SUPPLEMENTARY INFORMATION

S1. Raman spectra of pristine graphene, electrolyte and electrolyte covered graphene

Fig. 1a,b plot the Raman spectra for $V_{BG} = 0$ V and $V_{nBG} = 20$ V. Fig. 1c to 1f show the spectra recorded during the top gate experiment. Fig. 1c is the PEO Raman spectrum. This has three prominent peaks at $\sim$ 1282 cm^{-1} (P1), 1476 cm^{-1} (P2) and 2890 cm^{-1} (P3), which correspond to twisting, bending and stretching modes of the CH_2 bonds in the polymer[1]. Luckily they do not overlap the main features of graphene (see Figs. 1d,f). Furthermore these PEO Raman lines do not change with gating. Table 1 shows the comparison of G peak position, Pos(G), FWHM(G) and 2D/G height ratio, I(2D)/I(G), at zero gate voltage and the Dirac point for BG and TG.

It shows that at Dirac point we have the lowest Pos(G), maximum FWHM(G) and I(2D)/I(G). However, our sample has a lower I(2D)/I(G), FWHM(G) and higher Pos(G) than the most intrinsic samples measured to date[3, 2], due to the presence of charge impurities[3].

Table 1: G peak position, FWHM and 2D/G height ratio.

Gate Voltage	Pos(G) (cm^{-1})	FWHM(G) (cm^{-1})	I(2D)/I(G) (height ratio)
$V_{BG} = 0.0$ V	1586.7	8.7	2.0
$V_{nBG} = 20$ V	1584.0	12.6	3.1
$V_{TG} = 0.0$ V	1586.4	13.9	2.75
$V_{nTG} = 0.6$ V	1583.1	14.9	3.3

S2. Graphene gating through water

In order to check the influence of the top gate on the carrier concentration in graphene, experiments with graphene gated through water have been performed, following a similar procedure to that described in Ref. [4]. The samples are prepared by standard microexfoliation, followed by e-beam

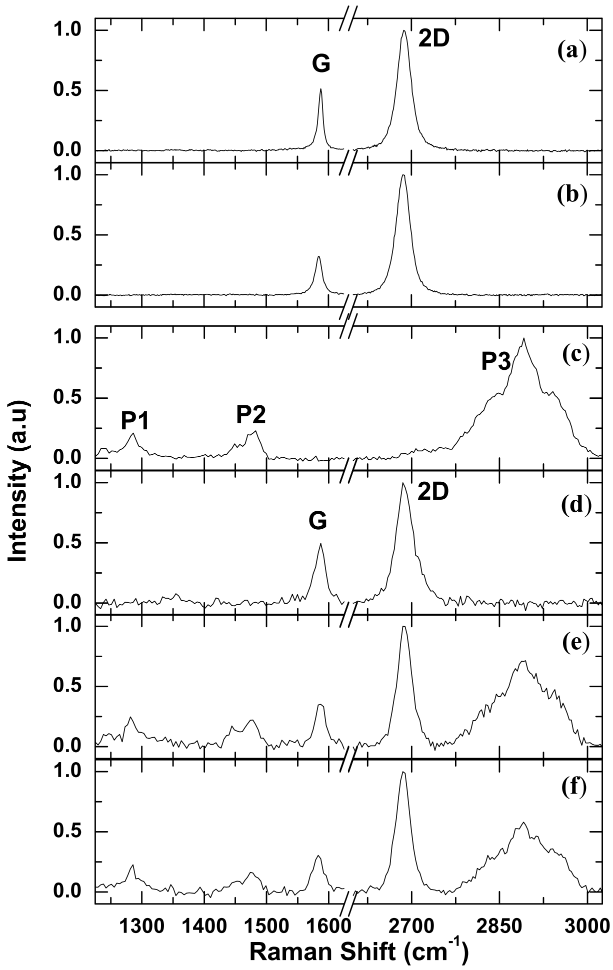

Figure 1: Raman spectra at (a) $V_{BG} = 0.0$ V and (b) V_{BG}=V_{nBG}=20V. (c) Raman spectra of PEO+$LiClO_4$ mixture. (d) Raman spectra of graphene before pouring the polymer electrolyte. (e) Raman spectra at $V_{TG} = 0.0$ V and (f) V_{TG}=V_{nTG}=0.6V. P1, P2 and P3 are the polymer peaks[1]

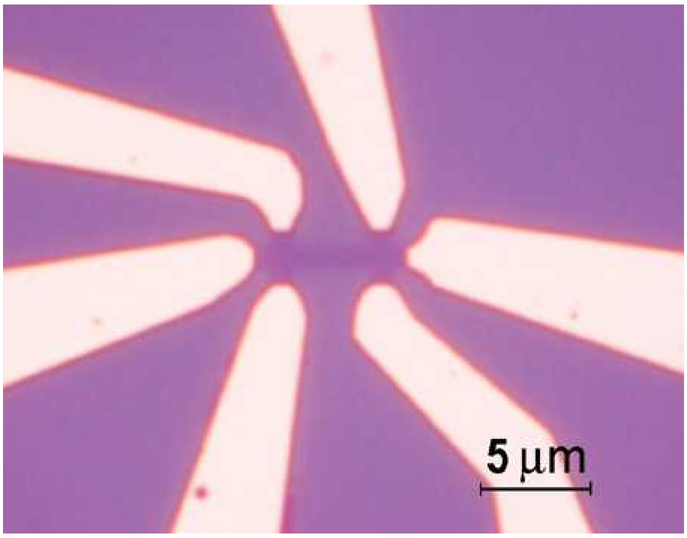

Figure 2: Optical micrograph showing the geometry of the graphene device used for water top gating

lithoghraphy, deposition of Ti/Au contacts and reactive plasma etching steps during which a set of contacts and Hall bar mesa structure are defined (Fig. 2). A small droplet of water (of about 10^{-4} ml) is then applied on the graphene surface. This usually dries out within 1 minute. Thus, this set up is not stable over time, nor suitable for extensive Raman measurements, unlike the solid polymer electrolyte discussed in the paper. We take special care to minimize the contact between water and the leads to the graphene sample, in order to decrease the leakage current. In this way the leakage current is kept below 50nAmp. An Au wire is then immersed into the droplet in order to apply the top gate voltage. Standard low-frequency lock-in is used to measure the resistance of the sample as a function of top-gate voltage, Fig 3. A top gate voltage as high as 5V can be applied, after which the leakage current increases above 50nAmp. The sample appears to be p-doped at zero top-gate voltage (as expected for doping by water[5]). However, ambipolar behavior is observed when finite top-gate voltage is applied (Fig. 3). The maximum resistance at the charge neutrality point does not change after applying water (Fig. 3). The ON/OFF ratio is found to be ~ 40 when water top gating is using, which is a record high for present large graphene devices. This demonstrates that the capacitive coupling between the water and graphene is very high.

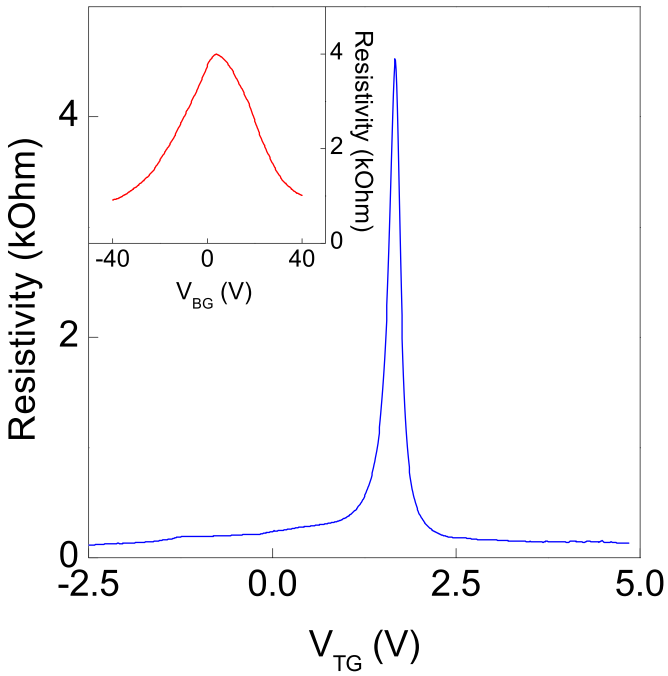

Figure 3: Resistivity as a function of back and top gate voltage

References

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