Standardization of surface potential measurements of graphene domains

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We compare the three most commonly used scanning probe techniques to obtain a reliable value of the work function in graphene domains of different thickness. The surface potential (SP) of graphene is directly measured in Hall bar geometry via a combination of electrical functional microscopy and spectroscopy techniques, which enables calibrated work function measurements of graphene domains in ambient conditions with values $\Phi_{1LG} \approx 4.55 \pm 0.02$ eV and $\Phi_{2LG} \approx 4.44 \pm 0.02$ eV for single- and bi-layer, respectively. We demonstrate that frequency-modulated Kelvin probe force microscopy (FM-KPFM) provides more accurate measurement of the SP than amplitude-modulated (AM)-KPFM. The discrepancy between experimental results obtained by different techniques is discussed. In addition, we use FM-KPFM for contactless measurements of the specific components of the device resistance. We show a strong non-Ohmic behavior of the electrode-graphene contact resistance and extract the graphene channel resistivity.

Mapping of the local electronic properties of graphene is necessary for control of growth parameters and for understanding device functionality. The accurate quantification of the measured values is essential in order for the properties of graphene to be reliably understood and compared.

The growth of graphene by the sublimation of Si from SiC is arguably the most advanced method for producing continuous, homogeneous large area graphene1. Control over the layer thickness has been demonstrated2, with sublimation being the method of choice for device manufacture, where a continuous, large area of single-layer (1LG) of graphene is required, i.e. nanoelectronics, sensing, THz applications, etc. Due to advances in the sample growth, it is generally possible to achieve homogenous single layer coverage over large areas (i.e. ~95% 1LG coverage for the sample presented in this work). However, even small inclusions of bi-layer graphene (2LG) leads to redistribution of carriers, inhomogeneous screening effects, and the corresponding nanoscale changes in the surface potential (SP) and work function ($\Phi$). Unambiguous determination of the layer thickness of epitaxially grown graphene using atomic force microscopy (AFM) is particularly challenging due to the stepped nature of the SiC substrate coupled with growth of graphene, which often nucleates at step edges1.

Scanning measurement techniques, such as Kelvin probe force microscopy (KPFM), are widely used for mapping the SP of graphene as well as identification of graphene layers. For example, KPFM has recently been used to distinguish between areas of 1LG, 2LG, few layer graphene (FLG) and the buffer or interfacial layer (0LG) and for validation of optical quality control methods for graphene4,5. However, KPFM does not generally provide reliably comparable values for differences in SP between layers with a wide variety of $\Delta V_{\text{KPFM}}$ values previously reported for 1-2LG. For example, for epitaxial graphene on SiC, Filletier et al.6 reported a $\Delta V_{\text{KPFM}} \approx 120$ mV in air. On the other hand, in case of exfoliated graphene on SiO2, Yu et al.6 reported a $\Delta V_{\text{KPFM}} \approx 120$ mV after accounting for environmental effects by measuring in ambient atmosphere and dry nitrogen, whereas Ziegler et al.7 reported a smaller value of $\Delta V_{\text{KPFM}} = 68$ mV in ambient conditions.

The effects of substrate on the charge transfer to graphene and subsequent change of the SP have been discussed in depth7-10. Additionally, change in the charge carrier concentration, whether it is intentional, by electrostatic or photochemical gates11,12, or incidental, as by uncontrolled adsorbates, modifies the measured $\Delta V_{\text{KPFM}}$ values (see ref. 6). For example, specific atmospheric gating can modify the $\Delta V_{\text{KPFM}}$ of epitaxial graphene from 0 to 100 mV on changing the environment from vacuum or pure nitrogen to >1 ppm NO2 in nitrogen mixture13. Moreover, atmospheric humidity gating has been shown to increase the $\Delta V_{\text{KPFM}}$ values14,15. While the reported discrepancy in the published values of $\Delta V_{\text{KPFM}}$ can be partly attributed to different substrate and environmental gating, here we primarily address the...
We analyze the ability to obtain quantified, comparable and accurate results of single-pass frequency-modulated (FM)-KPFM, conventional dual-pass amplitude-modulated (AM)-KPFM and electrostatic force spectroscopy (EFS) by performing measurements on a graphene Hall bar device after SP calibration of the AFM probe against gold electrodes. In contrast to many experimental studies, we aim to investigate graphene devices in standard ambient conditions (rather than in vacuum or specific gas atmosphere), as such conditions are the most representative both for general research and industrial lines. We find that conventional AM-KPFM, being a force sensitive technique, suffers from a spatial averaging effect of the SP due to a significant contribution of the cantilever base and cone to the capacitive coupling, which reduces \( \Delta V_{\text{CPD}}^{1-2\text{LG}} \) and leads to incorrect values of SP measured on a biased device. In contrast, FM-KPFM is sensitive to the force gradient and measures the SP of the area directly under the probe apex, demonstrating improved spatial resolution and absence of averaging effects. We conclude that FM techniques, such as FM-KPFM and EFS, provide more accurate measurement of the SP than AM-KPFM. Using calibrated FM-KPFM, we perform precise work function measurements of 1LG and 2LG, being \( \Phi_{1\text{LG}} = 4.55 \pm 0.02 \text{ eV} \) and \( \Phi_{2\text{LG}} = 4.44 \pm 0.02 \text{ eV} \), respectively, for the sample studied here. We also perform contactless measurements of the resistance of the graphene channel and two separate electrode-graphene lead contacts.

We demonstrate that the experimental approach presented here can be successfully used for standardization of measurements of the work function and obtaining reliable quantitative parameters not only in graphene but in many other electronic materials, i.e. semiconductors, photovoltaics, etc. Representing a surface state of a material rather than its bulk property, the work function, in graphene in particular, can be strongly affected by environmental conditions. To assure accuracy of measurements, recalibration

![Figure 1](image)

**Figure 1** | Schematic diagrams of the experimental techniques. (a) AM-KPFM; (b) FM-KPFM; topography of the graphene Hall bar is superimposed with SP maps on a 3D image. Plots show characteristic profiles, i.e. SP on top and topography on bottom along the horizontal line in the center of the image (not shown). (c) Typical parabolic change of the cantilever phase shift measured by EFS during DC voltage sweep at a fixed point on 1LG. (d) UPS data showing the work function of gold, \( \Phi_{\text{Au}} = 4.82 \text{ eV} \), for four different samples.
should be done upon any significant change of ambient (e.g. humidity).

**Results**

**Surface potential measurement techniques.** Surface potential maps of a sample can be obtained using KPFM, which measures the strength of the electrostatic forces between a conductive probe and the sample. There are different methods of detecting electrostatic forces, namely: AM-KPFM, which responds to the electrostatic force at a set frequency of probe oscillation (Figure 1a); and FM-KPFM, which responds to the electrostatic force, while maintaining constant amplitude of cantilever oscillation (Figure 1b). As we show below, the choice of the measurement technique significantly affects the accuracy of surface potential measurements on micrometer scale graphene. The schematic diagrams of the used techniques are shown in Figure 1; all techniques are discussed in detail in the Method section.

**AM-KPFM: experimental results.** Figure 2a shows a topography map of the graphene device. SiC step terraces are clearly visible running at a ~60° angle to the channel. Gold contacts are seen at the left and right hand sides of the image. The image reveals that it is generally rather difficult to determine the graphene layer thickness from the topography maps. The surface potential of the electrically grounded device was mapped using AM-KPFM in ambient environment (Figure 2b). Bright areas of 2LG are clearly visible on the 1LG background, whereas darker regions correspond to etched SiC. The value of the $\Delta V_{\text{CPD}} = 50$ mV is consistently measured over all areas of the sample (Figure 2c), which is comparable to previously published results on similar samples. The SP dip which can be seen to the right of the 2LG step in Figure 2c (as well as in Figure 3b and Figure 4b) is attributed to a small patch of resist residue, which is clearly observed in the topography map (circled in red in Figure 2a).

Further to this, we study the surface potential of a biased graphene device. Bias voltages of $V_{ch} = 0, \pm 0.5, \pm 1, \pm 1.5$ and $\pm 2$ V were applied to the left gold electrode and SP maps of the device were obtained in AM-KPFM mode. Figure 2d shows the plotted SP values along the marked line (Figure 2b) going through the center of the channel and connecting the gold leads. The raw data is plotted in Figures 2c and 2d, i.e. no calibration of the probe’s work function has been performed here. As a result, the measured SP values in Figure 2d are not centered at 0 V. A significant discrepancy between applied and measured voltages is observed using AM-KPFM, i.e. the total difference in surface potential values measured on the left gold electrode, when biased with $V_{ch} = +2$ and $-2$ V, is only $\sim 2.9$ V, i.e. 27.6% less than the expected 4 V. After taking into account the work function of the probe, the values of $\Delta V_{\text{CPD}}$ between the biased gold contacts are still smaller than expected. This discrepancy in applied and measured voltages can be explained by the spatial averaging of AM-KPFM due to the long-range nature of the electrostatic forces acting on the probe and leading to substantial contributions from the probe cone and the base. These parasitic contributions can affect the measured SP, as the area under the cantilever may not be directly over the gold leads, but instead averaging the SP over the channel device leading to a lower total value. For a given device geometry and using AM-KPFM, it is expected that scanning across the channel might somewhat decrease parasitic capacitive coupling between the cantilever and the gold electrodes. However, bearing in mind that the size of the cantilever (200 $\times$ 30 $\mu$m) is considerably larger than the device channel (50 $\times$ 5 $\mu$m) and intricate device electrode geometry (up to 6 electrodes and bonding pads of a complex shape), some coupling between the cantilever and the gold electrodes and bonding pads is unavoidable in any scanning direction. Moreover,
any inhomogeneity of the device itself (i.e. the presence of graphene domains of different thickness) will contribute to the averaging effect of AM-KPFM technique. It should be noted that the maintaining of absolute uniformity of graphene thickness over the length of 200 μm remains challenging.

**FM-KPFM: experimental results.** Surface potential mapping has been further carried out using FM-KPFM on the same device. Figure 3a shows the potential map of the grounded device. Areas of 1LG and 2LG are sharply outlined and better defined compared to the measurements taken with AM-KPFM. Values of \( \Delta V_{CPD}^{1-2LG} \) are 150 mV are recorded as shown in Figure 3b. This value is consistent over the device and significantly larger than \( \Delta V_{CPD}^{1-2LG} \) obtained with AM-KPFM. The larger \( \Delta V_{CPD}^{1-2LG} \) values can be accounted for by considering the measurement technique, which uses the force gradient rather than the force and also leads to improved spatial resolution of FM-KPFM compared to AM-KPFM. Figure 3c shows a line profile of the surface potential measured along the center of the channel with \( V_{ch} = 0 \), ±0.5, ±1, ±1.5 and ±2 V. The change in surface potential values measured on the left gold lead when biased with \( V_{ch} = +2 \) and −2 V is now ~4.18 V, i.e. 4.5% larger than the expected 4 V, suggesting that this technique provides improved SP measurements even over relatively small structures with a size of several micrometers. This result is in a very good agreement with recent finding, where a negligible averaging effect was demonstrated for graphene samples using FM-KPFM mode.

**Electrostatic force microscopy and spectroscopy: experimental results.** Figure 4a shows an EFM phase map of the grounded device. Due to the high spatial resolution, the edges of 2LG domains are sharp and well defined. Figure 4b shows the recorded SP values obtained from EFS measurement points over the area of 1LG and 2LG. As this is a spectroscopy rather than a mapping technique, values may be slightly affected by the exact position. In this instance, \( \Delta V_{CPD}^{1-2LG} \) is 110 mV is in a reasonable agreement with the results obtained by FM-KPFM, this being expected as both techniques are sensitive to the force gradient. The discrepancy can be attributed to only a few EFS experimental points obtained on the small isolated 2LG domain at the center of the channel, whereas a significantly larger number of points were measured with FM-KPFM. Further improvement of EFS method and better agreement with FM-KPFM can be achieved by decreasing the step size between measurement points. Results of measurements of 200 EFS spectroscopy points taken along the center of the channel between the two gold contacts with the left contact biased at \( V_{ch} = 0 \), ±1 and ±2 V are shown in Figure 4c.

Even the most accurate FM-KPFM and EFS techniques provide a non-zero reading of the surface potential on the grounded electrode, i.e. \( V_{CPD} = -365 \text{ mV} \) for FM-KPFM (Figure 3c) and \( V_{CPD} = -723 \text{ mV} \) for EFS (Figure 4c). This discrepancy is the result of a work function difference between the gold and PFQNE-AL probe. Further to this, we account for the resulting work function difference by subtracting the \( V_{ch} \) obtained from the grounded right contact from the experimental value of the \( V_{CPD} \), i.e. \( \Delta V = V_{CPD}(V_{ch}) - V_{CPD}(0) \). The procedure was performed using results of all three experimental techniques for the range of applied \( V_{ch} \) providing \( \Delta V \) for the left gold electrode (Figure 5). The measured potential drop is typically 27.6% lower than the actual \( V_{ch} \) for AM-KPFM, whereas it is 4.4% and 7.8% higher for FM-KPFM and EFS, respectively. The lower \( \Delta V \) measurements are consistent with spatial averaging, as the relatively large base of the cantilever weakly interacts with the device channel and the right contact, both of which are at a lower \( V_{CPD} \) than the left contact, as was discussed above. The higher \( \Delta V \) measurement with FM-KPFM could be a result of an overestimation of the SP due to a relatively large excitation voltage of \( V_{AC} \approx 8 \text{ V} \), whereas the discrepancy with EFS rises from un-optimized fitting parameters.

**Work function calibration.** We employed the use of force gradient techniques to provide accurate measurements of work function of 1LG and 2LG. Initially, work function of the PFQNE-AL probe was calibrated against the work function of the gold leads: \( \Phi_{probe} = \Phi_{Au} + \epsilon \Delta V_{CPD} \), where \( V_{CPD} \) was measured on the grounded gold electrodes. Ultraviolet photoelectron spectroscopy (UPS) measurements were carried out on four separate samples of gold deposited by e-beam evaporation under the same conditions as the deposition of the gold electrodes. The spectra were acquired with voltage of ~19.04 V applied to the sample. The Fermi edge was centered at 0 eV by measuring the offset from a high resolution Fermi edge spectrum of the silver calibration sample. The offset was used to correct the energy scale for all four Au spectra (Figure 1d). Using the indicated energies obtained from the spectra, we can determine the work function of the 1LG and 2LG domains.

\[
\begin{align*}
\Phi_{1LG} &= \Phi_{2LG} = \Phi_{Au} + \epsilon \Delta V_{CPD} \\
\Phi_{1LG} &= \Phi_{2LG} = \Phi_{Au} + \epsilon \Delta V_{CPD}
\end{align*}
\]

where \( \epsilon = 1.8 \) for the gold leads. The work function values were determined to be \( \Phi_{1LG} = 4.5 \pm 0.2 \text{ eV} \) and \( \Phi_{2LG} = 4.6 \pm 0.2 \text{ eV} \). The lower \( \Phi_{1LG} \) and higher \( \Phi_{2LG} \) values can be accounted for by considering the measurement technique, which uses the force gradient rather than the force and also leads to improved spatial resolution of FM-KPFM compared to AM-KPFM. Further improvement of EFS method and better agreement with FM-KPFM can be achieved by decreasing the step size between measurement points.
the samples, the work function was calculated. The difference in energy between the Fermi edge measured on a silver calibration sample and the cut off (x) is given by $x = h - \Phi$, where the energy of the incident photon is $h = 21.22$ eV. The cut off was obtained by fitting a line to the relevant part of each spectrum, determining its gradient and the point at which it crosses the energy-axis. The absolute value of $x = 16.40$ eV was established, thus measuring the work function of all four gold samples as $\Phi_{Au} = 4.82$ eV. UPS characterization performed in ultra-high vacuum (UHV) would include all irreversibly bound adsorbates (chemisorbed oxygen and physisorbed hydrocarbon) attached to the surface prior to the measurements. Some uncertainty may arise from the reversible adsorption of species, such as water, which may form surface dipoles leading to a small change of the $\Phi_{Au}$ on transferring the sample from UHV to ambient environmental conditions. Owing to its relatively high standard electrode potential of 1.52 eV\textsuperscript{18,19}, gold is very stable in air, being less prone to both oxidation and formation of submonolayer of water in ambient conditions (both these factors could potentially affect $\Phi_{Au}$) than most other good conductors. Following Ref. 20, we can estimate that $\Phi_{Au}$ may decrease by $\sim$3% (top estimation) as the relative humidity changes from 0 to 40%. Use of a more stable metal would be ideal for tip calibration, however gold remains one of the best electrode materials.

Using the measured work function of gold, we calculated the work function of the probe to be $\Phi_{probe} = 4.09$ eV using EFS (Figure 1c). Then, the work function of 1LG and 2LG were determined: $\Phi_{sample} = \Phi_{probe} - e\Delta V_{CPD}$ using the measured values of the SP extracted from the line profiles of the potential maps, i.e. $V_{CPD} = -454$ and $-344$ mV for 1LG and 2LG, respectively, see Figure 4b. This defines work functions of $\Phi_{1LG} \sim 4.55 \pm 0.02$ eV and $\Phi_{2LG} \sim 4.44 \pm 0.02$ eV. These work function values are within the range of previously reported results of 4.41–4.57 eV for 1LG measured with FM-KPFM\textsuperscript{21}. It should be noted that the work function of graphene depends on the carrier density and is, therefore exceptionally sensitive to substrate and environmental gating due to its two-dimensional nature. For example, the published values were reported to change with varying lab ambient, i.e. the change of work function and SP due to adsorbates being $<50$ meV\superscript{20}a and $\sim 130$ meV\superscript{21}, respectively.

**Contactless resistance measurements.** High accuracy of FM-KPFM technique provides an excellent contactless method for measuring the electrode-graphene contact resistance with no need for specifically patterned electrodes\textsuperscript{8}, which is typically used with the transmission line method. Using experimental results shown in Figure 3c (i.e. line profiles of $V_{CPD}$ at $V_{ch} = \pm 2$ V), contact and channel resistance can easily be deduced by normalizing these line profiles $[V_{CPD}(V_{ch}) - V_{CPD}(0)]/V_{ch} = \Delta V/V_{ch}$ as shown in Figure 6a. This procedure accounts for any intrinsic $V_{CPD}$ changes, i.e. variations in the work function of features, such as 1LG, 2LG and gold. The resulting normalized line profile is solely a consequence of the potential drop at electrode-graphene contacts and along the graphene channel due to changes in the resistance. Dependence of the normalized voltage drop $\Delta V/V_{ch}$ as measured across the left (right) contacts and graphene as well as across the graphene channel (i.e. points 1–2, 3–4 and 2–3, respectively, in Figure 6a) are plotted in Figure 6b. While the voltage drop within the graphene channel is constant for all applied $V_{in}$, this value changes linearly on electrode-graphene contacts. Careful inspection of the electrode-graphene potential drop for both contacts reveals a clear $V_{ch}$ dependence. Focusing on the left contact (points 1–2), relative change of the voltage on electrode-graphene channel is $\Delta V = 0.55$ and $-0.91$ V for $V_{ch} = + 2$ V and $-2$ V, respectively. However, at the right contact, the $\Delta V = 0.83$ and $-0.52$ V for $V_{ch} = + 2$ V and $-2$ V, respectively. From potential drop and $I-V_{ch}$

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**Figure 5** | Direct comparison of the surface potential measurements. Normalized SP values as measured by AM-KPFM, FM-KPFM and EFS techniques on the left gold electrode in dependence on the voltage applied to the same gold electrode.
could affect the flow of charge and, thus, the contact resistance. However, it is rather difficult to give a quantitative measure of this effect from our experiments.

**Discussion**

Using epitaxial graphene Hall bars with gold electrodes, we have demonstrated significant differences in accuracy and resolution between AM-KPFM and FM-KPFM techniques in determining the difference in surface potential between 1LG and 2LG. Values of $\Delta V_{\text{CPD,1-2LG}}$ measured with FM-KPFM demonstrate a threefold increase as compared to AM-KPFM. While AM-KPFM measures the electrostatic force on the cantilever, FM-KPFM is sensitive to the force gradient. Thus, AM-KPFM gives a weighted average of the signal, including contributions from the surface under the probe cone and cantilever. Sensitivity to the shorter range force gradient characteristic for FM-KPFM leads to spatially-confined contributions, which arise only from the probe apex, thereby reducing the spatial averaging of measured work functions observed in AM-KPFM. We experimentally demonstrate that FM-KPFM and consequently EFS have a greater degree of spatial resolution ($<20$ nm) than AM-KPFM. Improvement in spatial resolution is clear on comparing the sharpness of the potential maps obtained. Moreover, we show that use of FM-KPFM and a calibrated probe provide a simple and straightforward method of obtaining an accurate measure of the work function of 1LG and 2LG. Accuracy of measurements is provided by initial calibration of the KPFM signal against the gold electrodes, whose work function was measured independently by UPS, however, keeping in mind that the work function of gold may somewhat change on transferring the sample to ambient due to adsorption of reversible species as discussed above. This improvement in measurement technique enables greater accuracy in determination of the work function of 1LG and 2LG, with values of $\Phi_{\text{1LG}} \sim 4.55 \pm 0.02$ eV and $\Phi_{\text{2LG}} \sim 4.44 \pm 0.02$ eV, respectively, as valid for the particular sample studied here and in specified ambient conditions. Thus, the experimental procedure outlined in this paper can be implemented as a generic route for standardization of work function measurements applicable not only to graphene but to a wide class of nanoscale materials and devices, for example in light emission and solar devices, displays, etc.

FM-KPFM was also used to investigate: i) the contact resistance between the gold electrode and graphene, revealing a non-Ohmic behavior, and ii) the resistance of the graphene channel showing Ohmic behavior with $R_{\text{ch}} \sim 33$ k$\Omega$ and $\rho_{\text{ch}} \sim 2.7 \times 10^{-6}$ Ohm cm. This simple contactless method can be used to investigate the specific components of the total resistance, without fabricating devices for the transmission line method.

Our results unambiguously demonstrate that the measured values of the SP of single- and bi-layer graphene largely depend on the accuracy of the technique. However, we would like to stress that the obtained absolute values are not by any means the fundamental parameters for graphene and are strongly dependent on the state of the surface. The carrier concentration and correspondingly the work function of graphene are exceptionally sensitive to substrate (intrinsic) and environmental (extrinsic) gating due to its two-dimensional nature. Being representative of the surface state, the work function can be strongly affected by such factors as traces of gas contamination, temperature and, in particular, humidity. For example, by conducting experiments in the controllable humidity (outside of the scope of the present paper) we found that standard ($\sim 10–15\%$) variations corresponding to typical day-to-day change of the lab humidity, led to a corresponding change of $\Delta V_{\text{CPD,1-2LG}} \sim 25–30$ mV. Moreover, it was reported that 1LG and 2LG have different adsorption energies for gases, such as NO$_2$, and water vapour, which leads to the differences in the doping levels for these two domains. Thus, without reproducing substrate and environmental conditions, the SP and work functions of graphene

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**Figure 6** | Contactless resistance measurements with FM-KPFM.

(a) Normalized surface potential line profiles. Experimental values are obtained by FM-KPFM along the dashed line in Figure 3a. (b) $\Delta V/V_{\text{ch}}$ and (c) resistance measurements of left contact, right contact and across the graphene channel, i.e. points 1–2, points 3–4 and points 2–3, respectively, in (a). Dashed lines are guides for the eye. Inset in (b) shows the dependence of the total current ($I$) through the circuit on the bias voltage ($V_{\text{ch}}$).
obtained in different experiments cannot be adequately compared. This places an even stronger emphasis on the necessity of standardization in the measurements of the SP and work function for each particular sample of interest and for given environmental conditions.

**Methods**

Sample preparation. Nominally monolayer epitaxial graphene was prepared by sublimation of Si and the subsequent graphene formation on the Si-terminated face of an on-axis 4H-SiC(0001) substrate at 2000 °C and 1 bar argon gas pressure. Details of the growth and structural characterization are reported elsewhere. The specific synthesis route has been developed to provide large areas of homogeneous single-layer graphene. The resulting material is n-doped, owing to charge transfer from the interfacial layer adsorbed, with the measured electron concentration in the range n = 6–20 × 10¹⁶ cm⁻² and carrier mobility of μ ~ 3000 cm² V⁻¹ s⁻¹ at room temperature. The epitaxial graphene device was fabricated by electron beam lithography (PMMA/MMMA and ZEP520 resists), oxygen plasma etching and evaporation of Ti/Au (5/100 nm) electrodes. Details of the sample fabrication are reported elsewhere. The device comprises two cross with a channel width of 4.8 μm, surrounded by 1.6 μm-wide trench etched down into the SiC substrate. The transport measurements were performed in air, at room temperature, in a dark environment. Details of the measurements are reported in Ref. 26.

Standard lithography fabrication methods lead to a thin (1–2 nm) layer of a resist residue on top of the graphene. The residues can significantly affect the carrier density and even type, as shown in our previously published work in this area. For instance, we show exposure of the resist residue to 250-nm wavelength UV light for 20 minutes leads p-doping of the graphene, where n = 2.5 × 10¹⁴ cm⁻² with δμL = 4.68 eV. Subsequently, cleaning the residues using contact-mode AFM restored the n-type characteristics of the graphene device. Oxygen plasma etching and evaporation of Ti/Au (PMMA/MMA and ZEP520 resists) are performed to prepare relevant 2D and 3D electrodes, which was measured by ultraviolet photoemission spectroscopy (UPS) and monitored by atomic force microscopy. Moreover, 1.6–20 nm of Ti/Au is evaporated to measure the work function of the graphene sample. Calibrated work function measurements of graphene were obtained with EFS by calibrating the work function of the probe against the known work function of gold electrodes, which was measured by ultraviolet photoemission spectroscopy (UPS), see Figure 1d.

**Amplitude-modulated KPFM.** The AM-KPFM, discussed here, is performed as a dual-pass technique; topography line profile is mapped with tapping mode AFM during the first pass, which is then traced at a set lift height above the surface performing the surface potential measurement (Figure 1a). During the second pass of AM-KPFM, the mechanical drive to the cantilever is disabled and an AC bias voltage (V_{AC} = 2 V) is applied to the probe at the mechanical resonance f₀ of the cantilever. The V_{AC} causes the cantilever to oscillate due to the attractive and repulsive electrostatic interaction (E_{ST}) between the probe and the sample:

\[ F_{ST} = -\frac{1}{2} \frac{dC}{dz} (V_{DC} - V_{AC}) + \frac{V_{AC}}{\sin(\phi)} \tag{3} \]

where \( V_{DC} \) is a DC bias voltage and \( V_{AC} \) is a constant potential difference between the probe and sample. A proportional-integral-derivative (PID) feedback loop monitors and simultaneously maintains the amplitude of the cantilever oscillation and a compensating \( V_{DC} \) to the probe to cancel the probe-sample electrostatic forces, i.e., \( E_{ST} = 0 \) at each pixel. The applied \( V_{DC} \) is recorded at each point, providing a map of the SP. This conventional dual-pass KPFM is a well-established technique, used widely for quantitative probing of the surface potential of graphene,32,33,35,36. Generally, the technique suffers from a poor lateral resolution, 50–70 nm, see e.g. Ref. 31.

**Frequency-modulated KPFM.** FM-KPFM, discussed here, is a single-pass technique, which gives a larger degree of spatial resolution than AM-KPFM as it measures the force gradient (dF/dz)² rather than the force acting on the entire cantilever. The probe–sample electrostatic forces affect the resonance frequency of the cantilever, i.e., \( f_{0} \) MOD. Therefore, recording \( V_{DC} \) at each pixel. In a similar fashion to AM-KPFM, the probe-sample electrostatic forces are nullified when \( V_{DC} = V_{CPD} \), therefore generating the SP map. However, in contrast to AM-KPFM, FM-KPFM typically requires stiffer, higher frequency cantilevers. FM-KPFM offers a higher spatial resolution of ~20 nm as a result of force gradient localization to the probe apex and higher sensitivity to frequency shifts.6,25 While AM-KPFM is usually performed as a dual-pass technique where first topography and then SP are measured along the same line in an alternating fashion, FM-KPFM is most often performed as a single-pass technique, where topography and potential are recorded simultaneously, thus improving the speed of image capture. However, it should be noted that being either single- or dual-pass is not a definition of the techniques, as other examples have been demonstrated previously.9,36,37

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**Author contributions**

O.K. designed the research, R.Y. grew the samples, V.P. fabricated nanodevices, V.P. and O.K. analyzed the data. All authors discussed the results, participated in writing and commented on the manuscript. All authors reviewed the manuscript.

**Additional information**

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