Dynamics of the Electric Double-Layer in Electrolyte Gated Graphene FETs

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The electric double-layer (EDL) of graphene FETs is studied in the time/frequency domain over electrolyte concentrations (c_{ion}) ranging from ($1 < c_{ion} < 150$ mM) (Fig.1). From impedance data the effective gate capacitance is modelled, using the graphene quantum capacitance (including a residual charge < $5x10^{11}$ cm^{-2}) in series with a bias-independent EDL capacitance (C_{EDL}), itself a series combination of Stern (C_{St}) and diffuse layer (C_{diff}) capacitances (Fig.2). A core tenet of FET biosensor principles is the notion that the Debye 'screening length', determined by c_{ion}, also controls C_{EDL} and therefore the potentiometric sensitivity of the device. In contrast to this assumption, our measured data is consistent with a model in which C_{EDL} is independent of c_{ion}, suggesting that the inner 'Stern' layer of the EDL is controlling the charge coupling of the channel (Fig.2) [1]. The electrical bandwidth of the FETs is then studied via a single-source mixing technique [2] (Fig.3), again as a function of c_{ion} (>1 mM). It is seen that the -3dB cut-off frequency of the FET transconductance can be described by a single-pole RC filter fit in which only the solution + graphene resistance is varying (C_{EDL} is constant). The transient dynamics of the EDL are examined via electrically pulsed gating, revealing that the time required to reach a steady-state current in the channel increases as c_{ion} reduces, ranging from ~10 μs (x1 PBS=150 mM) to ~1 ms in de-ionised water, timescales that are complementary to the cut-off frequencies (Fig.4).

References

[1] Dankerl *et al*. Phys. Rev. Lett. 106, 196103 (2011)
[2] Kulkarni *et al*. Acc. Chem. Res 2016, 49, 11, 2578
[3] Bazant *et al*. Phys. Rev. E 70, 021506 (2004)

Figures

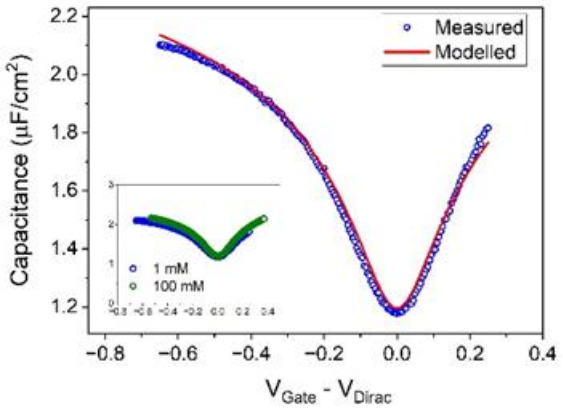

Figure 1: Gate capacitance as a function of c_{ion}. The form of the C-V curve does not depend on c_{ion} (inset)

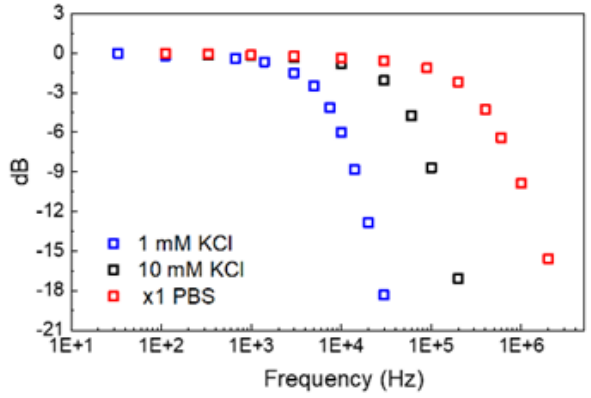

Figure 3: FET transconductance vs mixing frequency for various c_{ion}

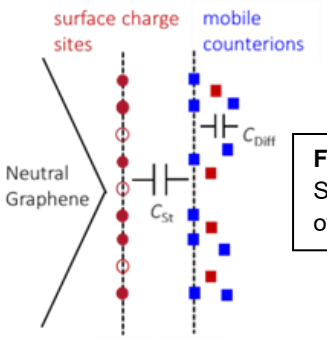

Figure 2: Schematic of EDL

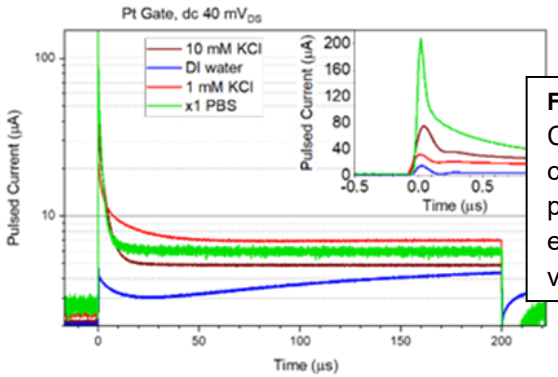

Figure 4: Channel current under pulsed excitation with varying c_{ion}