

The pH Dependence of Ultrafast Charge Dynamics in Graphene Oxide Dispersions

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S1. Atomic force microscope measurements.

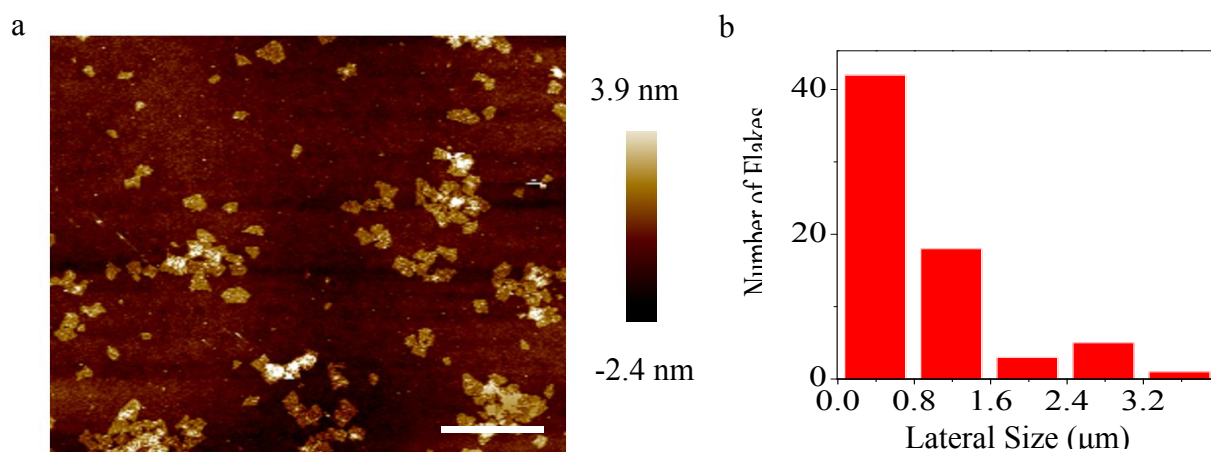


Figure S1 a) The AFM height image of as-prepared graphene oxide flakes deposited on SiO₂/Si substrate. Scale bar: 4 μm; b) the lateral size distribution of the graphene oxide flakes. The lateral sizes of flakes were estimated by taking the square root of the area of each flake measured with the NanoScope Analysis software.

S2. Absorbance and photoluminescence spectra & photoluminescence decay transients.

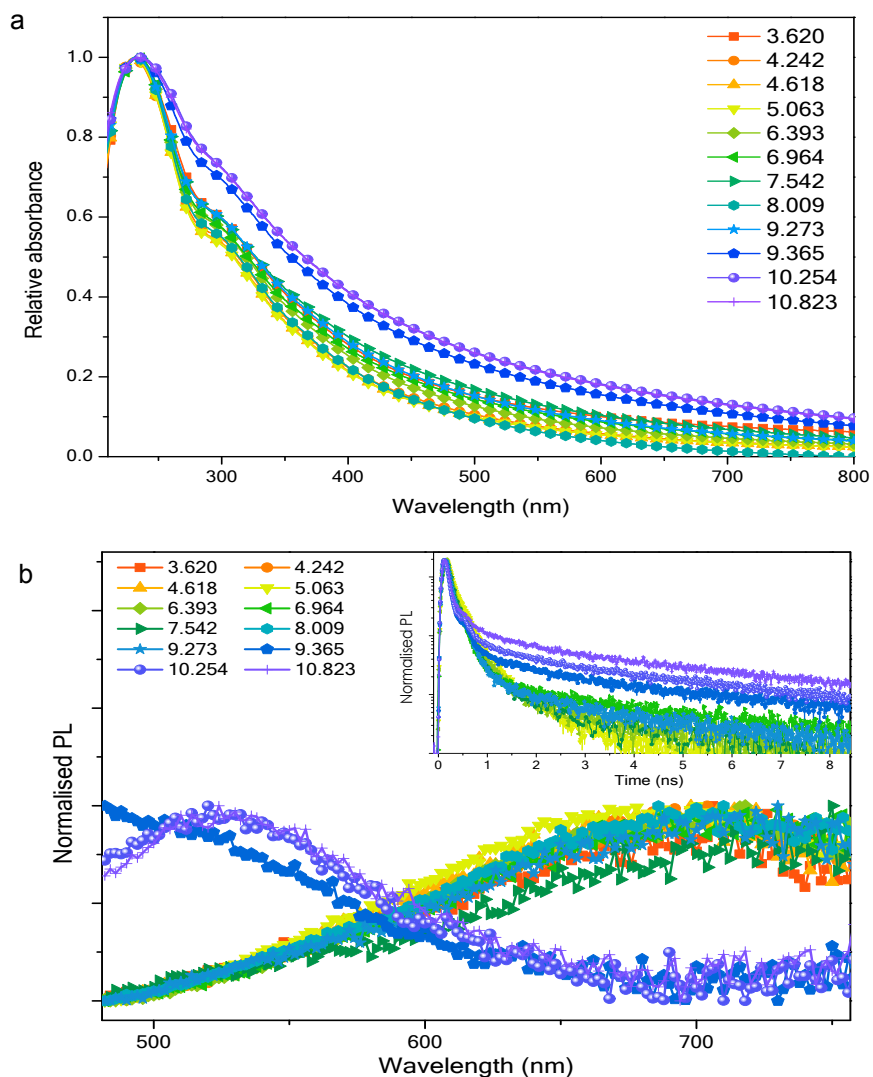


Figure S2 pH dependence of (a) absorbance spectra, normalised to the π - π^* absorbance feature at 230 nm, (b) normalised photoluminescence spectra for excitation at 400 nm, and (inset) photoluminescence transient measurements for emission at 580 nm.

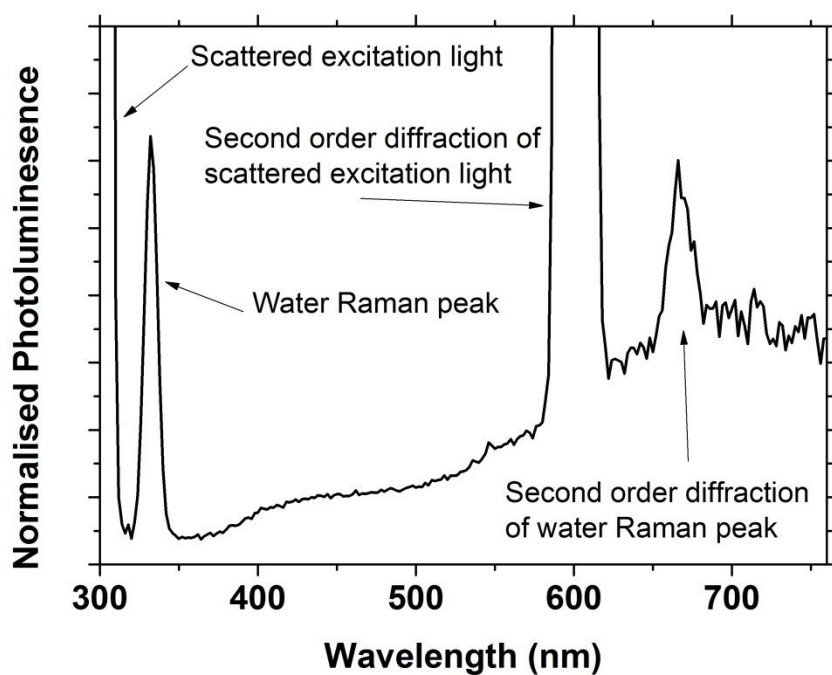


Figure S3. Spectrum for the pH=4.2 GO dispersions excited at 300 nm showing broad photoluminescence, and Rayleigh and Raman scattered excitation light that has undergone both first and second order diffraction within the spectrometer.

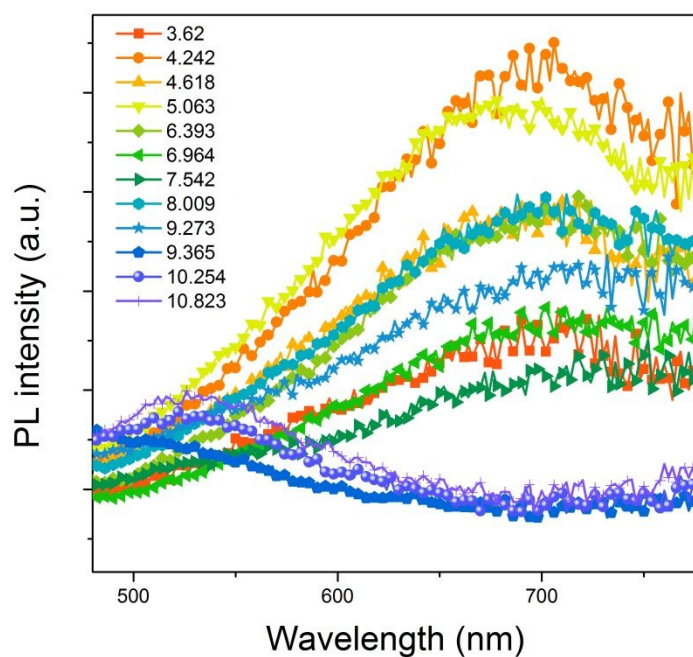


Figure S4. Steady-state photoluminescence of dispersions of GO at a range of pH levels, for excitation at 400 nm and without normalization.

S3. Excitation spectra.

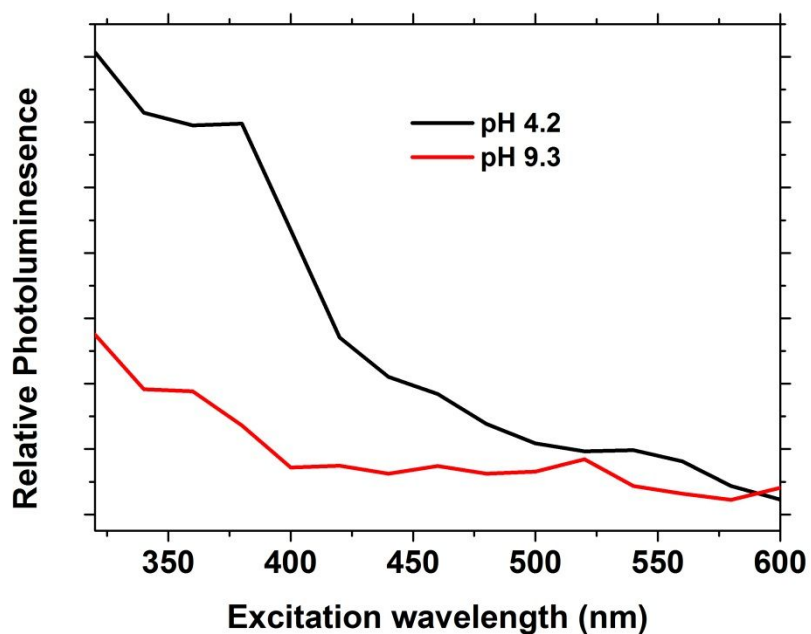


Figure S5. Excitation spectra for the pH 4.2 and pH 9.3 samples for a detection wavelength of 620 nm.

S4. Differential absorption spectra.

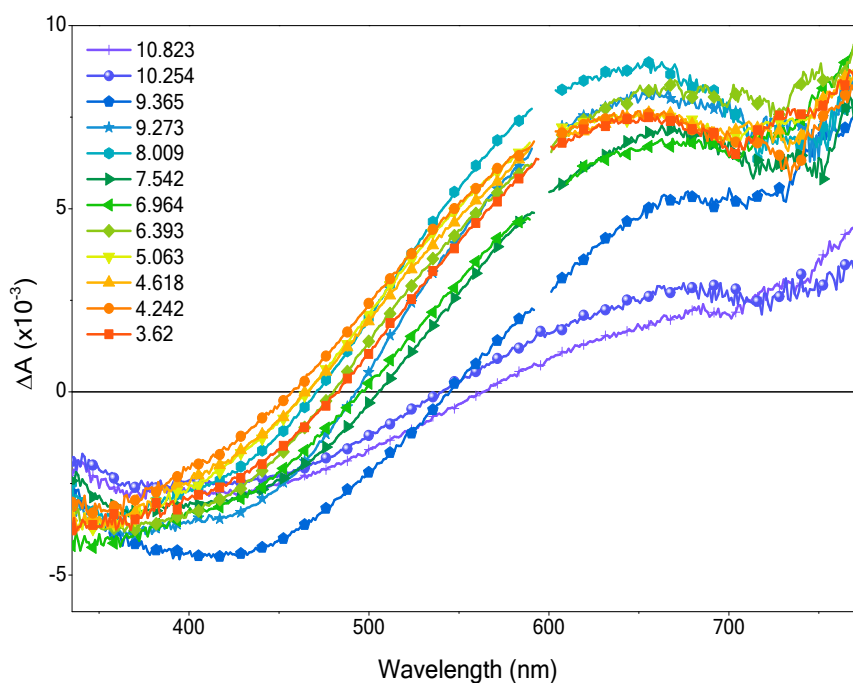


Figure S6 Differential absorption, ΔA , spectra between 330 – 750 nm following excitation at 300 nm. Each spectrum is taken at its respective maximum amplitude, around 2 ps after excitation.

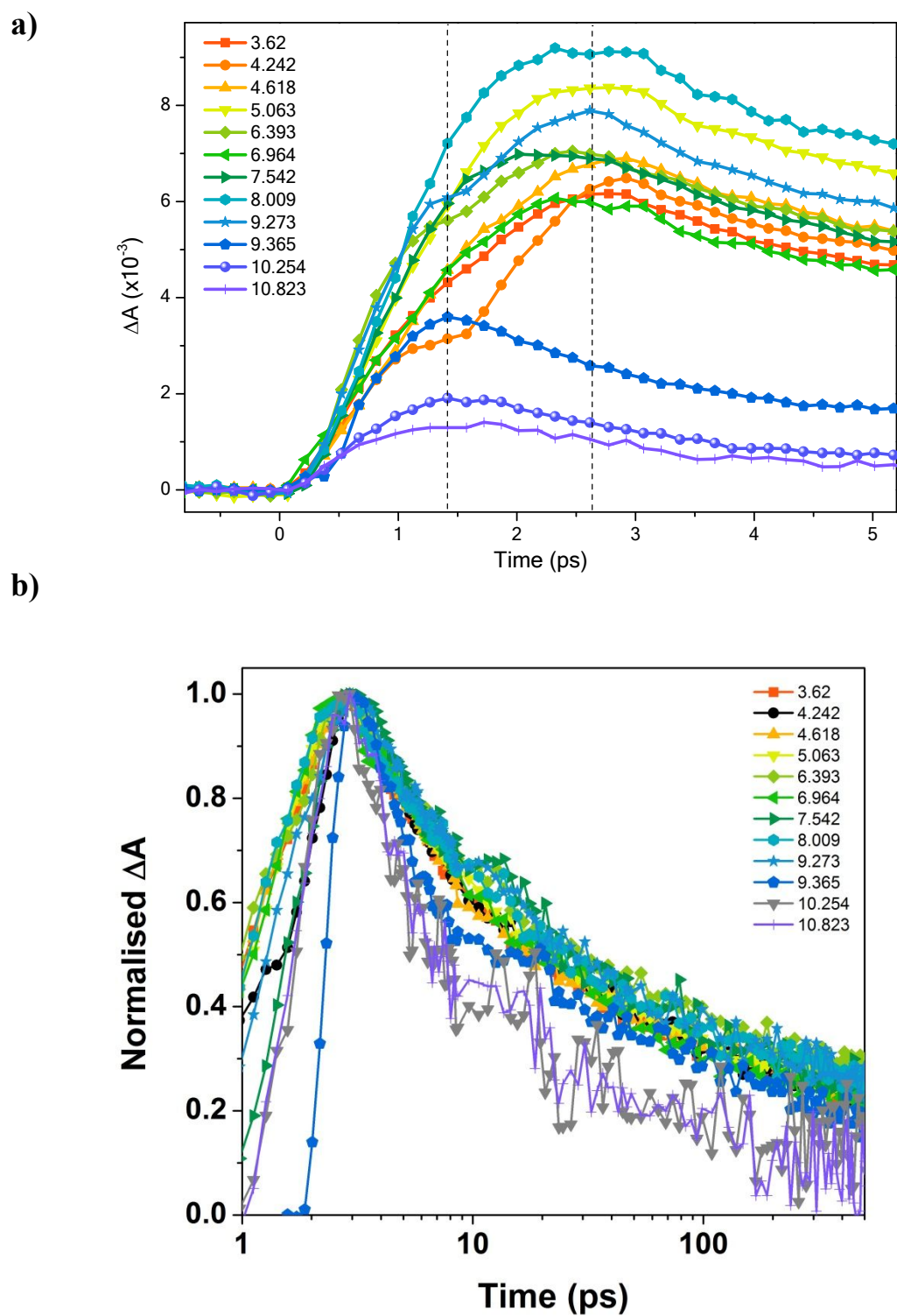


Figure S7 Differential absorption transients showing (a) stepwise growth for $\text{pH} < 9.3$ and monotonic growth for $\text{pH} > 9.3$, and (b) more rapid decay for $\text{pH} > 9.3$.

S5. Decay component amplitudes.

Globally fitting tri-exponential decays to figure 4b yields time constants $\tau_1 = 2.0 \pm 0.1$ ps, $\tau_2 = 15 \pm 1$ ps and $\tau_3 = 140 \pm 10$ ps. The variation in amplitude of each component with pH is plotted in Figure S, in which it can be seen that the amplitude associated with τ_1 increases significantly for $\text{pH} > 9.3$, with the amplitudes associated with the other time constants drops at the same point.

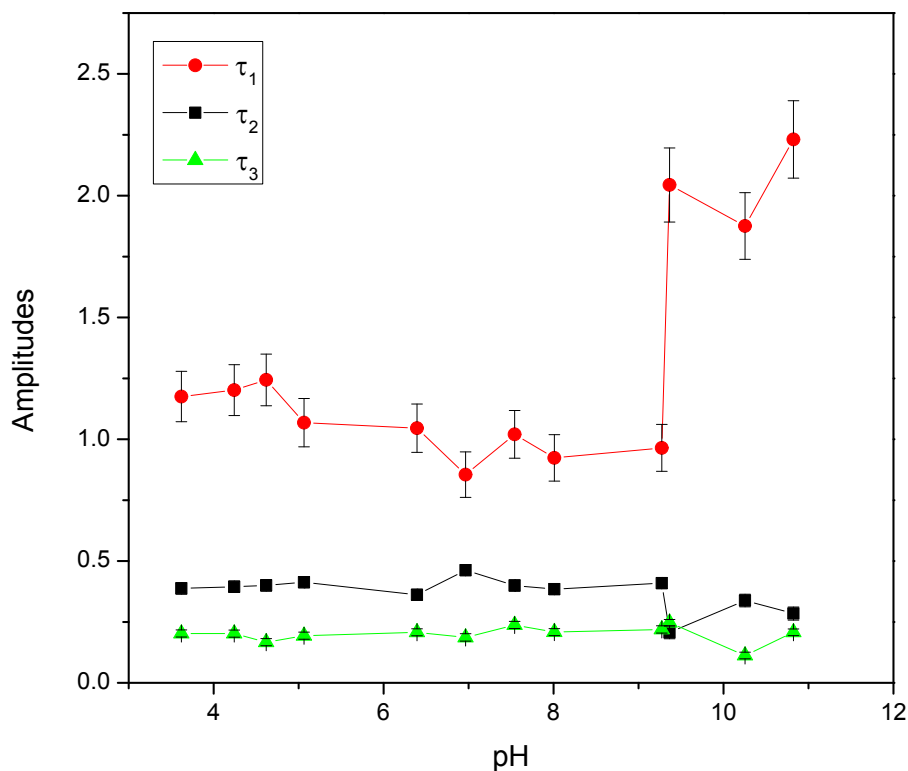


Figure S8. Amplitudes associated with the three exponential time constants (τ_1 , τ_2 , τ_3) fitted to the transient absorption decay curves of GO dispersions at a range of pH levels.