

Supplementary Information: Laser-induced tuning of graphene field- effect transistors for pH sensing

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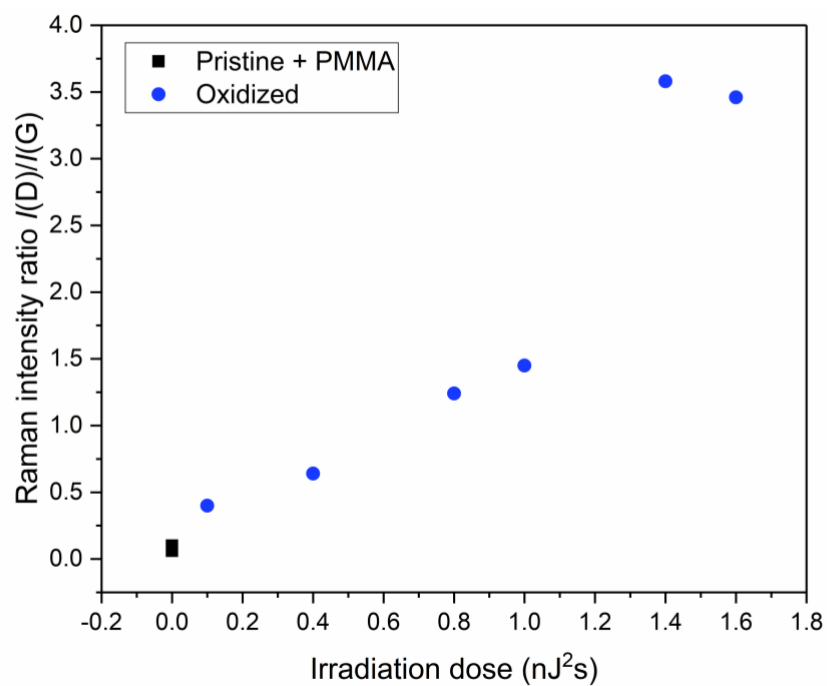
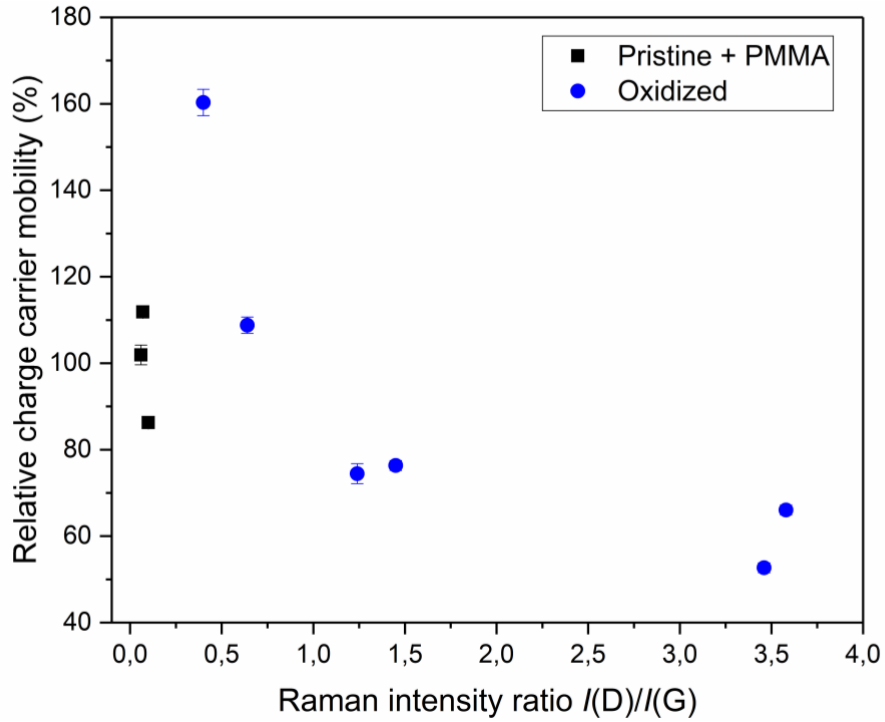


Fig S1: The dependency of the $I(D)/I(G)$ ratio on the laser irradiation dosage used in 2PO with the reported other parameters. Notice that this graph is only valid for this work and cannot be generalized, as the oxidation level achieved with 2PO depends also on other conditions. (See chapter 2.2 of the article for the detailed list of parameters used)

Table S1: Fitting parameters of the Raman spectra for all the samples. Lorentzian peak shape was used for all the bands

Sample $I(D)/I(G)$ ratio		0.06	0.07	0.10	0.40	0.64	1.24	1.45	3.46	3.58
Peak	Parameter									
Laser dose (nJ²s)		0.0	0.0	0.0	0.1	0.4	0.8	1.0	1.6	1.4
D	Location	1345	1344	1343	1350	1348	1346	1346	1344	1344
	FWHM	26.93	40.00	40.00	23.18	22.56	21.06	20.16	30.30	20.69
	Area	1499	2040	2412	7891	11769	19446	17674	76415	45617
	Height	35.44	32.48	38.38	216.72	332.06	587.90	558.11	1605.57	1403.57
G	Location	1591	1583	1584	1602	1599	1596	1588	1590	1586
	FWHM	12.37	16.81	20.43	11.61	12.75	14.23	18.28	33.16	21.99
	Area	11847	11441	12323	9844	10438	10603	11081	24178	13552
	Height	609.75	433.25	383.98	539.64	521.26	474.32	385.98	464.12	392.40
D'	Location	1618	1625	1624	1627	1627	1625	1627	1620	1623
	FWHM	9.30	6.31	20.00	4.20	8.90	6.70	11.26	17.54	13.58
	Area	352	139	661	103	422	489	1096	6815	3647
	Height	24.06	14.03	21.04	15.54	30.18	46.49	61.99	247.31	170.98
2D	Location	2683	2675	2678	2692	2689	2686	2685	2682	2680
	FWHM	31.23	33.00	32.17	32.47	35.68	34.47	31.76	42.36	30.89
	Area	28658	36463	30265	17030	21153	20741	27539	19579	29311
	Height	584.18	703.33	598.90	333.90	377.39	383.10	552.05	294.27	604.10
Sensitivity (mV/pH)		20.0 ± 0.9	21 ± 2	22 ± 2	20 ± 4	19 ± 2	22 ± 3	25 ± 3	24 ± 1	25 ± 2

a)



b)

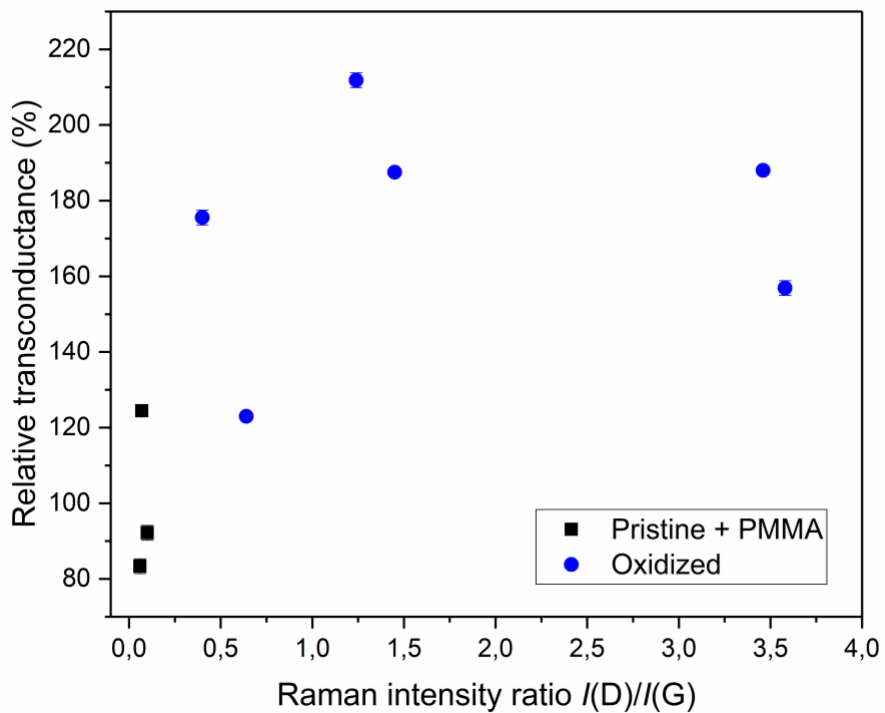
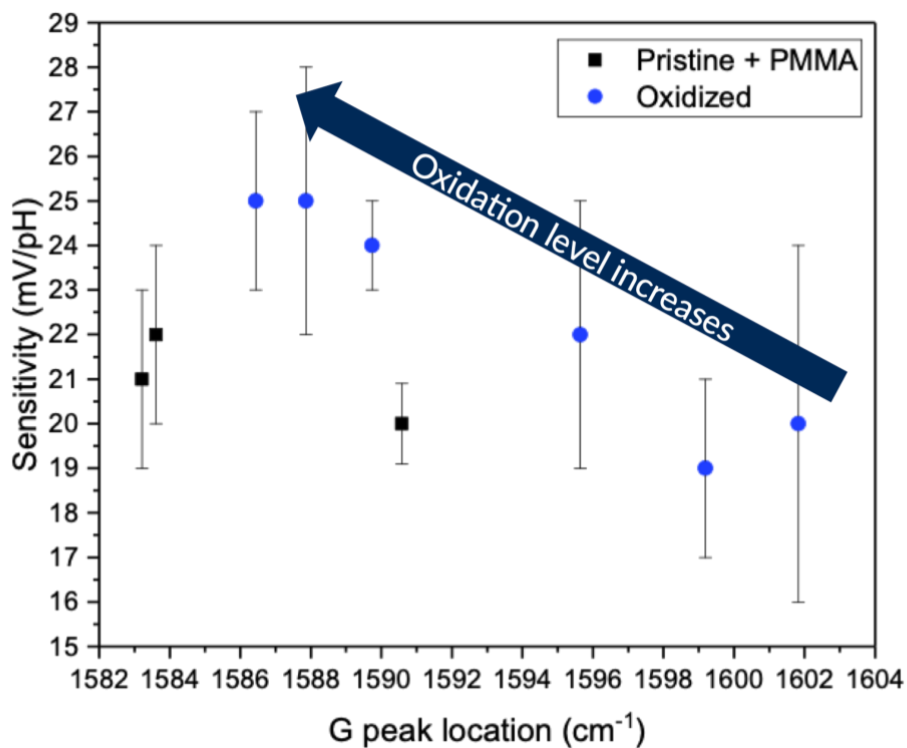


Fig S2: a) The maximum relative transconductance and b) the corresponding relative charge carrier mobility when $U_G < U_{Dirac}$ vs. Raman intensity ratio $I(D)/I(G)$, which reflects the oxidation level. Each point is an average of all the repetitions measured with an individual device and the error bars represent the standard error of the mean. Values have been normalized so that the average of the pristine samples is 100 %.

a)



b)

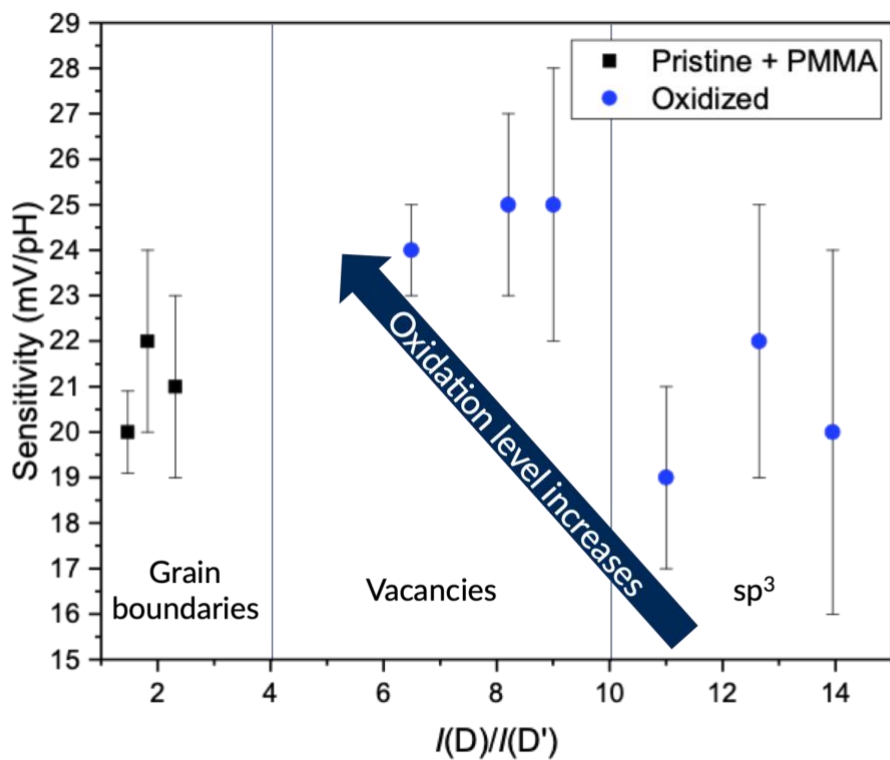


Fig S3: a) Sensitivity vs. G peak location and b) sensitivity vs. $I(D)/I(D')$. The trend of the oxidation level for the oxidized samples is indicated with an arrow. The regions where different defect types are dominant have been named and approximately separated with lines.³⁻⁵

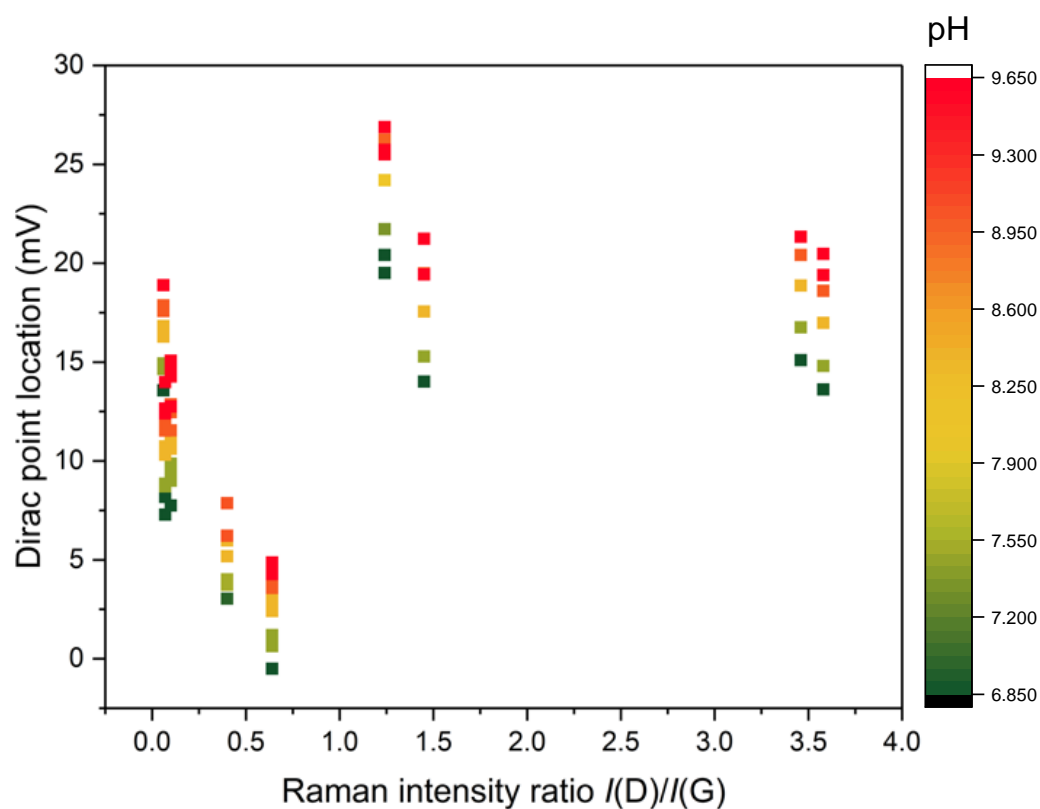
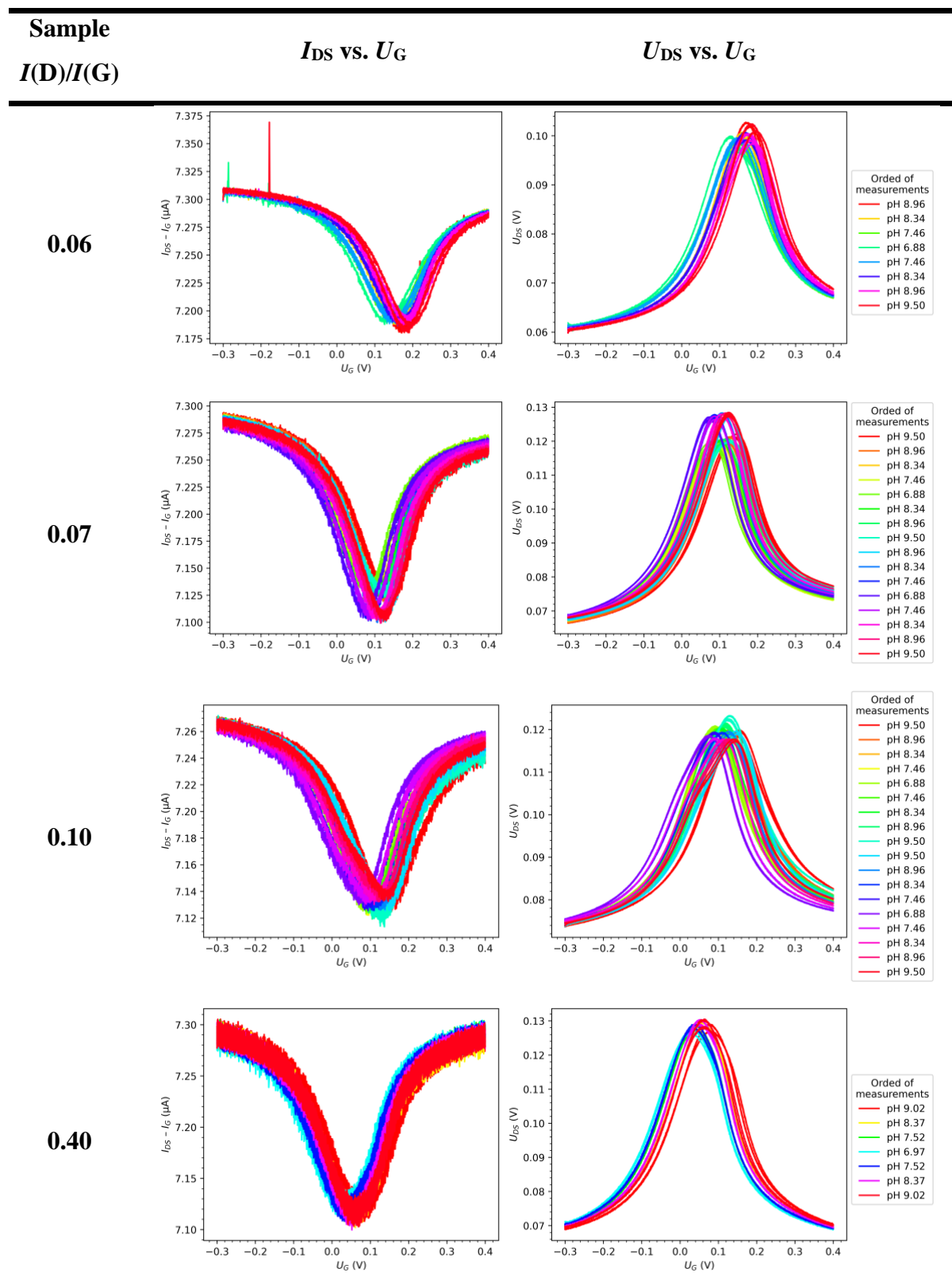


Fig S4: Scatter plot of all the data used for the pH sensitivity measurements in this publication with the Dirac point location plotted against the $I(D)/I(G)$ ratio at different pH.

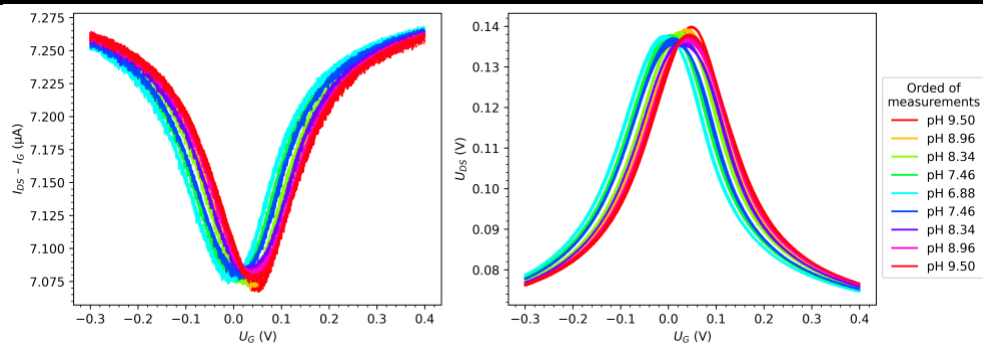
Table S2: Sample history for all the samples used and related identification tags. The letter in the ID refers to the chip on which the device was on and the number to the device

Sample $I(D)/I(G)$	ID
0.06	A1
0.07	B1
0.10	B2
0.40	C1
0.64	D1
1.24	E1
1.45	A1
3.46	B2
3.58	A2

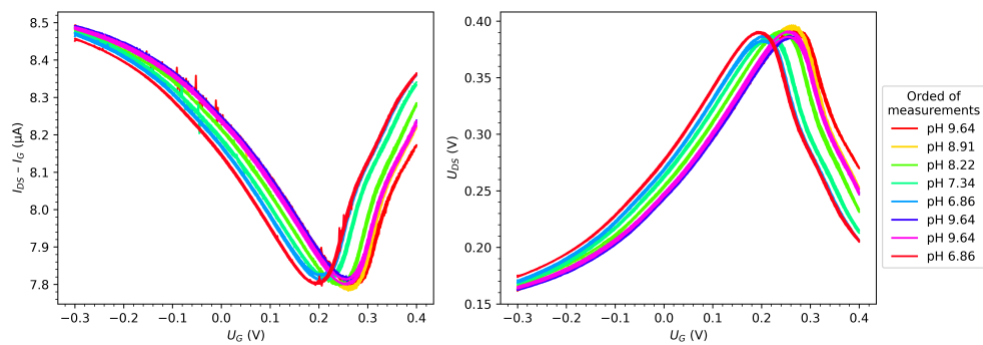
Table S3: Raw data for all of the pH measurements used in this study



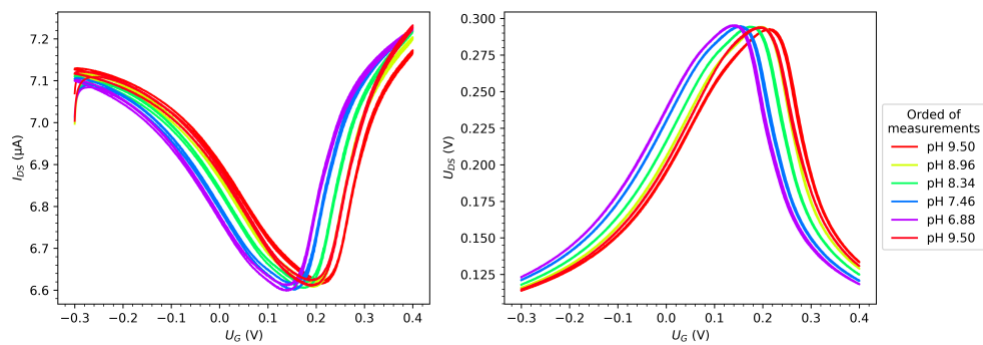
0.64



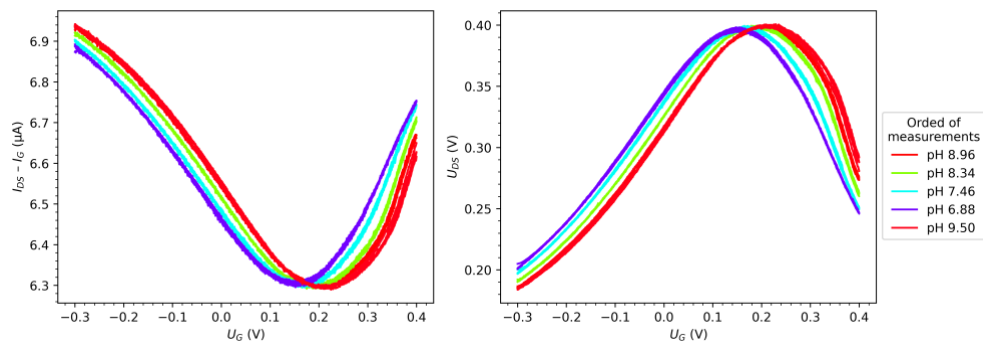
1.24



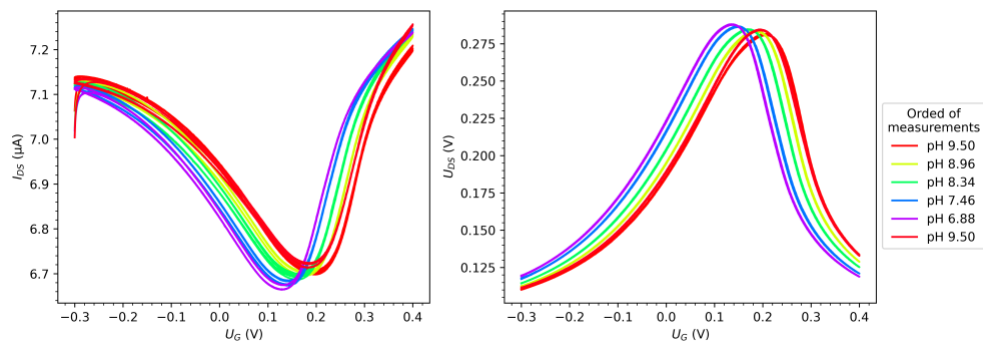
1.45



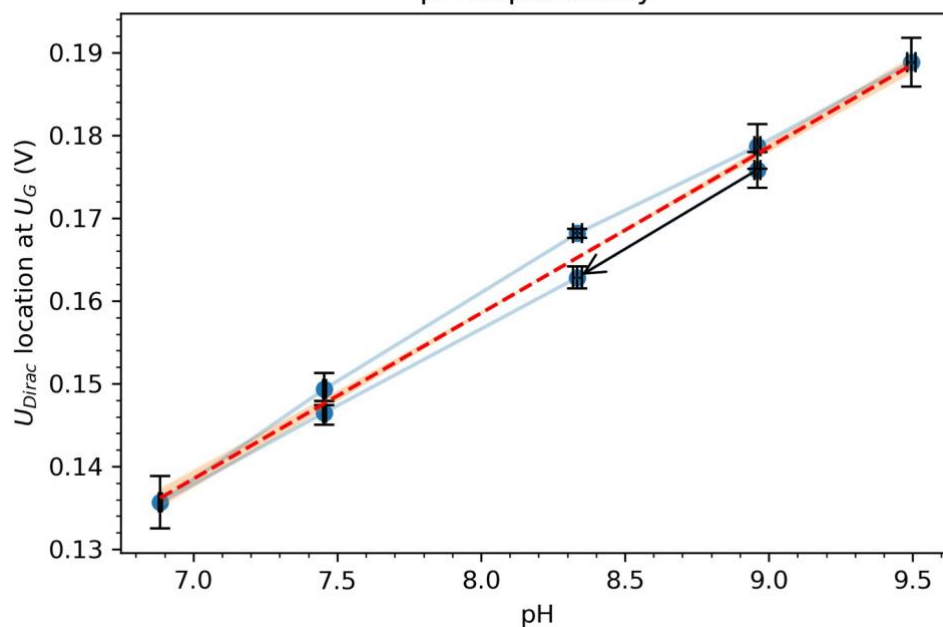
3.46



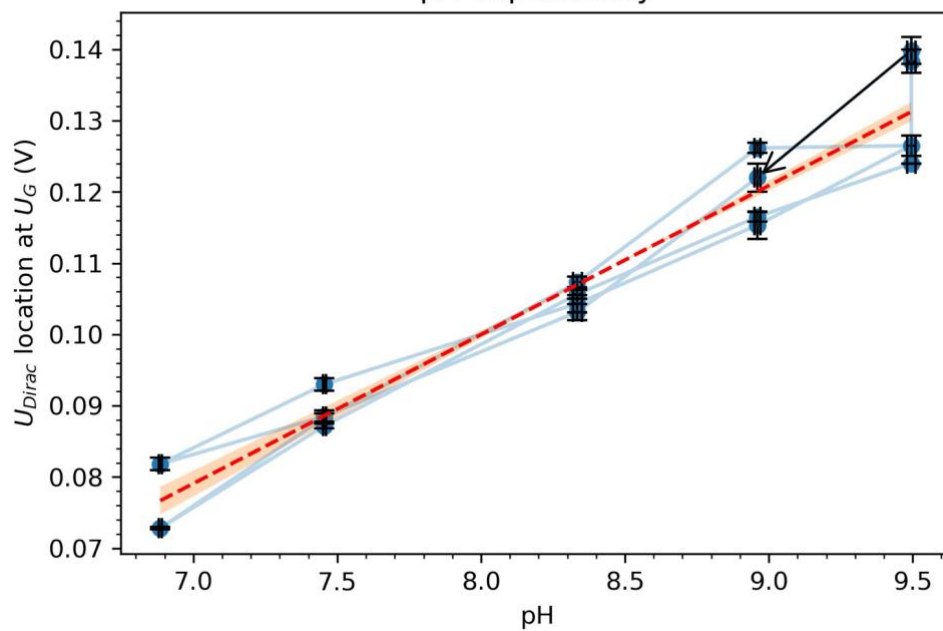
3.58



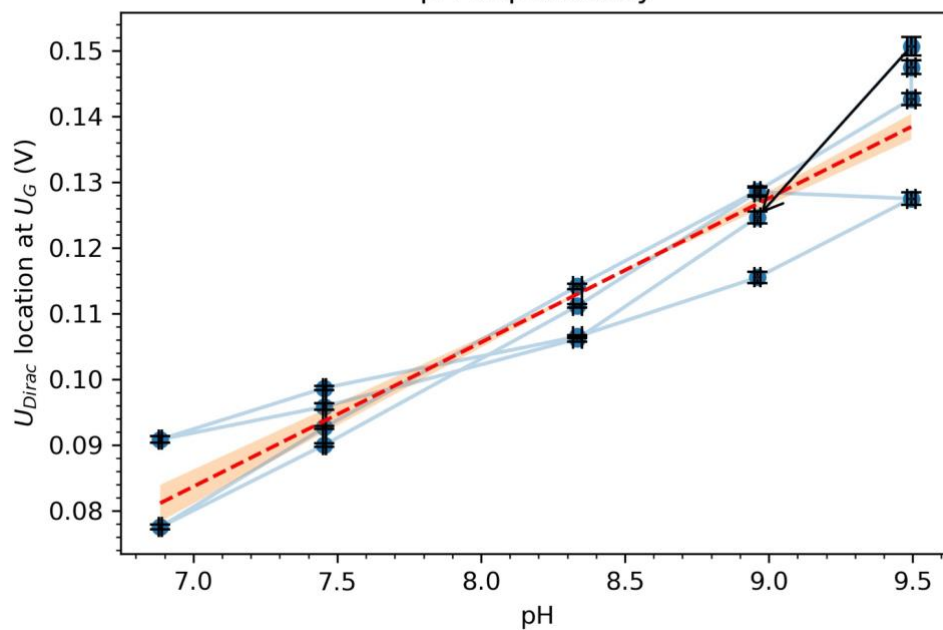
Sample 0.06 $y = (-0.002 \pm 0.007) + (0.02 \pm 0.0009)x$
 $R^2 = 0.989829$
pH dependency



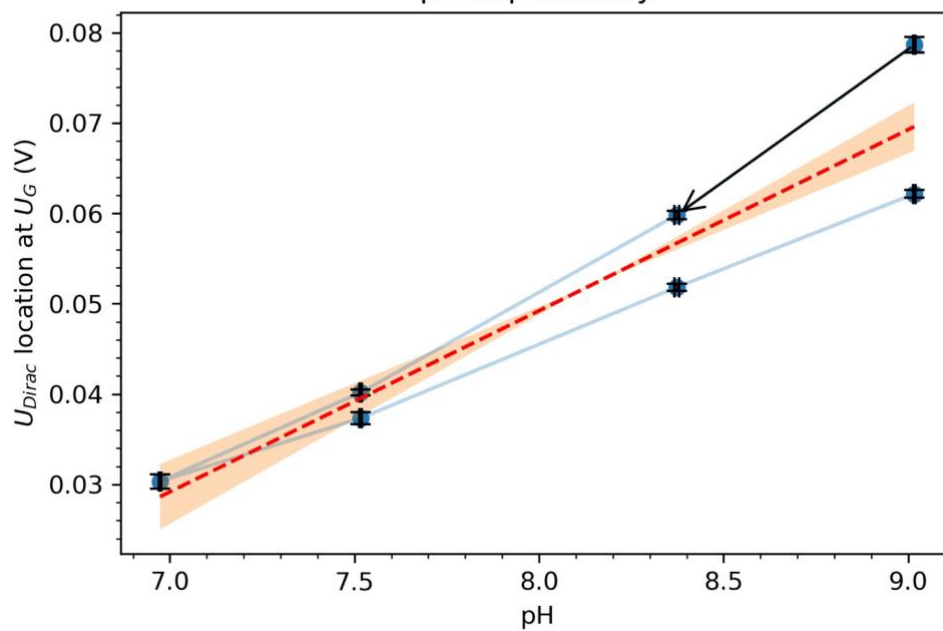
Sample 0.07 $y = (-0.07 \pm 0.02) + (0.021 \pm 0.002)x$
 $R^2 = 0.944998$
pH dependency



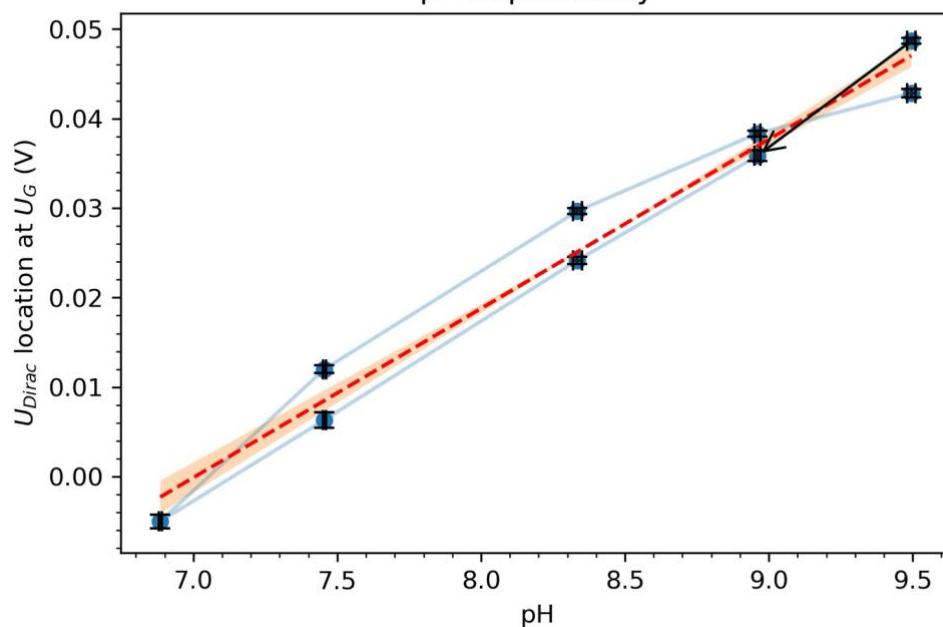
Sample 0.10 $y = (-0.07 \pm 0.02) + (0.022 \pm 0.002)x$
 $R^2 = 0.901343$
pH dependency



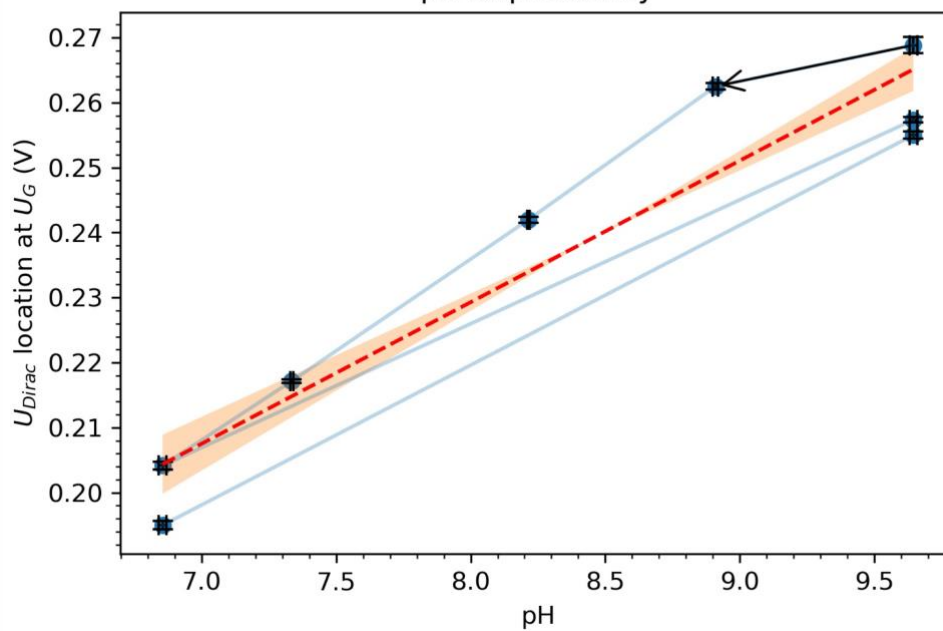
Sample 0.40 $y = (-0.11 \pm 0.03) + (0.02 \pm 0.004)x$
 $R^2 = 0.894569$
pH dependency

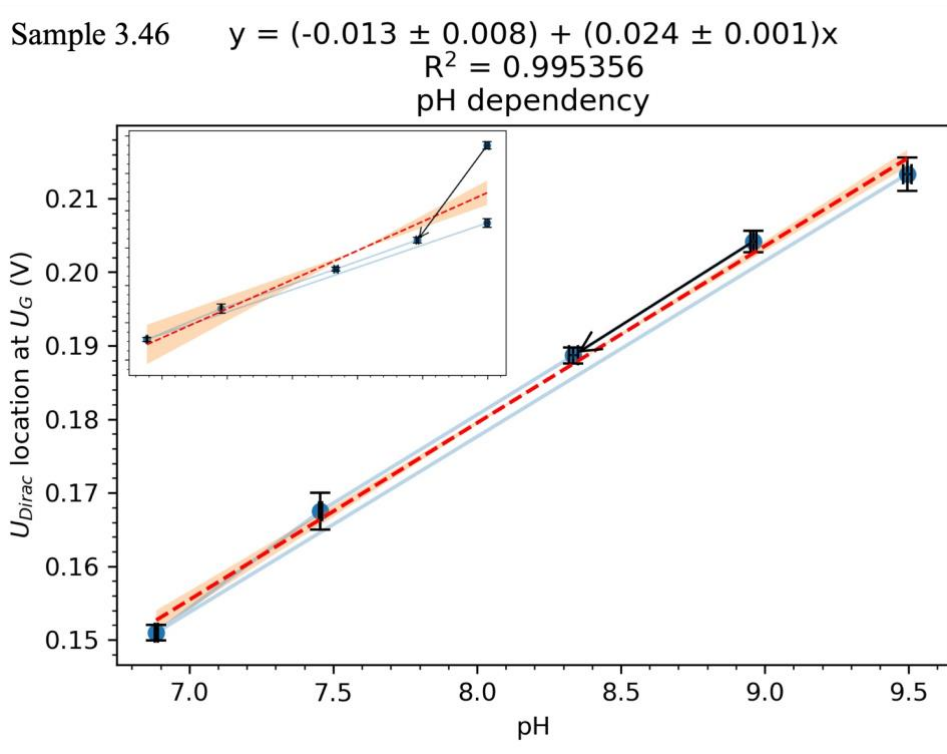
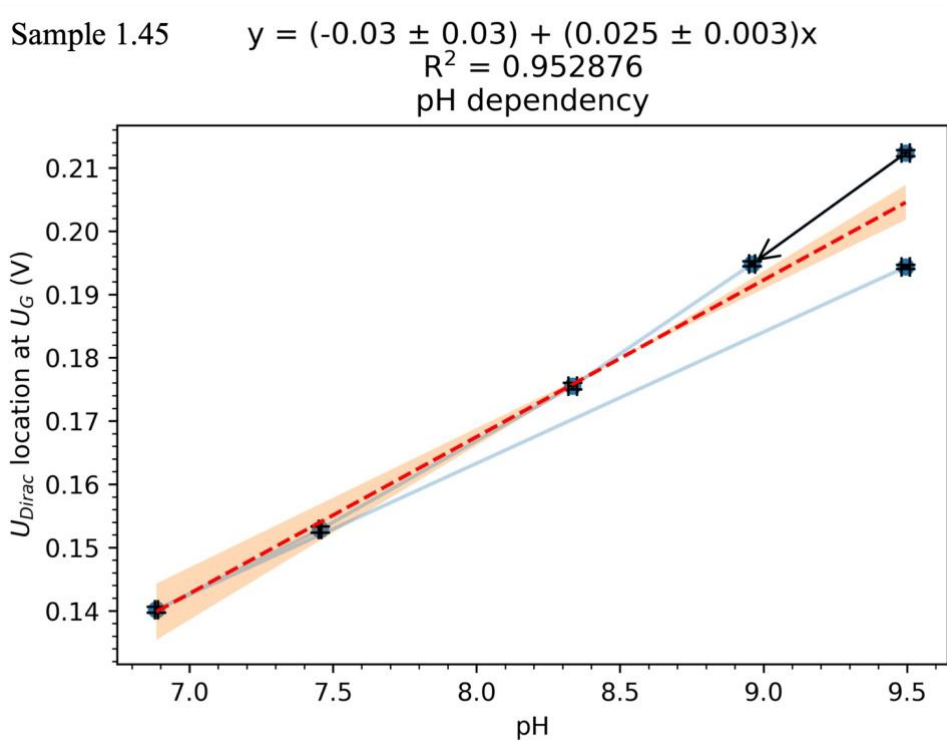


Sample 0.64 $y = (-0.13 \pm 0.01) + (0.019 \pm 0.002)x$
 $R^2 = 0.973349$
pH dependency



Sample 1.24 $y = (0.06 \pm 0.03) + (0.022 \pm 0.003)x$
 $R^2 = 0.909213$
pH dependency





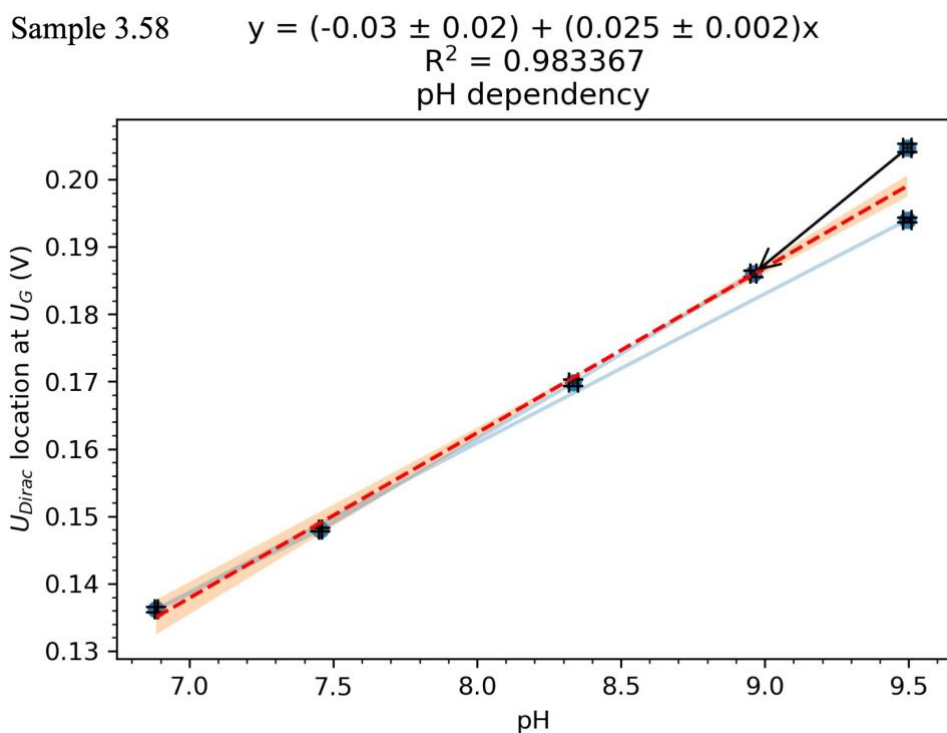


Fig S5: All the pH dependency fittings with their corresponding fitted linear equation and parameters. The sample is identified with their $I(D)/I(G)$ ratio in the top left corner of each graph. Sample 3.46 has an inset showing the left-out outlier point. The error bars represent the standard error of the mean for all the $I_{DS}-U_G$ scans at the one pH value. The arrow indicates the measurement order by pointing from the first point to the second, followed by the line in order.

Supplementary references

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