Electronic Supplementary Information

Optical imaging of the potential distribution at transparent electrode/solution interfaces

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Experimental Section

Fabrication of single-layer graphene electrode on glass: Single-layer graphene was synthesized and transferred to glass according to previous report.[46] In brief, single-layer graphene was synthesized by chemical vapor deposition (CVD) technique on copper foils as a catalyst and substrate as reported previously. The synthesis is carried out at 1050 ℃ with hydrogen (50 sccm) and methane (20 sccm) mixed gas as sources. As-deposited graphene was transferred onto a glass slide (25mm×75mm) using a standard wet method with PMMA (average Mw ~996,000, dissolved in ethyl lactate with a concentration of 4 wt%) as removable adhesive layer. The graphene ribbon electrode was further fabricated by selectively etching the graphene under oxygen plasma coupled with a photomask.

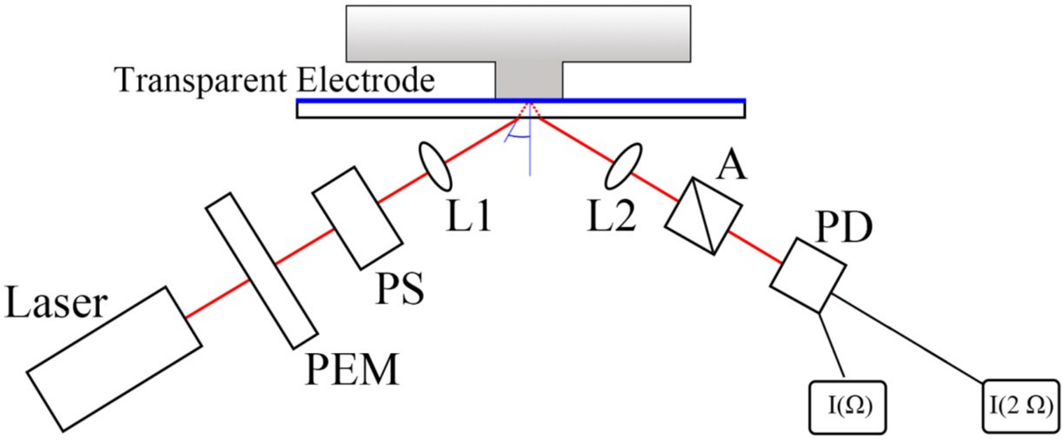

Fig. S1. Cartoon showing the home-built OIRD instrumental setup. A 632.8 nm laser from He-Ne laser passed through a polarizer is introduced to a photoelastic modulator (PEM), which modulates the polarization of the laser between p and s at a frequency of 50 kHz. The phase difference between p- and s- polarized components in the incident laser is adjusted by a phase shifter. The light is focused by an optical lens (L1) to the surface of the transparent electrode at an incident angle of 60°. The phase and amplitude of reflected light are analyzed by using a polarization analyzer (A) coupled with photodiode detector (PD). Two OIRD signals, namely I(Ω) and I(2Ω) are collected by two lock-in amplifiers. The transparent electrode and whole electrochemical cell was mounted on a motorized stage, which enables scanning of the whole surface of the electrode at a spatial resolution of 4 μm.

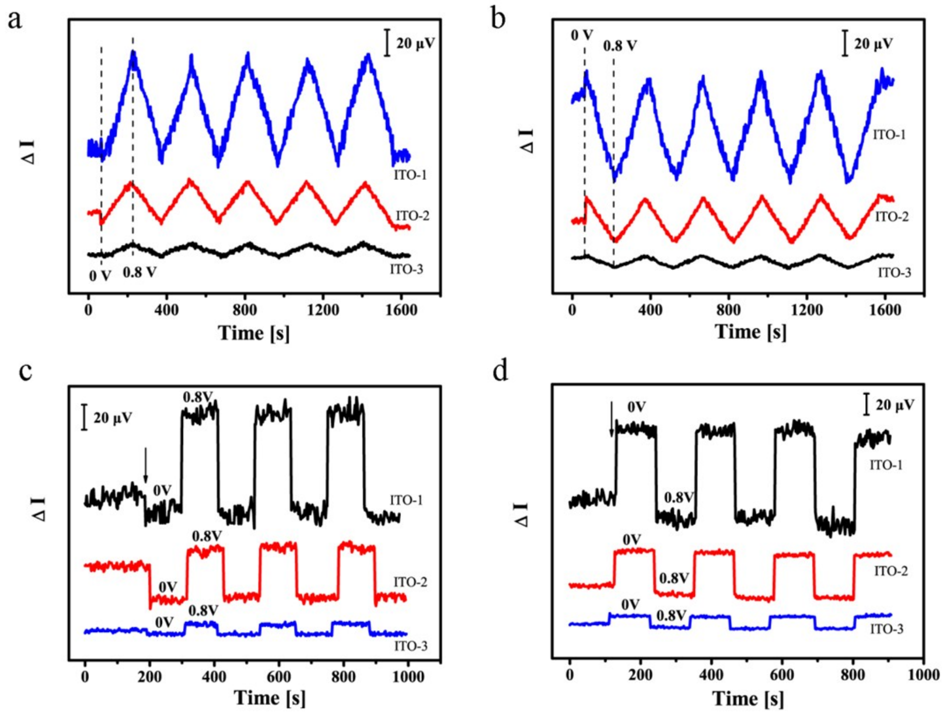

Fig. S2. Simultaneous OIRD I (Ω) signals (a, c) and I (2Ω) signals (b, d) recorded on three types of ITO electrodes with different conducting layer thickness upon continuous CV scanning from 0 to 0.8 V at a scanning rate of 5 mV s^{-1} (a, b) and potential step from 0 to 0.8 V (c, d) in 3 M KCl solution. ITO thicknesses and conductivities of ITO-1, ITO-2 and ITO-3 are ~400 nm, 3±1Ω/□; 210 nm, 6±1Ω/□; and 120 nm, and 14±1Ω/□, respectively.

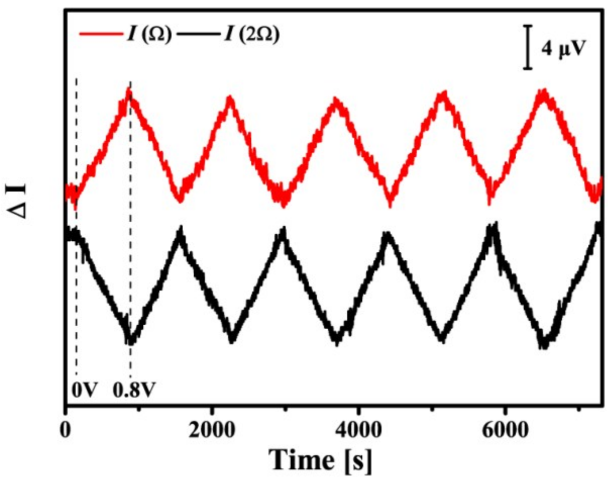

Fig. S3. Simultaneous OIRD signals recorded on ITO-3 electrode during continuous CV scanning from 0 to 0.8 V at a scanning rate of 1 mV s^{-1} in 3 M KCl solution.

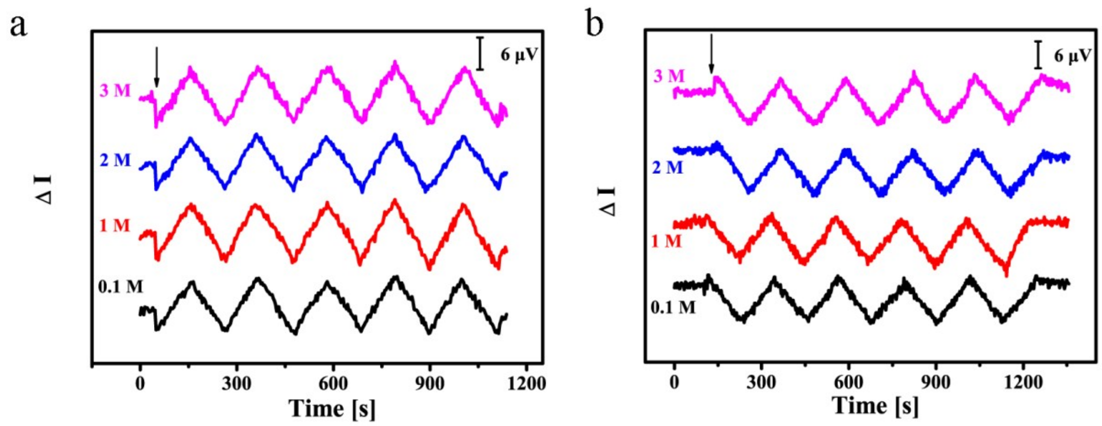

Fig. S4. Simultaneous OIRD I (Ω) (a) and I (2Ω) signal (b) recorded on ITO-3 electrode during continuous CV scanning from 0 to 0.8 V in KCl solution with different concentrations at a scanning rate of 5 mV s^{-1}.

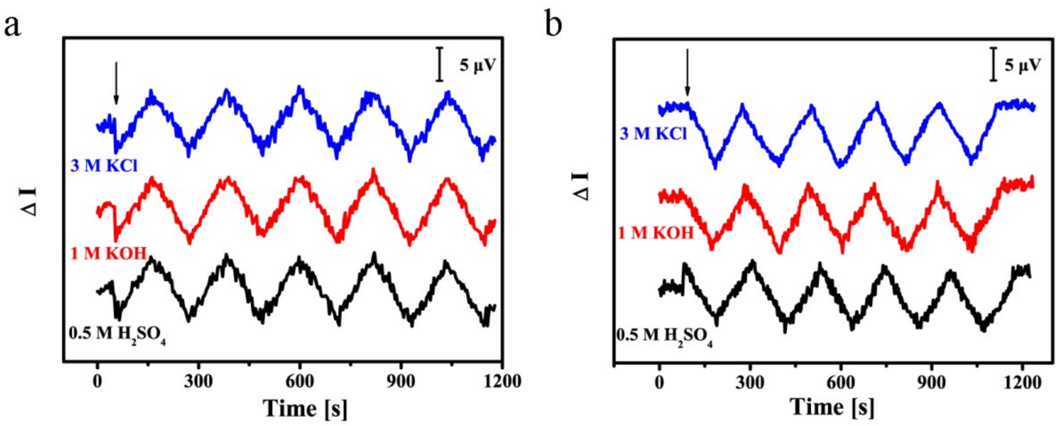

Fig. S5. Simultaneous OIRD I (Ω) (a) and I (2Ω) signals (b) recorded on ITO-3 electrode during continuous CV scanning from 0 to 0.8 V at a scanning rate of 5 mV s^{-1} in different solutions.

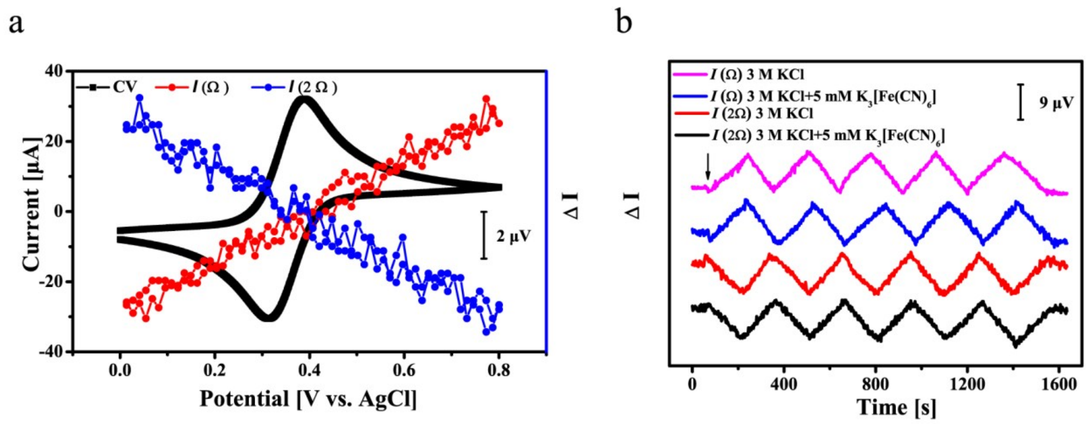

Fig. S6. (a) Simultaneous OIRD signals and current recorded on ITO-3 electrode during potential scanning from 0 to 0.8 V at a scanning rate of 5 mV s^{-1} in 5 mM $K_4Fe(CN)_6$/ $K_3Fe(CN)_6$ with 3 M KCl; (b) Comparison of OIRD signals collected on ITO-3 electrode during continuous CV cycling from 0 to 0.8 V at a scanning rate of 5 mV s^{-1} in 3 M KCl with and w/o 5 mM $K_4Fe(CN)_6$/ $K_3Fe(CN)_6$.

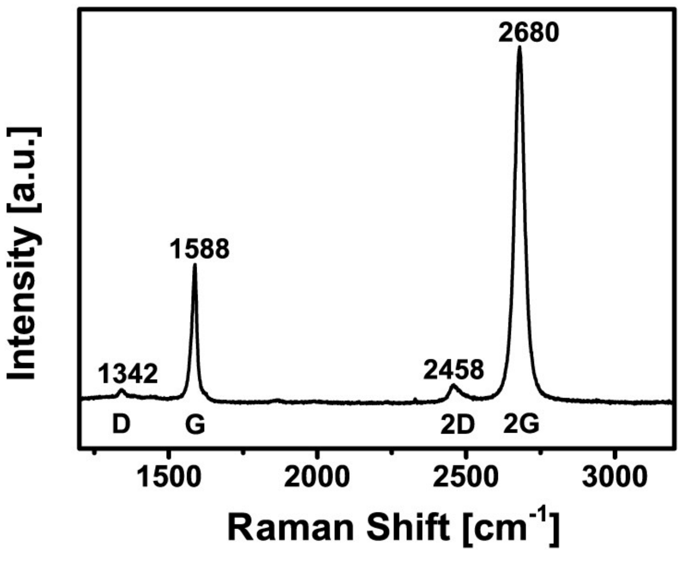

Fig. S7. Raman spectrum of single-layer graphene transferred to glass surface.

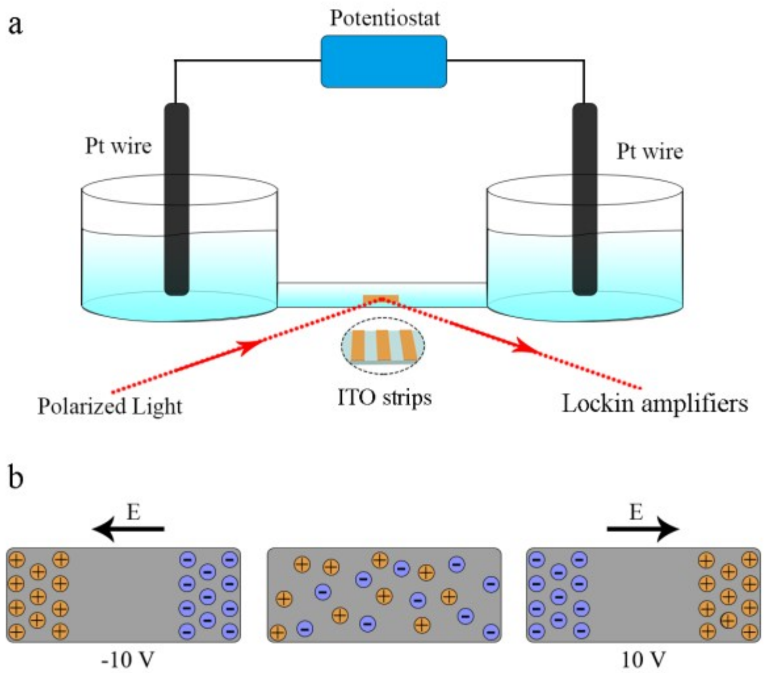

Fig. S8. (a) Scheme illustrating the OIRD setup for imaging of the potential distribution on a bipolar system; (b) Potential gradient is generated on ITO strip coupled with redistribution of electron density when a voltage is applied to two Pt driving electrodes.

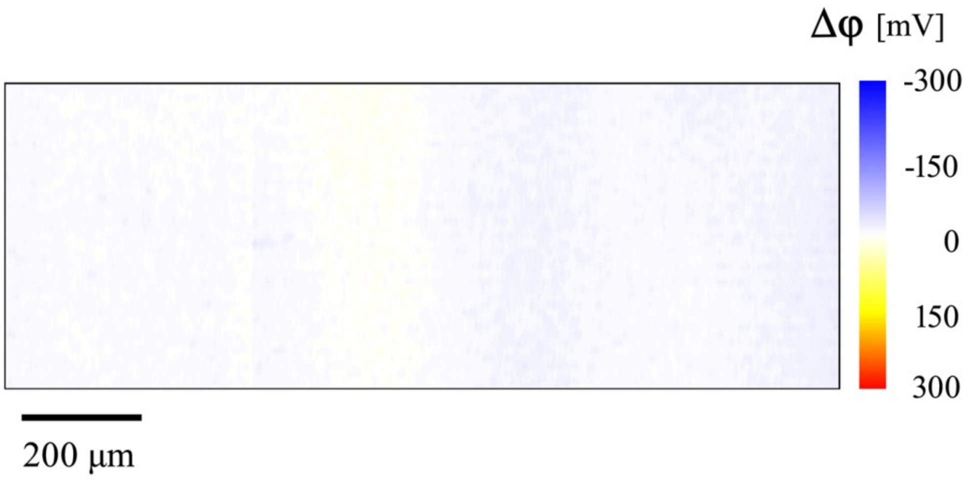

Fig. S9. Potential distribution image converted from OIRD I(Ω) images of a ITO-1 strip electrode in a bipolar system without a driving voltage.

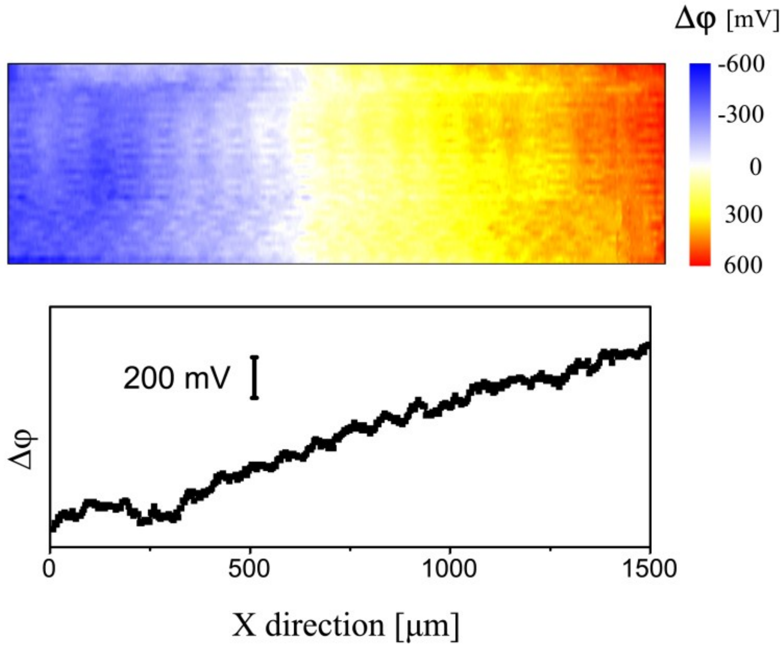

Fig. S10. Potential distribution image and corresponding potential profile converted from OIRD I(2Ω) images of a ITO-1 strip electrode in a bipolar system at a driving voltage of 10 V.

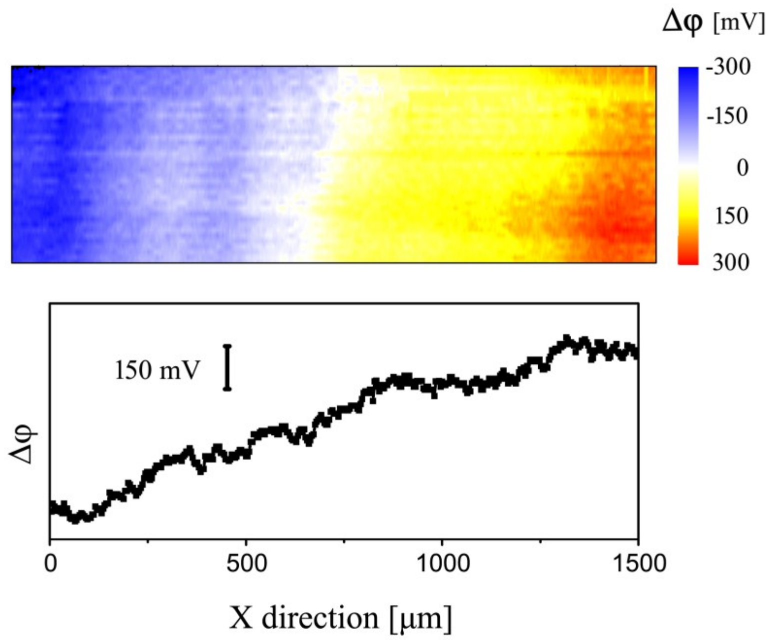

Fig. S11. Potential distribution image and corresponding potential profile converted from OIRD I(Ω) images of a ITO-1 strip electrode in a bipolar system at a driving voltage of 5 V.

Table S1. OIRD sensitivity and S/N ratio on five types of transparent electrodes. [a)]

Electrode	Thickness [b)] (nm)	Conductivity [b)] (ohm/☐)	I(Ω)				I(2Ω)			
			ΔI (μV)	SD (μV) [c)]	S/N	Sensitivity (μV/V) [d)]	ΔI (μV)	SD (μV)	S/N	Sensitivity (μV/V)
ITO-1	400	3±1	81.36	3.22	25.27	101.70	79.63	2.25	35.39	99.54
ITO-2	210	6±1	39.87	2.21	18.04	49.84	41.22	1.98	20.82	51.53
ITO-3	120	14±1	8.96	0.84	10.67	11.20	10.27	0.74	13.88	12.84
FTO	350	6±1	9.29	0.85	10.93	11.61	9.53	0.91	10.47	11.91
Graphene	N.A.	N.A.	8.79	1.05	8.37	10.99	10.55	1.00	10.55	13.19

[a)] Derived from multi-potential step response from 0 to 0.8 V for ITO-1 as in Fig. 1e while from potential step response from 0 to 0.8 V for other four electrodes; [b)] data provided by the suppliers; [c)] standard derivation at 0 V; [d)] sensitivity defined as the amplitude of OIRD signal upon 1 V potential change.