Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Current transport in graphene/AlGaN/GaN vertical heterostructures probed at nanoscale

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1. Graphene transfer

Graphene was grown by Chemical Vapour Deposition on a ~25 μm thick polycrystalline copper foil at a temperature of 1000 °C by using CH_4/H_2 as precursors. As schematically illustrated in Fig.S1, a 15×15 mm foil was coated with Poly(methyl methacrylate) (PMMA), which works as carrier layer, by spin coating for 60 seconds at 1000 rounds per minutes and baked at 150°C for 10 minutes. Electrochemical delamination of graphene from copper foil [1] was performed in a 0.2 M KOH water solution with an applied voltage of 5 V between the copper foil, used like cathode, and an anode of gold. Graphene sustained on PMMA was printed on the target substrate and PMMA was removed in Acetone at room temperature.

2. Determination of graphene/AlGaN separation by AFM

The separation between a graphene layer and AlGaN was determined by tapping mode atomic force microscopy, adopting the following procedure, which was already demonstrated to measure the separation between graphene and SiO_2 [2]. To this aim, graphene mechanically exfoliated from HOPG onto AlGaN was used. Analyses were carried out on few layers of graphene (FLG) samples including monolayer and n-layers regions in a step-like fashion. This is illustrated in Fig.S2, reporting the morphology (a) and phase (b) images of a region including bare AlGaN and AlGaN coated by the FLG. Fig.S2(c) shows the height profile measured along the indicated line in Fig.2(a).

The height separation between graphene monolayer and AlGaN and between the stacked layers is also indicated. In Fig.S2(d) the height values measured with respect to the AlGaN baseline level were plotted as a function of the layer number N. By linear fitting of the data, the interlayer separation ($h=0.34\pm0.01$ nm) and the intercept with the height axis ($t_0=0.07\pm0.04$ nm) were extracted. The separation between a graphene single layer from the AlGaN was extimated as $t=h+t_0$, i.e. $t\approx0.41$ nm.

3. Graphene carrier density in a biased Gr/AlGaN/GaN heterostructure

The energy bandstructures of the Gr/AlGaN/GaN heterostructure at equilibrium ($V_{tip}=0$) and under reverse ($V_{tip}<0$) or forward polarization ($V_{tip}>0$) are schematically depicted in Fig.S3(a), (b) and (c), respectively. Here a very thin ($t\sim0.41$ nm) vacuum barrier has been assumed to separate the graphene layer from the AlGaN surface, which exhibit a donor-like surface states density n_d.

The carrier density $n_s(V_{tip})$ of the 2DEG at the AlGaN/GaN interface can be directly obtained by capacitive measurements, as discussed in the main text (see Fig.6(a)). The graphene carrier density under reverse bias can be evaluated by applying the Gauss' law, as schematically illustrated in Fig.S3(b).

In particular, the Gauss' law applied at the interface between graphene and the vacuum barrier yields:

$$\frac{\varepsilon_0\Delta}{t} = -qn_{gr} \tag{S1}$$

Applied to the vacuum barrier/AlGaN interface, it yields:

$$-\frac{\varepsilon_0\Delta}{t} + \frac{\varepsilon_0\varepsilon_{AlGaN}\Delta V}{d} = q(n_d - \sigma) \tag{S2}$$

Finally, the application to the AlGaN/GaN interface gives:

$$-\frac{\varepsilon_0\varepsilon_{AlGaN}\Delta V}{d} = q(\sigma - n_s) \tag{S3}$$

By combining Eqs. (S1), (S2) and (S3), the following relation between graphene and AlGaN/GaN 2DEGs carrier densities is obtained:

$$n_{gr} = n_d - n_s \tag{S4}$$

According to Eq.(S4), the surface donors density at $V_{tip}=0$ is $n_{d0}=n_{gr0}+n_{s0}$, where n_{gr0} and n_{s0} are the graphene and AlGaN/GaN 2DEGs carrier densities at $V_{tip}=0$, which have been determined as discussed in the main text. Assuming that $n_d \approx n_{d0}$, $n_{gr}(V_{tip})$ can be expressed as:

$$n_{gr}(V_{tip}) = (n_{gr0} + n_{s0}) - n_s(V_{tip}) \quad . \tag{S5}$$

By analogous consideration $n_{gr}(V_{tip})$ under forward bias can be determined.

4. Quantum capacitance contributions to the capacitance-voltage characteristics of Gr/AlGaN/GaN heterostructures

As discussed in the main text, the capacitance of the Gr/AlGaN/GaN heterostructure can be described as the series combination of three contributions: (i) C_{AlGaN}, i.e. the capacitance of the plane capacitor with the metal tip and the 2DEG as electrodes; (ii) $C_{Q,ord}$, i.e. the quantum capacitance of the AlGaN/GaN "ordinary" 2DEG [3]; (iii) $C_{Q,gr}$, i.e. the graphene quantum capacitance [4].

The two latter contributions can be expressed as:

$$C_{Q,ord}=m_{eff}q^2/(\pi\hbar^2) \tag{S6}$$

and

$$C_{Q,gr}(n_{gr}) = \frac{2q^2}{\hbar v_F \sqrt{\pi}} \sqrt{n_{gr}} \tag{S7}$$

and are reported together with the measured capacitance of the Gr/AlGaN/GaN heterostructure (C_{tot}) in Fig.S4. This comparison shows that both $C_{Q,ord}$ and $C_{Q,gr}$ are much larger than C_{AlGaN}.

Figure Captions:

Fig.S1. Schematic representation of the used graphene transfer procedure from metal foil to target substrate via (a) PMMA deposition, (b) electrochemical delamination, (c) PMMA/Gr printing on the target substrate and (d) PMMA elimination.

Fig.S2. AFM morphology (a) and phase (b) images of a region including bare AlGaN and AlGaN coated by few layers of graphene (1, 4, 5, 6 and 7 layers layers in a step-like fashion). (c) Height profile measured along the indicated line in (a). (d) Height values measured with respect to the AlGaN baseline level as a function of the layer number N. By linear fitting of the data, the interlayer separation (h=0.34±0.01 nm) and the intercept with the height axis (t_0=0.07±0.04 nm) were extracted.

Fig.S3 The energy bandstructures of the Gr/AlGaN/GaN heterostructure at equilibrium ($V_{tip}=0$) (a), under reverse ($V_{tip}<0$) (b) or forward polarization ($V_{tip}>0$) (c)

Fig.S4 Measured capacitance of the Gr/AlGaN/GaN heterostructure (C_{tot}) compared with the quantum capacitance of the AlGaN/GaN 2DEG ($C_{Q,ord}$) and the graphene quantum capacitance ($C_{Q,gr}$)

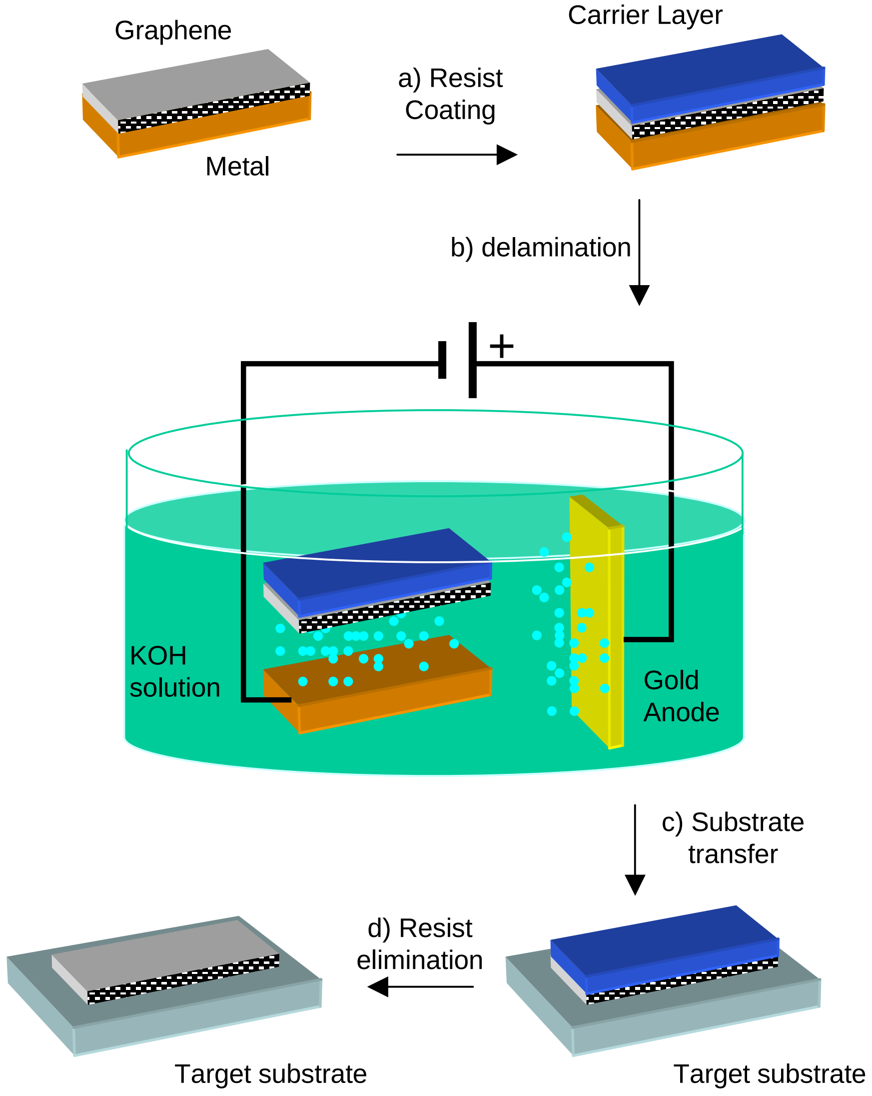
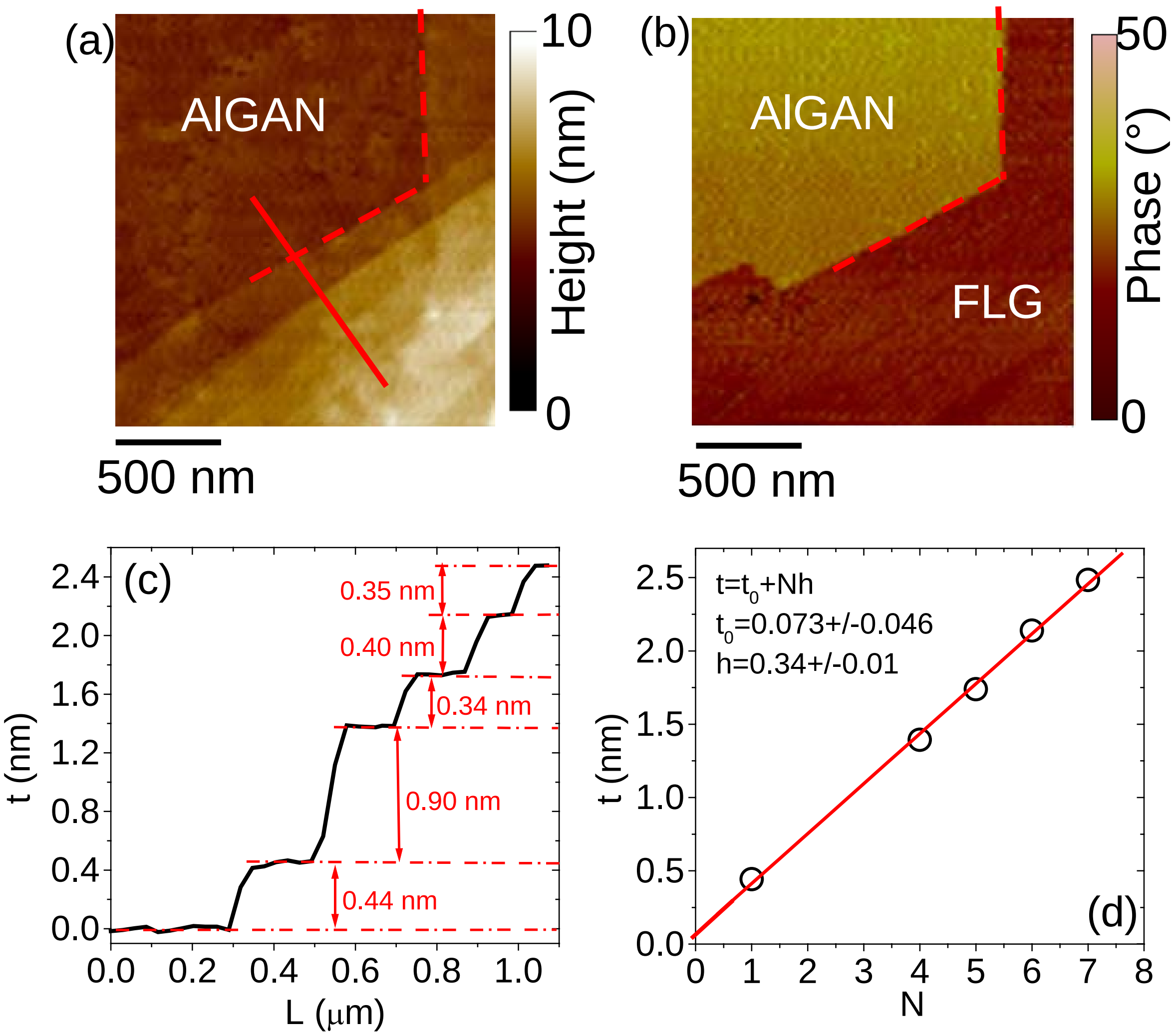
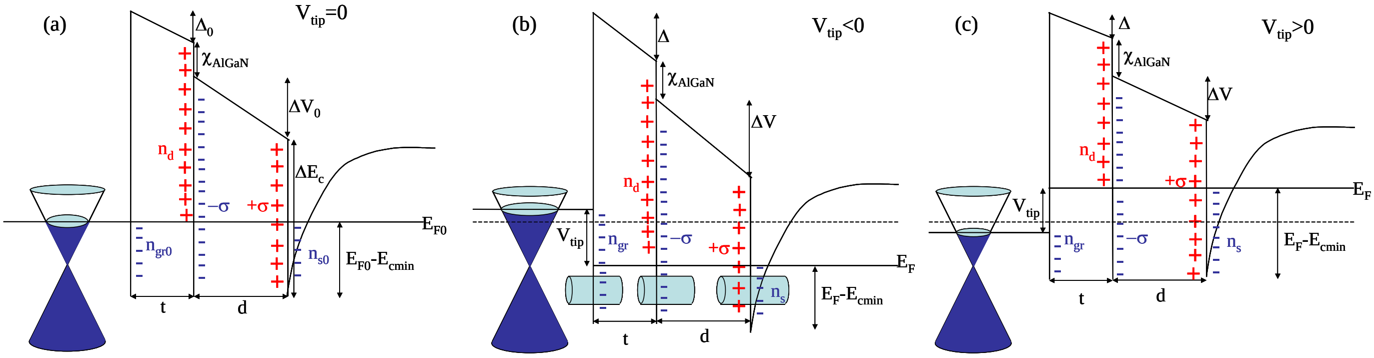

Graphene
Metal
a) Resist
Coating
Carrier Layer
b) delamination
+
KOH
solution
Gold
Anode
c) Substrate
transfer
d) Resist
elimination
Target substrate
Target substrate

Fig.S1



(a)
AlGAN
10
Height (nm)
0
500 nm
(b)
AlGAN
FLG
50
Phase (°)
0
500 nm
(c)
0.35 nm
0.40 nm
0.34 nm
0.90 nm
0.44 nm
t (nm)
L (μm)
(d)
t=t0+Nh
t0=0.073+/-0.046
h=0.34+/-0.01
t (nm)
N

Fig.S2



(a)
$V_{tip}=0$
Δ_0
χ_{AlGaN}
ΔV_0
n_d
ΔE_c
$-\sigma$
$+\sigma$
E_{F0}
n_{gr0}
n_{s0}
$E_{F0}-E_{cmin}$
t
d
(b)
$V_{tip}<0$
Δ
χ_{AlGaN}
ΔV
n_d
V_{tip}
n_{gr}
$-\sigma$
$+\sigma$
E_F
n_s
E_F-E_{cmin}
t
d
(c)
$V_{tip}>0$
Δ
χ_{AlGaN}
ΔV
n_d
$+\sigma$
E_F
V_{tip}
n_{gr}
$-\sigma$
n_s
E_F-E_{cmin}
t
d

Fig.S3

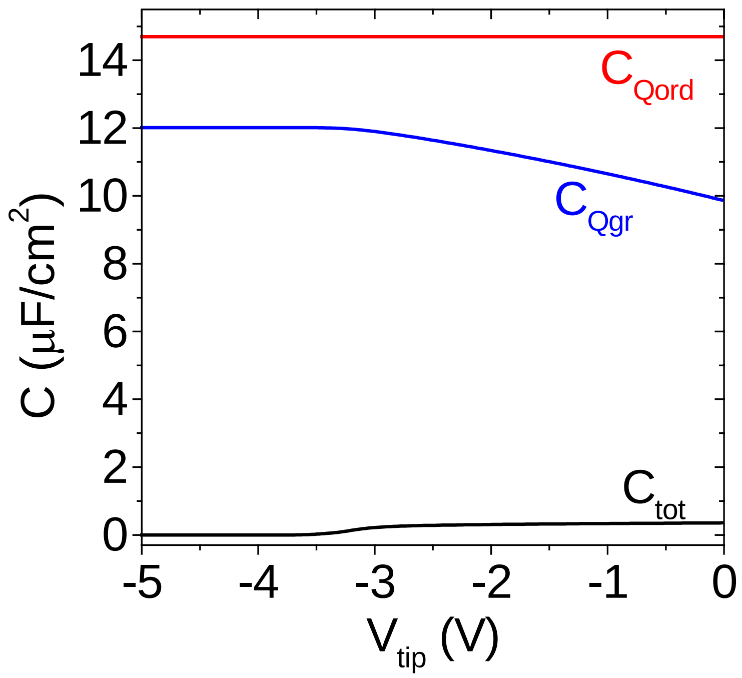


Fig.S4

References:

[1] *Electrochemical Delamination of CVD Grown Graphene Film: Toward the Recyclable Use of Copper Catalyst*, Y. Wang, Y. Zheng, X. Xu, E. Dubuisson, Q. Bao, J. Lu, K. P. Loh, ACS Nano, 5, 9927–9933 (2011).

[2]*Optical, morphological and spectroscopic characterization of graphene on SiO_2*, F. Giannazzo, S. Sonde, V. Raineri, G. Patanè, G. Compagnini, F. Aliotta, R. Ponterio, and E. Rimini, Phys. Status Solidi **7**, 1251-1255 (2010).

[3] *Quantum capacitance devices*, S. Luryi *Appl. Phys. Lett.* 52, 501-503 (1988).

[4] *Measurement of the quantum capacitance of graphene*, J. Xia, F. Chen, J. Li, N. Tao, Nature Nanotechnology 4, 505 - 509 (2009).