Supplementary Figures

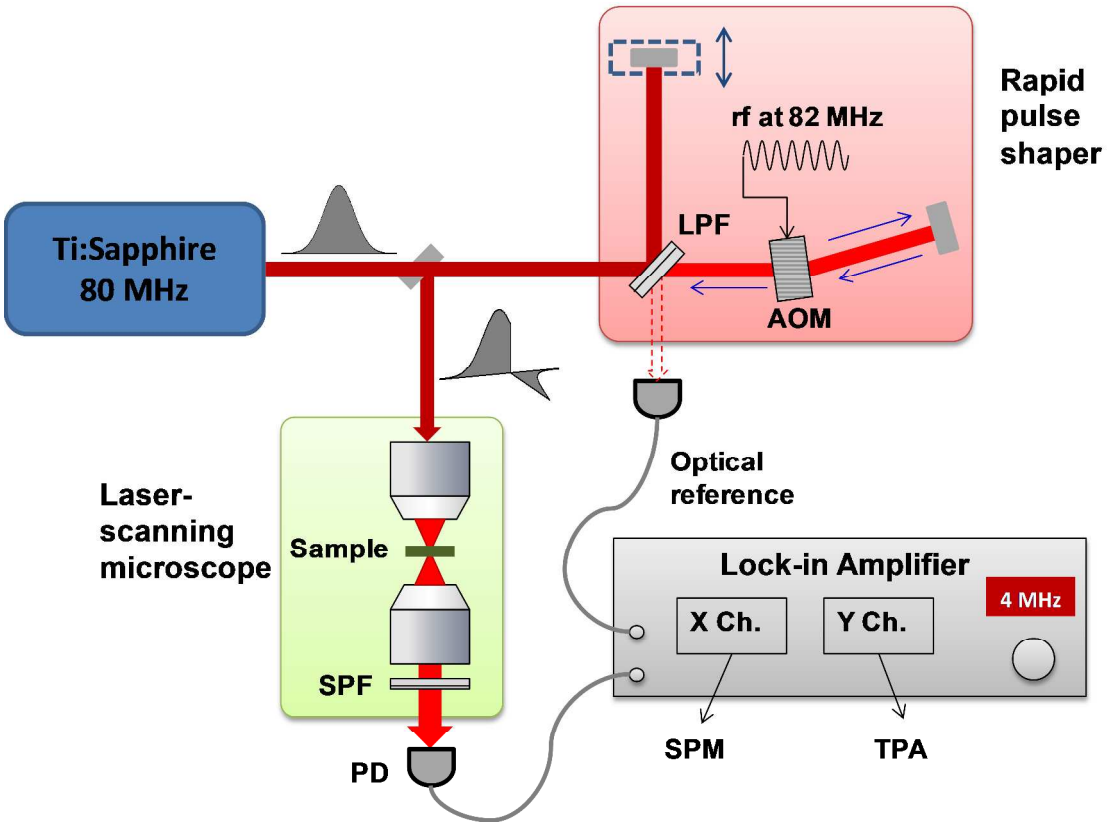

Figure S1 | Experimental setup for spectral reshaping nonlinear optical microscopy. A train of femtosecond laser pulses are converted into the desired pulse shapes by a rapid pulse shaper and sent into a home-built laser scanning microscope, where graphene samples are examined in a transmission mode. A lock-in amplifier is used to record the nonlinear optical signals with a reference produced by the remaining output from the pulse shaper. LPF, long-pass filter; AOM, acousto-optic modulator; SPF, short-pass filter; PD, amplified photodetector.

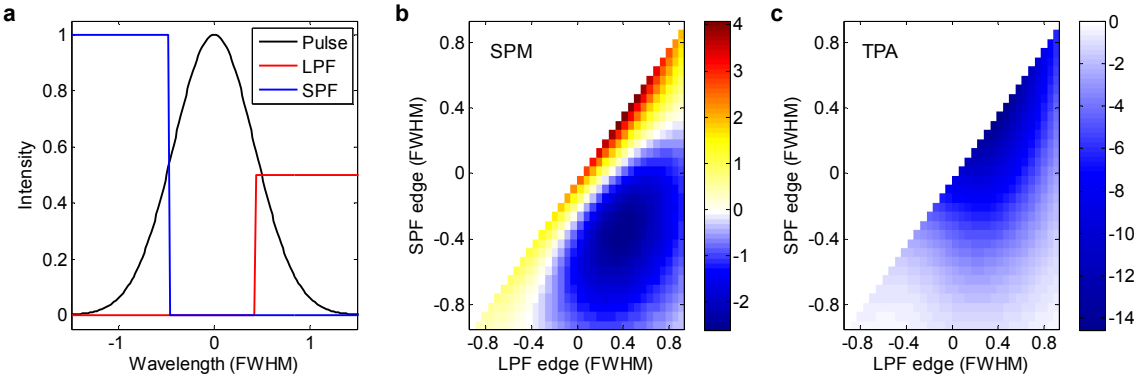

Figure S2 | Numerical calculation of SPM and TPA signals. a, short-pass filter (SPF) and long-pass filter (LPF) position. **b, c** SPM and TPA signals as a function of SPF and LPF edge positions.

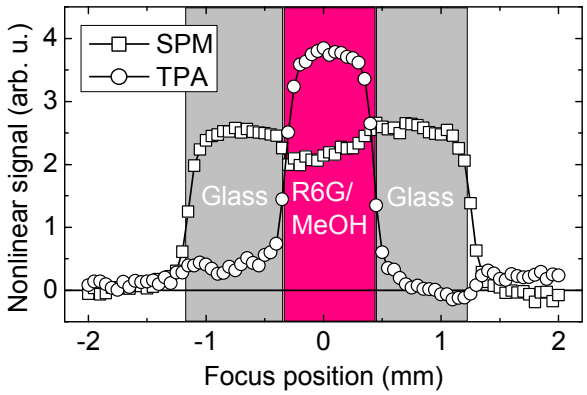

Figure S3 | SPM and TPA measurement of a quartz cuvette filled with 20 mM Rhodamine 6G in methanol. Input power is 1.5 mW, and lock-in time constant is 100 ms. From this result, β is estimated to be 1.7.

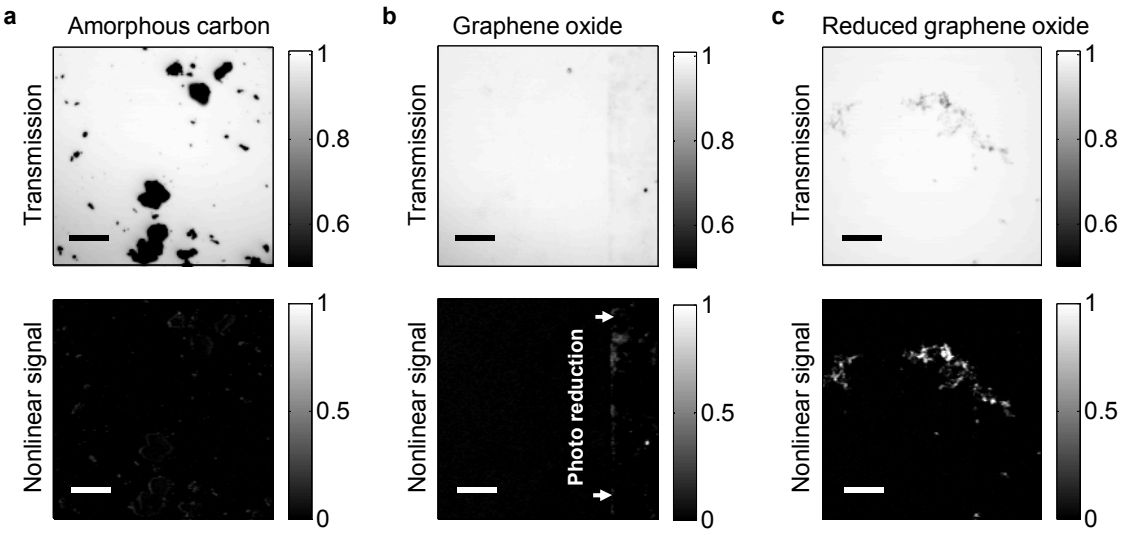

Figure S4 | Transmission and nonlinear optical images of amorphous carbon (a), graphene oxide (b) and reduced graphene oxide (c). No appreciable nonlinear optical signal is generated in amorphous carbon and graphene oxide. Long-time exposure to stronger laser power levels causes photo-induced reduction in graphene oxide and produces nonlinear optical signals.

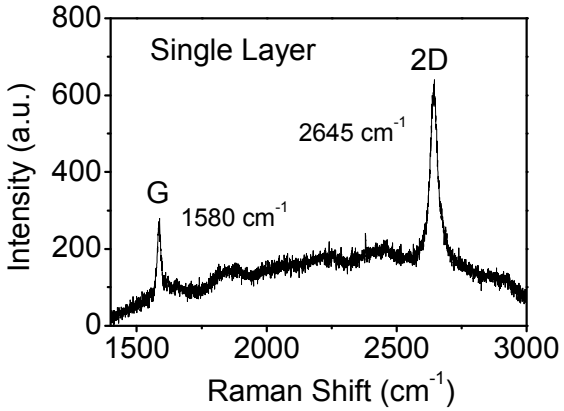

Figure S5 | Raman Spectrum of the single layer graphene in Fig.2b region 1. The 2D peak is higher than (almost twice as large as) the G peak, showing we have a single layer in this region.

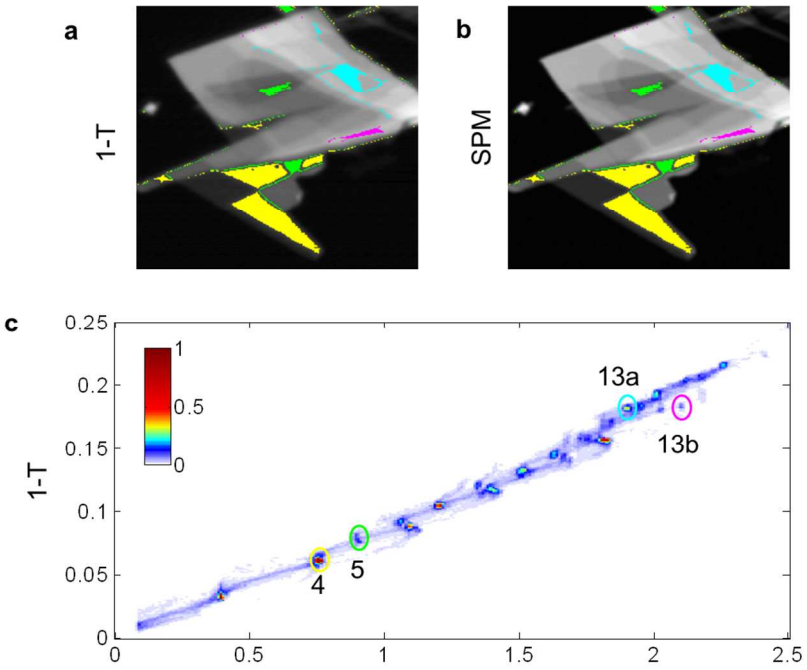

Figure S6 | Layer counting in few layer graphene flakes. a, Transmission contrast. **b**, SPM contrast in the same region. **c**, 2D histogram of transmission and SPM contrast, where colored circles label the region with certain number of layers obtained from transmission. These regions are highlighted in **a**, **b** in the corresponding colors. 13A and 13B denote regions with same number of layers at different locations. They produce different SPM signal because they experienced slightly different laser power.

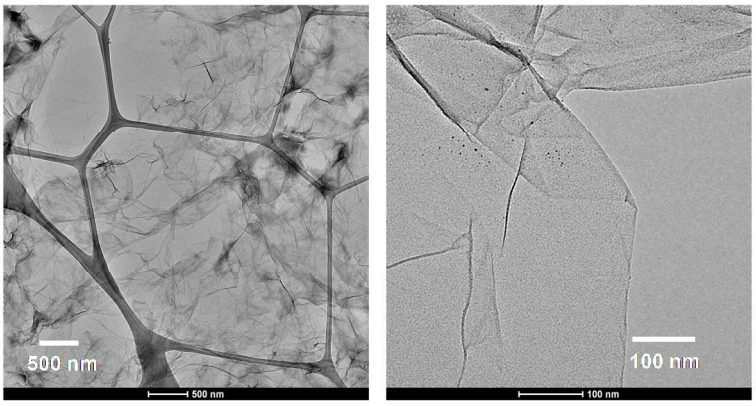

Figure S7 | TEM images of reduced graphene oxide