Supplementary materials

Cyclic Voltammetry and Impedance Measurements of Graphene Oxide Thin Films Dip-Coated on n-Type and p-Type Silicon

SEM measurements

SEM images of GO films on n-and p-type Si substrates are reported in Figures S1 and S2, respectively.
SEM analysis was accomplished with an FEI Quanta FEG 400 FESEM microscope (Eindhoven, The Netherlands).
As shown in Figure S1, large GO sheets are identified on Si substrates.

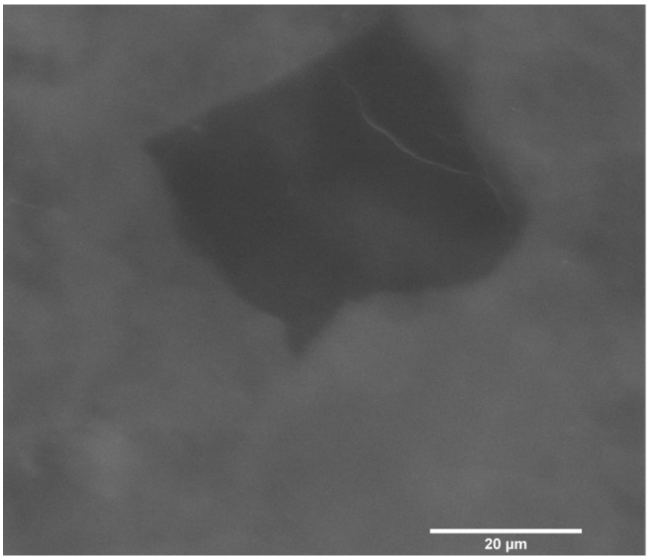

Figure S1. Scanning electron microscopy image of GO films on Si substrates (n-type).

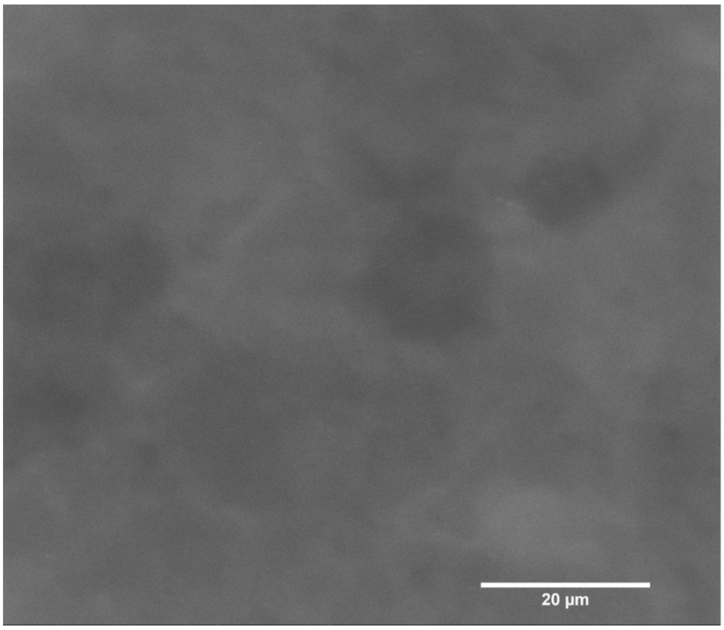

Figure S2. Scanning electron microscopy image of GO films on Si substrates (p-type).

Dynamic light scattering measurements

Dynamic light scattering gives a mean hydrodynamic diameter that results from the mean of the three translational diffusion coefficients.

In short, the linearly polarized light beam of a 35 mW HeNe laser (Spectra Physics mod. 127) was focused using a 300 mm focal length lens on the sample cell, which consisted of a quartz

cylindrical cuvette (2 cm inner diameter).

The autocorrelation functions of the scattered light were constructed using a Brookhaven Instruments 2030AT digital correlator, processing the photocurrent pulse produced by a 9863A Thorn-EMI photomultiplier with a built-in amplifier–discriminator. The light was collected at the scattering angle of 90° and a light collimator with a 50 mm pinhole allowed us to operate always with at least one coherence in the regime of pure homodyne detection. The autocorrelation function was Laplace inverted by the CONTIN routine.

We measured a Gaussian size distribution ranging from 800 nm to 7 μm, as can be seen in Figure S3.

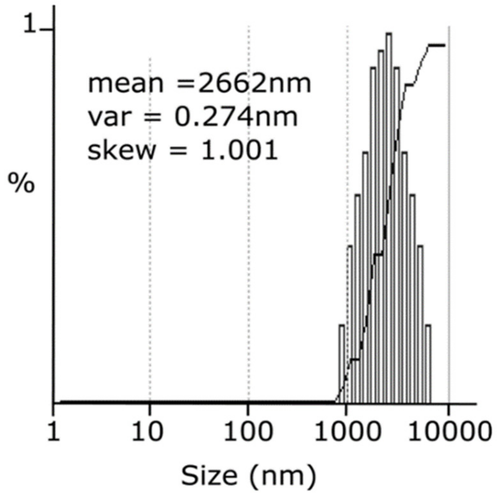

Figure S3. Particle size distribution of very diluted GO flakes in water.

Variable Angle Spectroscopic Ellipsometry Measurements

The thicknesses of the samples were estimated using Variable Angle Spectroscopic Ellipsometry measurements (VASE). The spectra of the ellipsometric angles ψ and Δ were acquired using a V-Vase (J.A. Woollam, Lincoln, NE, USA) ellipsometer in the (250-1000) nm wavelength range at different incident angles at room temperature. These angles were chosen because VASE measurements are usually made around Brewster's angle air substrate, which, for silicon, is about 65°, to have better contrast. The optical model and the best fitting values were calculated with WVASE32 (J.A. Woollam, Lincoln, NE, USA) application by means of the nonlinear Levenberg–Marquardt algorithm, which determines the minimum value of the mean square error (MSE).

We estimated the optical model of the substrates using the models implemented in the WVASE32 software.

In Figures S4 and S5, the experimental and generated values of ψ (a) and Δ (b) for GO on Si substrates (n-and p-type) are reported.

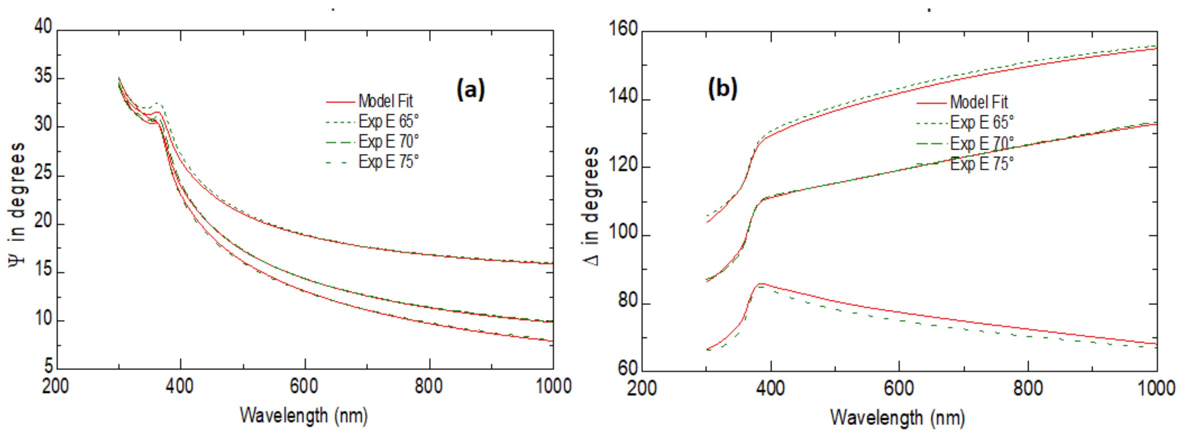

Figure S4. Variable angle spectroscopic ellipsometry measurements of graphene oxide films on Si substrates (n-type). Experimental and model generated ψ (**a**) and Δ (**b**) data fit at different angles of incidence.

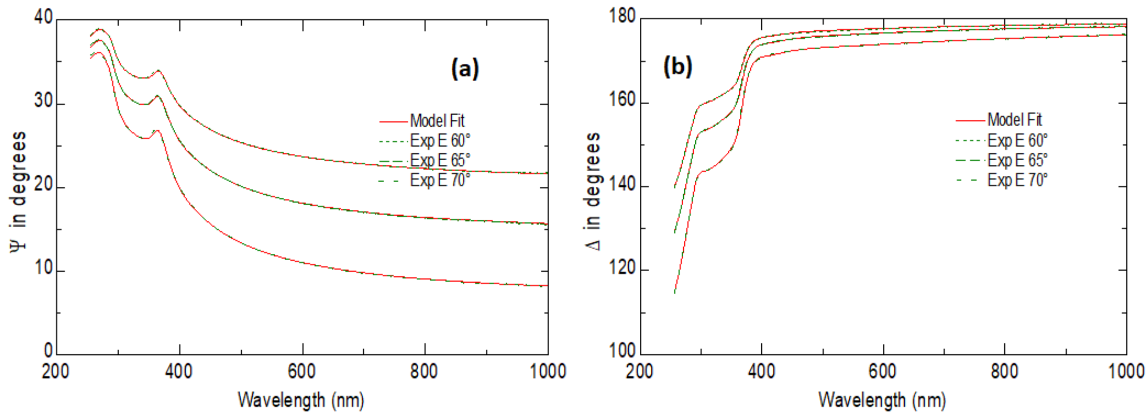

Figure S5. Variable angle spectroscopic ellipsometry measurements of graphene oxide films on Si substrates (p-type). Experimental and model generated ψ (**a**) and Δ (**b**) data fit at different angles of incidence.

GO films on the n-type and p-type Si substrates were modeled as the sum of three Lorentz oscillators to keep consistency with the Kramers–Kronig relations. The complex dielectric function is characterized by the relation:

$$\tilde{\varepsilon}(h\nu) = \varepsilon_1 + i\varepsilon_2 = \varepsilon_\infty + \sum_{k=1}^{N} \frac{A_k}{{E_k}^2 - E^2 - i\Gamma_k E}$$

where E is the energy of the incident photons, ε_∞ is the real part of the dielectric function when $E \rightarrow \infty$, A_k is the strength expressed in eV^2, Γ_k is the broadening in the eV end, E_k is the central energy of the k-th oscillator. A_k also indicates the contribution of each oscillator k to the whole system.

This oscillator model provides thickness data fittings around 10 nm with a low MSE for GO on n- and p-type Si.

Additional electrical measurements on n-type Si and GO/n-type Si samples

In Figures S6 and S7, impedance and cyclic voltammetry measurements of n-type Si and GO/n-type Si samples under dark and light illumination conditions are reported. The thickness of this additional GO/n-type Si sample was around 8 nm. The thickness was estimated using VASE.

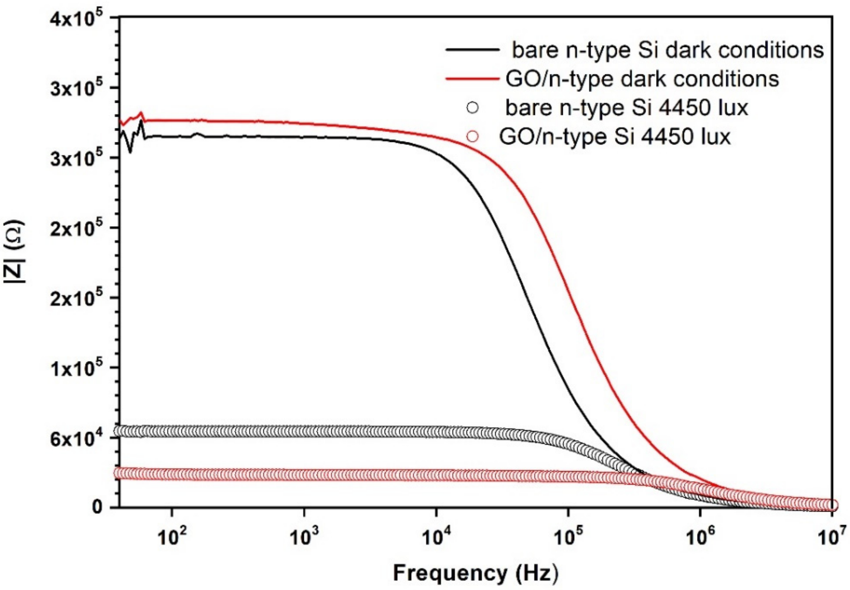

Figure S6. Impedance measurements of n-type Si and graphene oxide /n-type Si samples under dark and light illumination conditions.

As can be seen in Figure S6, the bare n-type Si shows an identical impedance to that of GO/n-type Si in dark conditions. In the presence of light radiation, there is an increase in conductivity of about four times for GO/n-type Si in comparison to bare n-type Si. As shown in figure 7 of the manuscript, the conductivity is quintupled for GO/n-type Si samples (thickness around 10 nm) in comparison to bare n-type Si under light conditions.

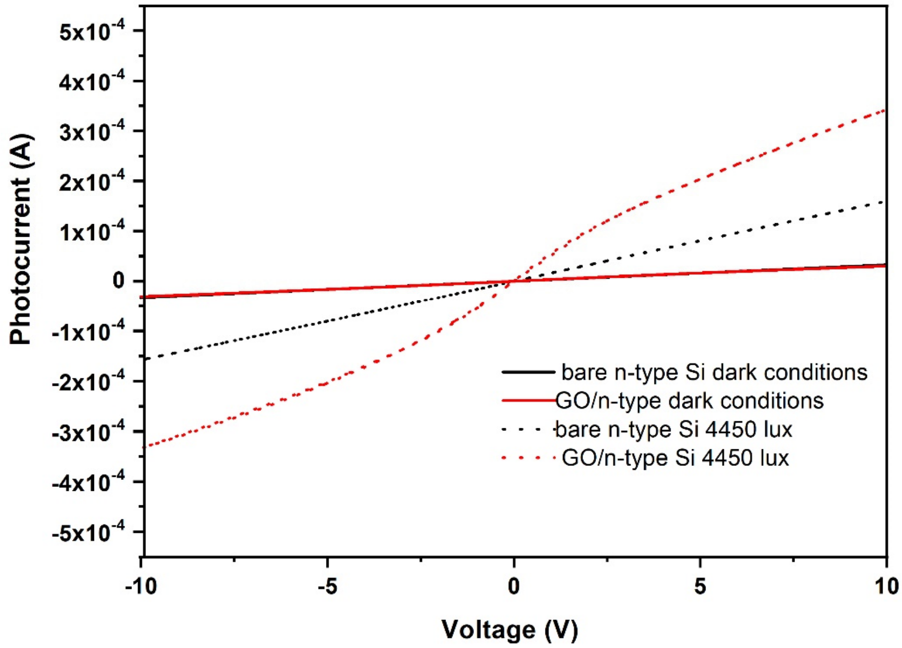

Figure S7. Cyclic voltammetry measurements on n-type Si and graphene oxide /n-type Si samples under dark and light illumination conditions.

As can be seen in Figure S7, the n-type Si sample shows almost the same current behavior in dark conditions. The photocurrent of GO/n-type Si samples doubled in comparison to bare n-type Si samples under light illumination. As shown in Figure 5 of the manuscript, the I–V characteristics show that the photocurrent of the GO/n-type Si sample (thickness around 10 nm) more than tripled in comparison to bare n-type Si samples under light illumination. This could be due to a better homogeneity of the sample.