Electronic Supplementary Information

In situ FTIR and Raman spectroelectrochemical characterization of graphene oxide upon electrochemical reduction in organic solvents

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For the IR spectroelectrochemical measurements a 12.5 mm diameter ZnSe hemisphere crystal was used as working electrode. First the ZnSe crystal was polished using 1/4 µm diamond paste and polishing cloth. The polished crystal was thereafter cleaned with oxygen (3 min) and hydrogen (1 min) plasma using the Harrick (PDC-3XG) plasma cleaner. For electrical contact a gold layer (40 nm) was evaporated on the cleaned crystal surface using the Edwards E306A coating systems. As shown in Figure S1 a 2 mm optical window was left in the center of the crystal. The crystal surface was finally covered with a thin sputtered gold layer (<10 nm).

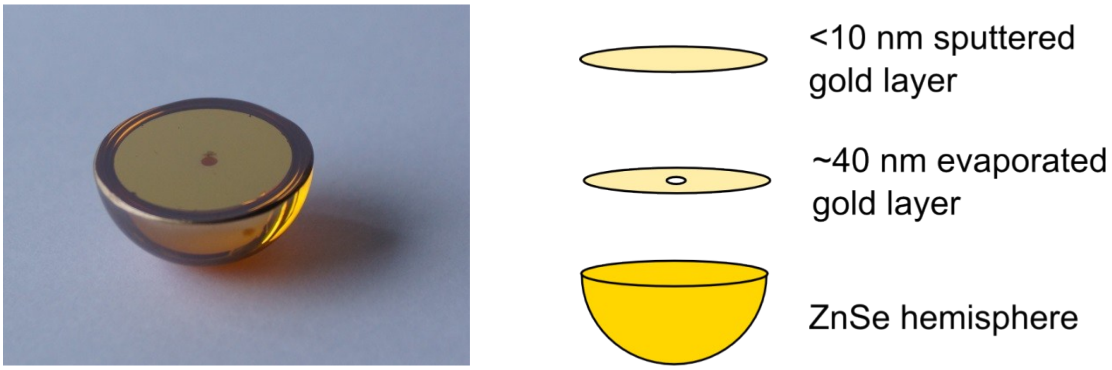

Fig. S1. Image (left) and schematics (right) of the ZnSe element used as working electrode for the IR spectroelectrochemical measurements.

The electrochemical cell was constructed from Teflon™. The cell was fitted with a Pt-wire counter electrode and a Ag/AgCl reference electrode as well as inlet and outlet for electrolyte solution (Fig. S2). The Au covered ZnSe crystal acted as a working electrode and sealing from the top-side of the cell was ensured using a Kalrez o-ring (3.99 mm diameter).

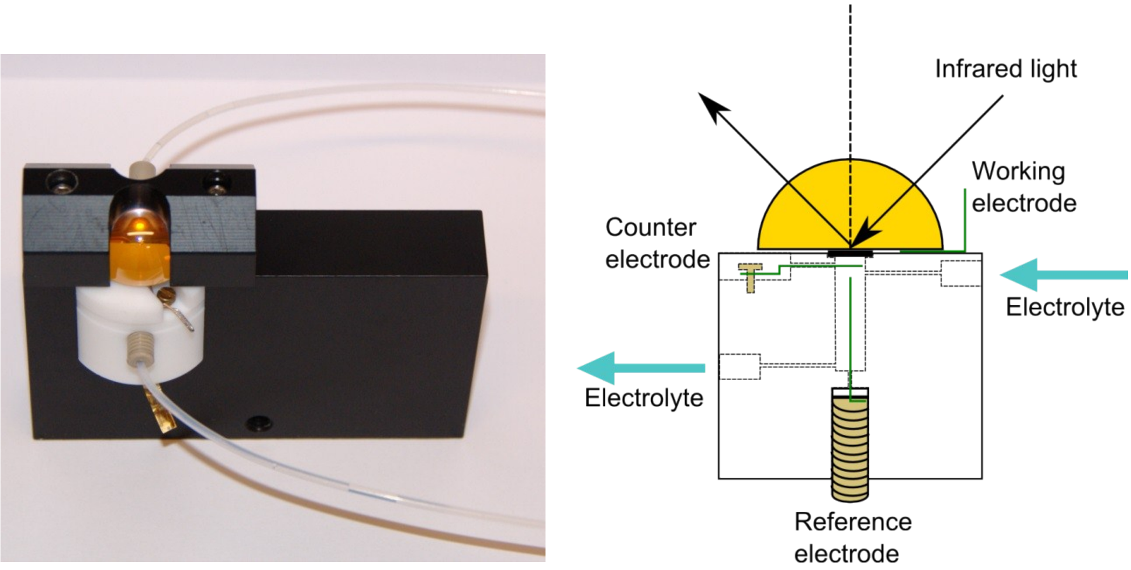

Fig. S2. Photograph (a) and schematics (b) of the electrochemical cell used for the in situ spectroelectrochemical FTIR measurements.

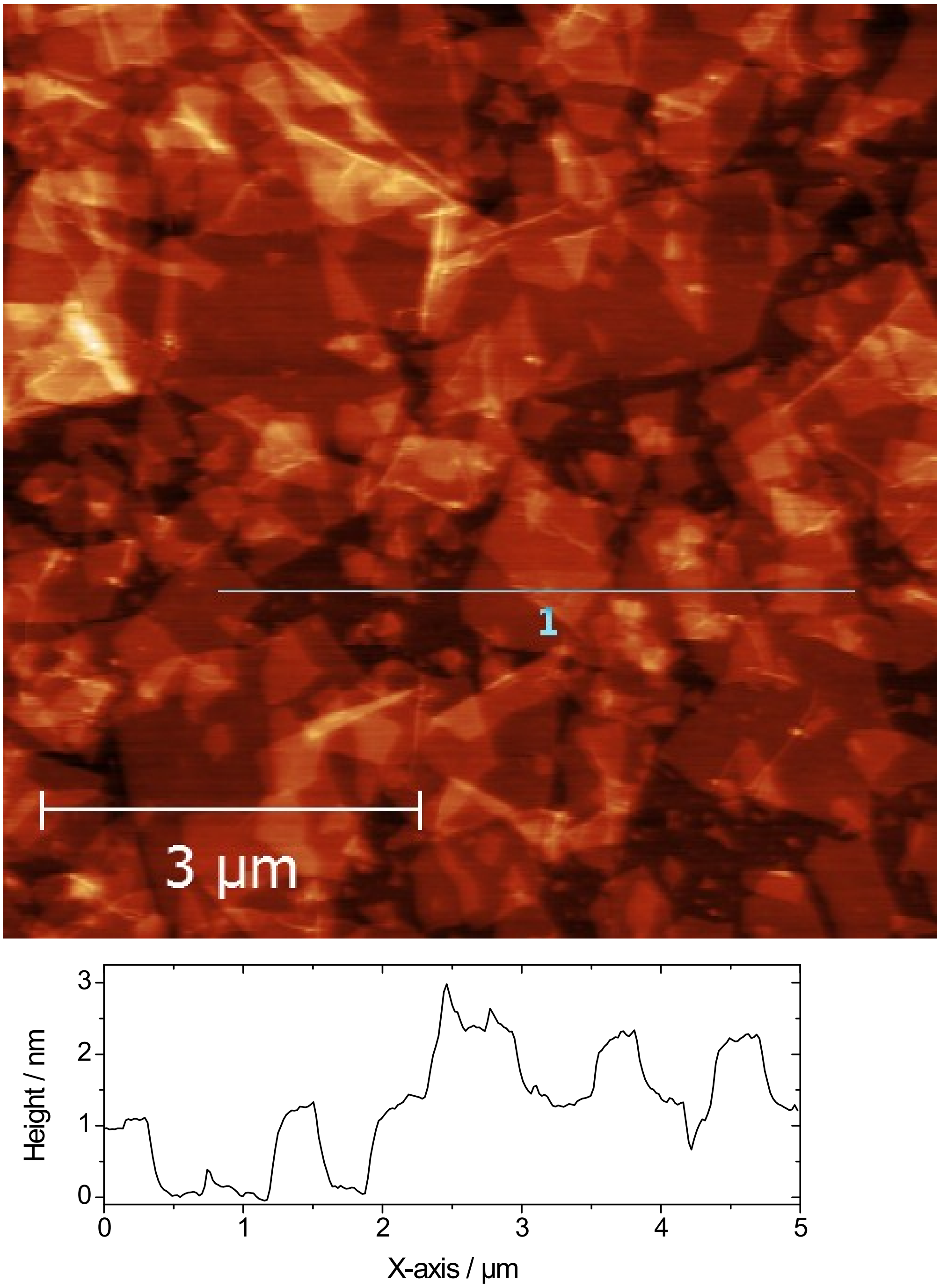

Fig. S3. AFM image of GO sheets (Au/MEA/GO) prepared according to a modified Hummers' method. The line in the AFM image indicates where the shown height profile has been recorded.

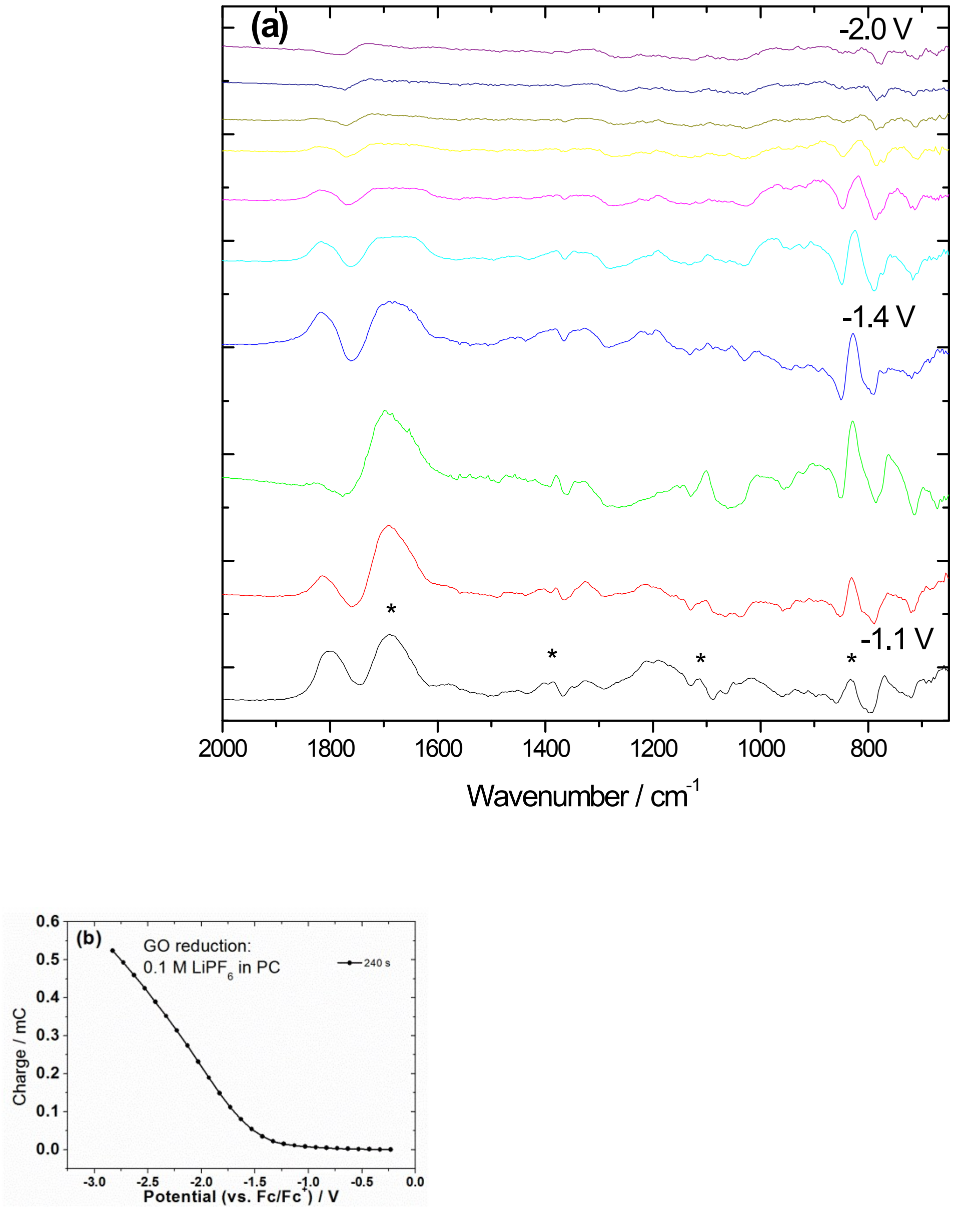

Figure S4. (a) In situ FTIR spectra recorded during electroreduction of GO in 0.1 M LiPF$_6$ (PC). For clarity the spectra are separated and only those gathered in the potential range -1.1 to -2.0 V are shown. The absorbance related to previous applied potential is used as reference. The main solvent peaks are market with an *. (b) The recorded change in reduction charge with potential during the spectroelectrochemical experiment.

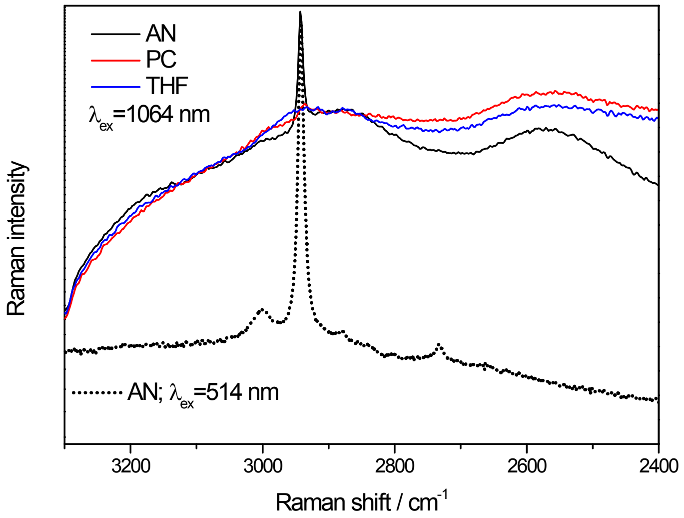

Figure S5. Raman spectrum of GO film on Au at -0.2/-0.4 V (vs. Fc/Fc$^+$) in AN, PC, and THF obtained in the range 2400-3300 cm^{-1}. Both the FT-Raman and dispersive technique was used and in the figure the dashed line represents the measurement in AN at 514 nm laser excitation.

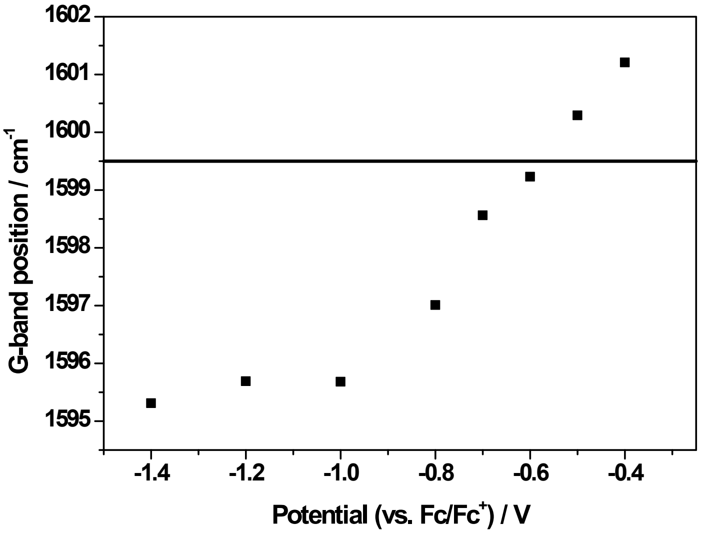

Figure S6. Changes in G band position during in situ electrochemical reduction of GO in aqueous solution. The horizontal line indicates the G band position at -0.4 V (vs. Fc/Fc$^+$) after reduction of the GO-film. [To facilitate comparison to our earlier results it is assumed that the potential of Ag/AgCl-reference electrode vs. Fc/Fc+-couple is ca. -0.44 V (+0.197 V Ag/AgCl vs. NHE and +0.64 V Fc/Fc+ vs. NHE)]

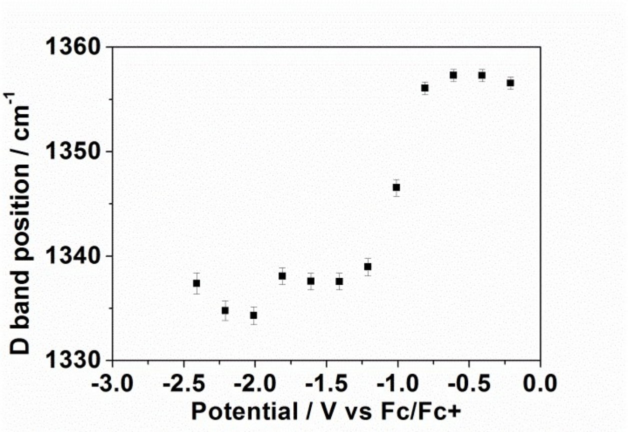

Figure S7. Changes in D band position during in situ electrochemical reduction of GO in AN using 514 nm laser excitation.