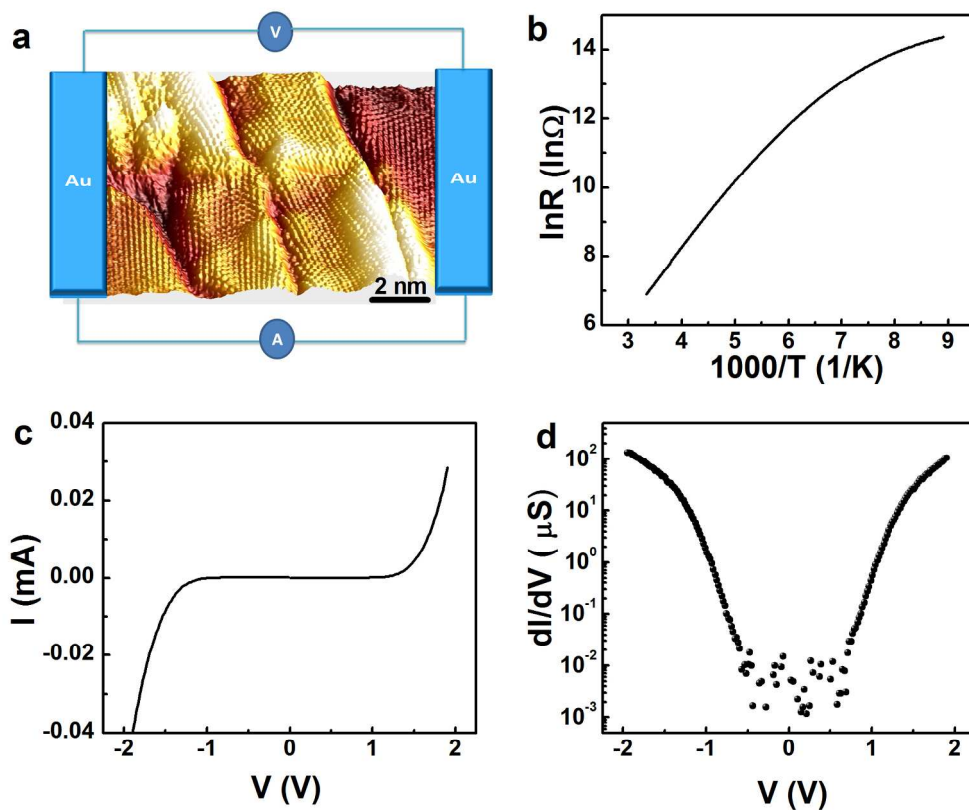


Figure 4| Electrical detection of the opening of a transport gap in Bernal stacked trilayer graphene on a vicinal SiC substrate. (a) Schematic drawing of the nano-gap device. (b) R-T curve measured with a bias voltage of 0.5 V. (c) I-V curve measured at 10 K and (d) the corresponding dI/dV curve to demonstrate the existence of a transport gap.

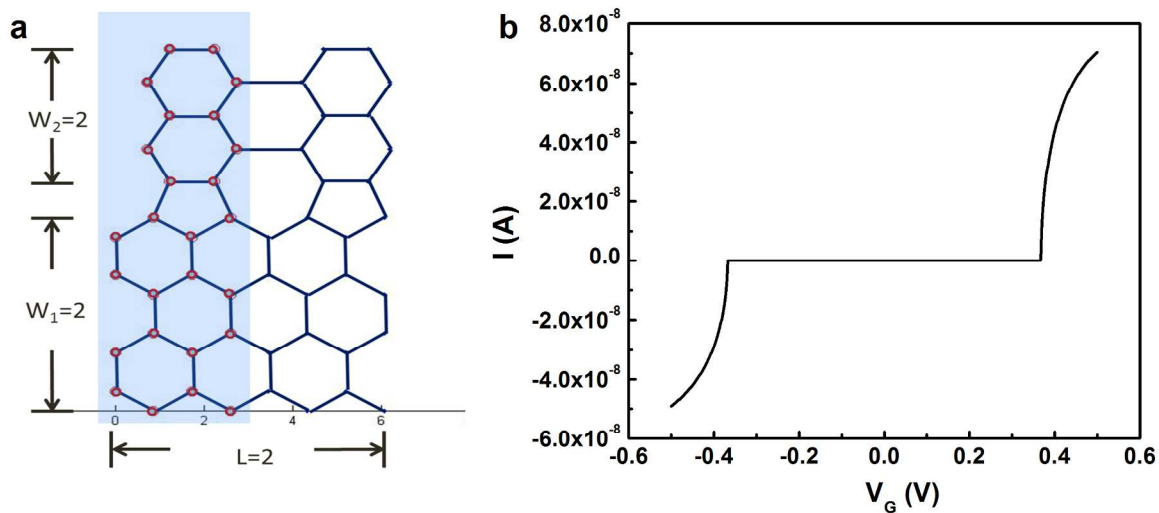


Figure 5 | Current as a function of gate voltage. (a) Schematic drawing of the model used, where L is the length of the NB, W_1 is the width of armchair structure and W_2 is the width of zigzag structure. (b) Current as a function of back voltage calculated from first-principle simulation.

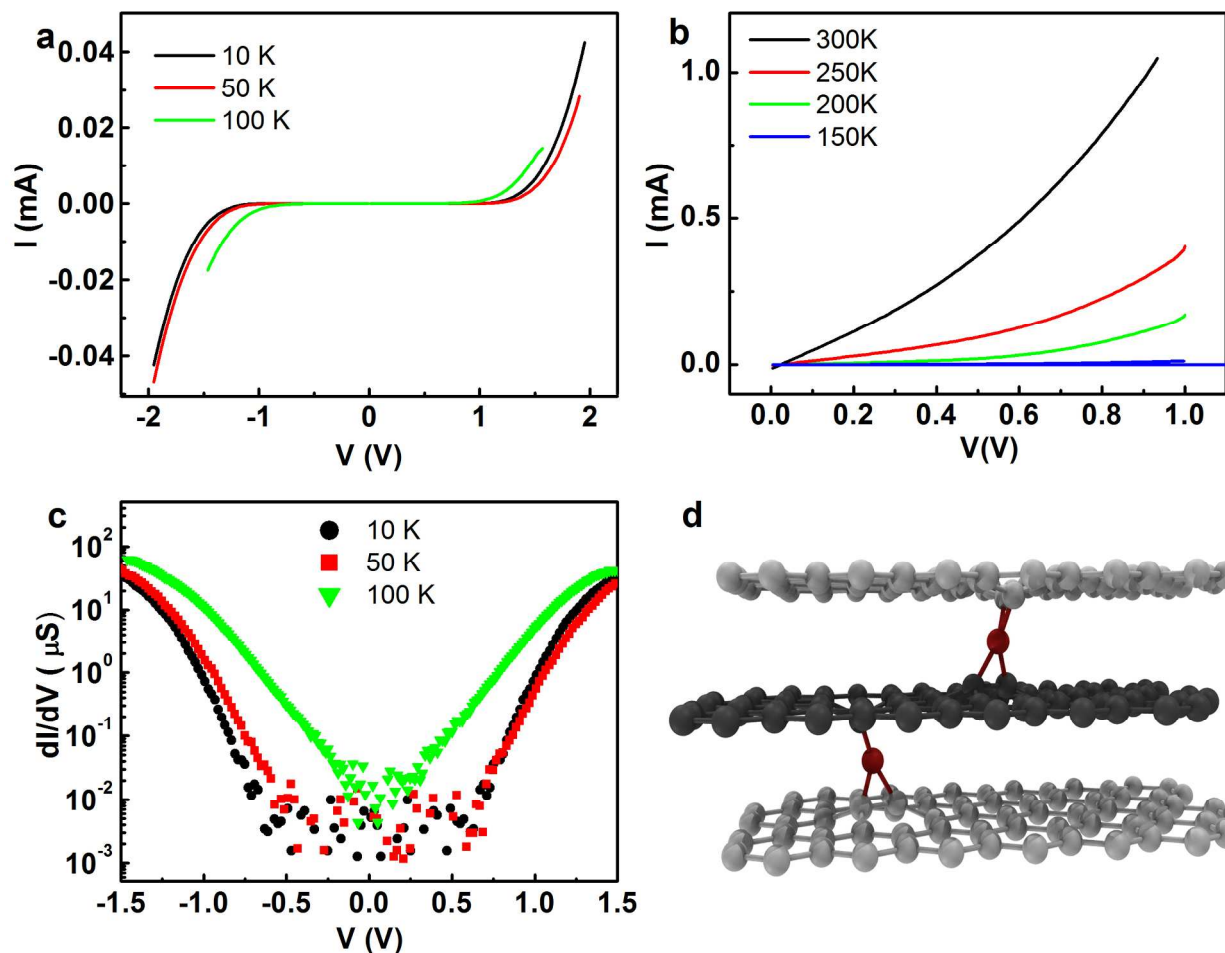


Figure 6 | I-V curves measured at different temperatures with current across the self-aligned NBs. (a) I-V curves measured at 10 K, 50 K, and 100K. (b) I-V curves measured at 150 K, 200 K, 250 K and 300K. (c) Corresponding dI/dV curves for temperatures below 150 K. (d) Schematic drawing of interstitial defects in trilayer graphene.

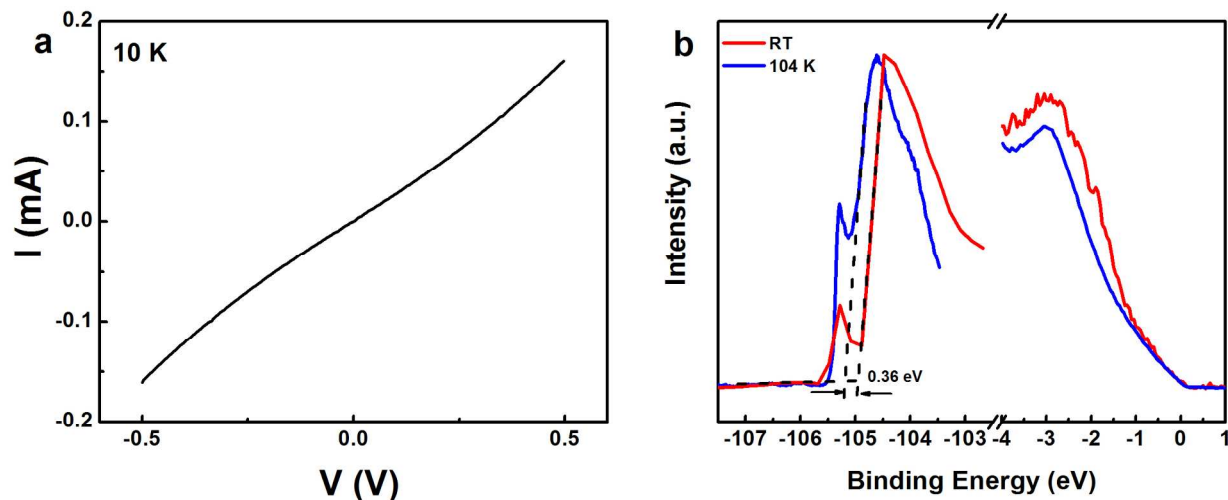


Figure 7 | I-V curve measured with current applied along the NBs and photoemission measurements at different temperatures. (a) I-V curve measured at 10 K along NBs. (b) SEC and valence band edge taken at 104 and 300 K at a photon energy of 110 eV. The sample was biased at -18.2 and -8.42 V at 104 and 300 K, respectively.