Supporting Information for

Determination of the thermal noise floor of graphene biotransistors

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1. Parasitic capacitance

To quantify parasitic capacitance, one GFET device was fully covered by photoresist. The photoresist acts as an insulating layer that blocks the electrolyte from touching the graphene. For this control device, the response of the circuit is dominated by the capacitive coupling between the electrolyte and the encapsulated metal electrodes. To quantify this parasitic capacitance, we apply a 25-mV ac bias between the electrolyte and the electrodes and measure the resulting ac current, $I^{\text{parasitic}}$. Figure S1 shows real and imaginary components of $I^{\text{parasitic}}$ (blue and green symbols, respectively). For comparison, the real and imaginary components of an exposed GFET device have been plotted (black and red symbols, respectively). These "raw currents" from exposed graphene devices are much larger than $I^{\text{parasitic}}$. As expected, $I^{\text{parasitic}}$ is independent of gate voltage (data not shown). When analyzing the properties of the graphene-electrolyte interface, we first measure "raw currents" and then subtract $I^{\text{parasitic}}$.

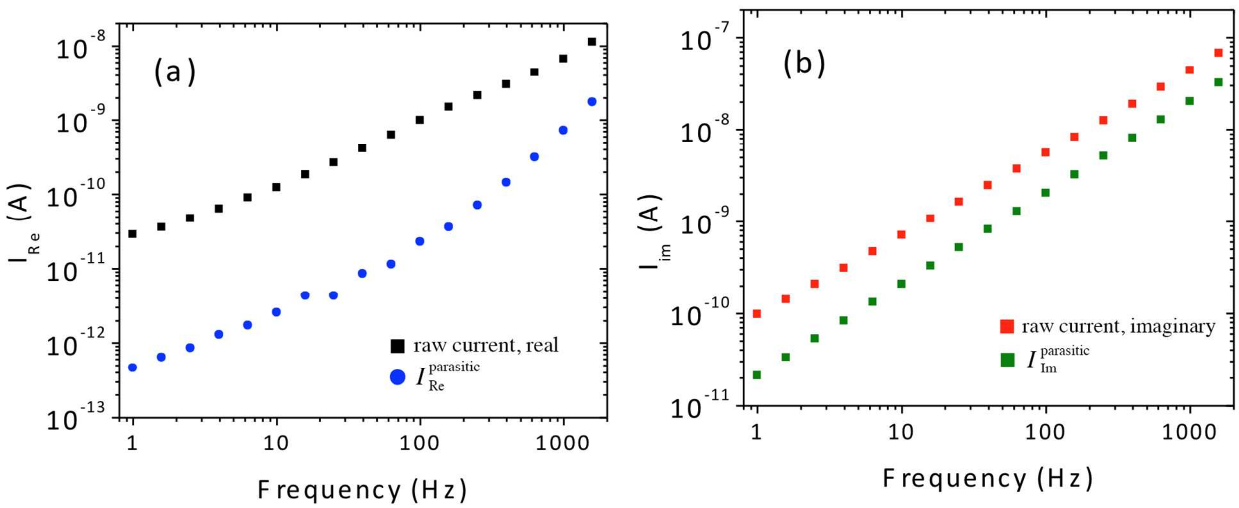


Figure S1. Real and Imaginary components, I_{re} and I_{im}, of current flowing between the electrolyte and a GFET device. The parasitic current is measured using a GFET device that is fully covered by photoresist (no direct contact between liquid and graphene). The "raw current" is measured from a device that has a window in the photoresist that exposes the graphene to electrolyte.

2. Quantum capacitance of graphene

A theoretical prediction for C_Q is found using the tight binding model to calculate the density of states in graphene,

$$C_Q = e^2 D(E_F) = e^2 \frac{2|E_F|}{\pi(\hbar v_F)^2} \quad , \qquad \text{(S1)}$$

where $v_F \approx 10^6$ ms^{-1}. Equation S1 predicts $|dC_Q/dV_g| = \alpha \cdot 23$ μF/V·cm^2, where $\alpha = C_{dl}/(C_{dl} + C_Q)$ describes the gate coupling efficiency ($E_F = \alpha e(V_g\text{-}V_D)$). The measured slope, $|dC_Q/dV_g| = 18$ μF/V·cm^2, agrees well with this theory (see Fig. 2d of main text). The measured minimum in quantum capacitance, $C_{Q,min} = 2$ μF/cm^2, is consistent with electrostatic disorder of order 100 meV (spatial fluctuations in E_F). Similar electrostatic disorder has been observed by other authors[1].

3. Impedance as a function of device area

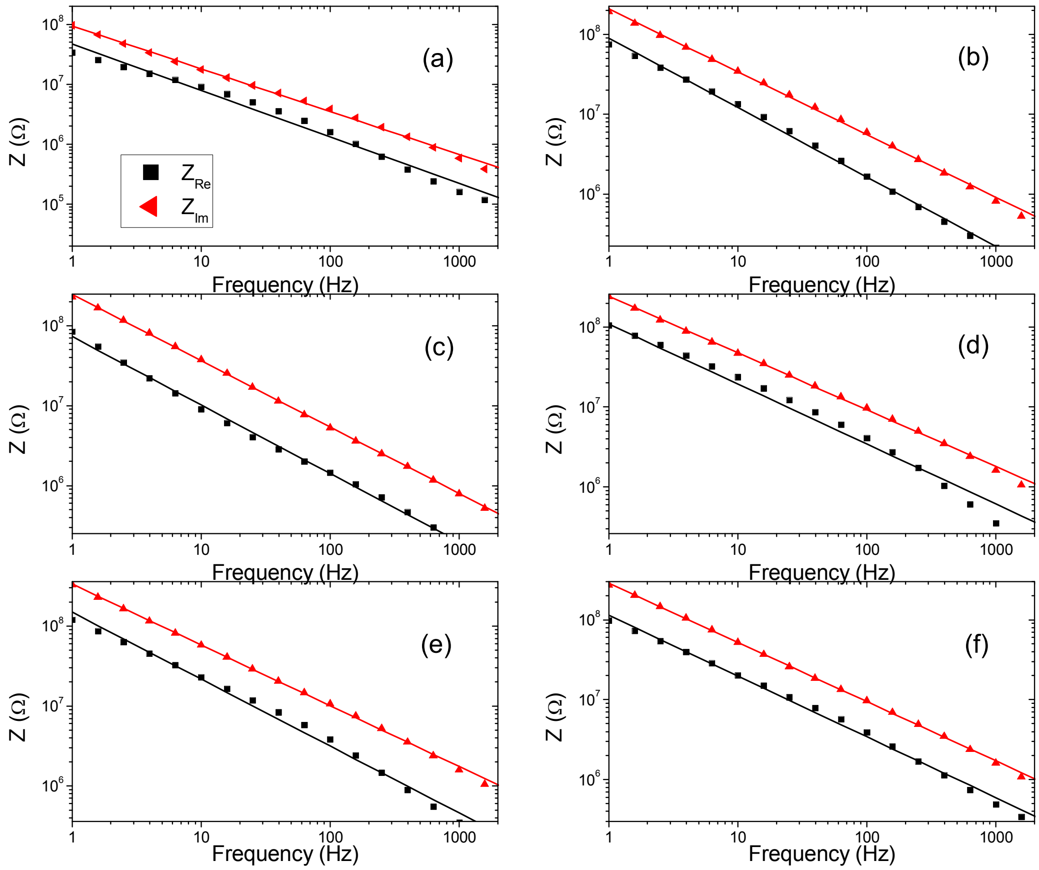


Fig. S2. Impedance vs. frequency for six devices, of areas a) 23,400 μm^2, b) 19,200 μm^2, c) 15,800 μm^2, d) 7,500 μm^2, e) 6,100 μm^2, and f) 5,000 μm^2.

4. Phase angle as a function of gate voltage

The phase angle between current and voltage, $\phi = \tan^{-1}(I_{im}/I_{re})$, is approximately independent of gate voltage.

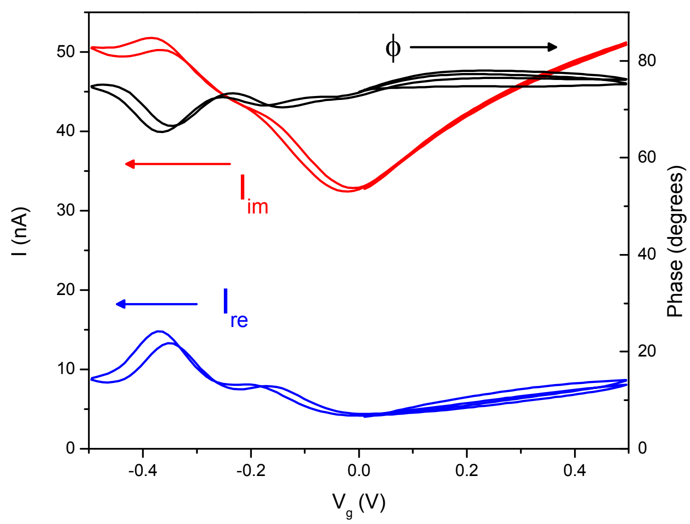


Fig. S3. Real and Imaginary current vs. gate voltage, and phase angle vs. gate voltage, for a device with graphene-electrolyte contact area A = 5040 μm^2.

5. Current fluctuations in a GFET device

GFET biosensors are designed to convert a small change in gate voltage to a large change in source-drain current, I_{sd}. Therefore, it is important to be aware of processes that add noise to the I_{sd} signal.

Fluctuations in I_{sd} are quantified by the power spectral density $S_I(f)$. The lower limit for $S_I(f)$ will be a superposition of two independent Johnson noise sources,

$$S_I(f) = S_I^{\mathrm{LG}}(f) + S_I^{\mathrm{CR}}, \qquad \text{(S2)}$$

where $S_I^{\mathrm{LG}}(f)$ is associated with liquid-gate Johnson noise, and S_I^{CR} is associated with channel resistance (CR) Johnson noise.

Current fluctuations generated by liquid-gate Johnson noise are given by

$$S_I^{\mathrm{LG}}(f) = V_{\mathrm{sd}}^2 \left(\frac{dG}{dV_{\mathrm{g}}}\right)^2 S_{V,\mathrm{th}}(f). \qquad \text{(S3)}$$

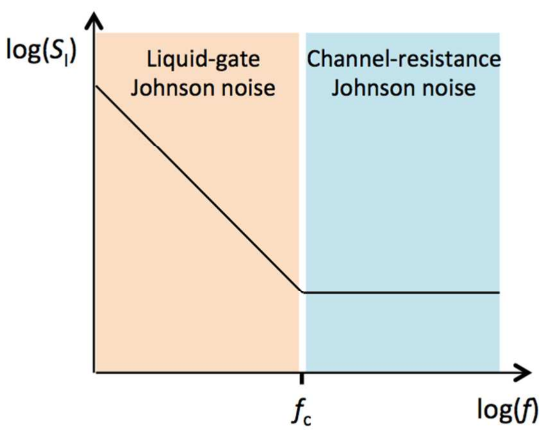
where $S_{V,th}(f)$ is the voltage noise on the gate, dG/dV_g is the transconductance of the GFET device, and V_{sd} is the source-drain bias. The magnitude of $S_I^{\mathrm{LG}}(f)$ depends on both frequency and device area, A.

Current fluctuations generated by channel-resistance Johnson noise are given by

$$S_I^{\mathrm{CR}} = 4kTG. \qquad \text{(S4)}$$

where G is the channel conductance. When $G = 1/(5\ \mathrm{k}\Omega)$, we find $S_I^{\mathrm{CR}} \sim 3 \times 10^{-24}\ \mathrm{A^2/Hz}$. This value is independent of frequency. Moreover, if graphene channels are fabricated in a square geometry (channel length = channel width), S_I^{CR} is also independent of A.

For typical device sizes and measurement bandwidths, liquid-gate Johnson noise is the dominant source of current fluctuations. We define the corner frequency, f_c, as $S_I^{\mathrm{LG}}(f_c) = S_I^{\mathrm{CR}}$ (see Fig. S4). Liquid-gate Johnson noise dominates when $f < f_c$. Figure S4 shows estimated values of f_c for a variety of device areas. To calculate f_c we use the lower limit $S_{V,th}(f)$ shown in Fig. 4 of the main text and assume $dG/dV_g = 1$ mS/V, $V_{sd} = 25$ mV.



A (μm²)	f_c (kHz)
1	4,700
10	310
100	21
1000	1.4

Fig. S4. Schematic plot illustrating the power spectral density of current fluctuations, S_I, as a function of frequency, f. In the absence of charge-trap noise, we expect the low frequency spectrum to be dominated by liquid-gate Johnson noise, while the high frequency spectrum is dominated by channel-resistance Johnson noise. The table shows estimated values of the corner frequency f_c for relevant device areas.

Supporting Information References

S1. Xia, J., Chen, F., Li, J. & Tao, N. Measurement of the quantum capacitance of graphene. *Nat. Nanotechnol.* **4,** 505–9 (2009).