Supporting Information

CF_4/H_2 Plasma Cleaning of Graphene Regenerates Electronic Properties of the Pristine Material

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S1. C1s spectrum obtained from CVD graphene grown on Cu foil

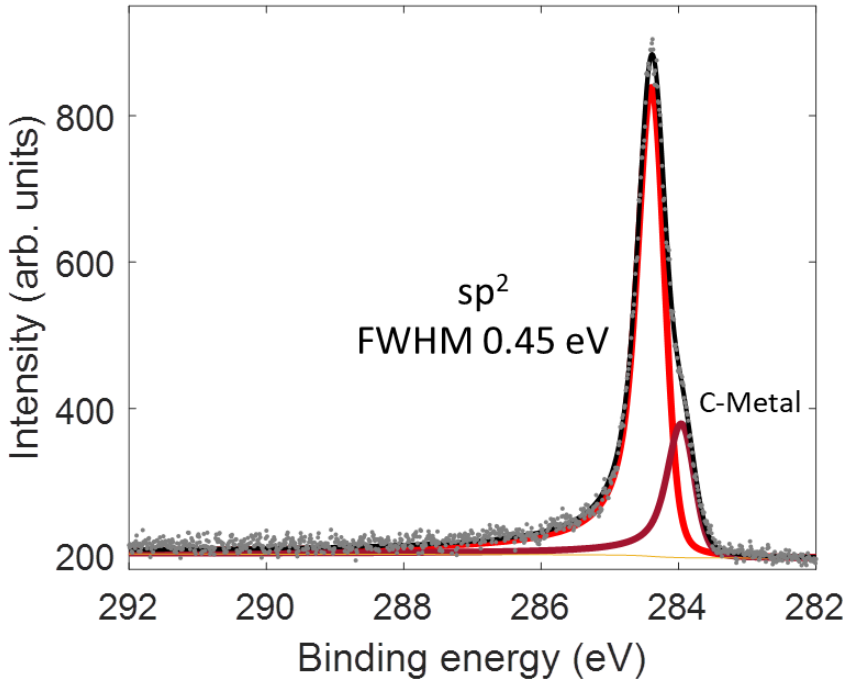

Figure S1: C1s core level spectrum of CVD graphene grown on Cu showing FWHM of 0.45 eV

S2. AM-KPFM imaging of pre-treated CVD graphene transferred onto 5nm-SiO_2/Si

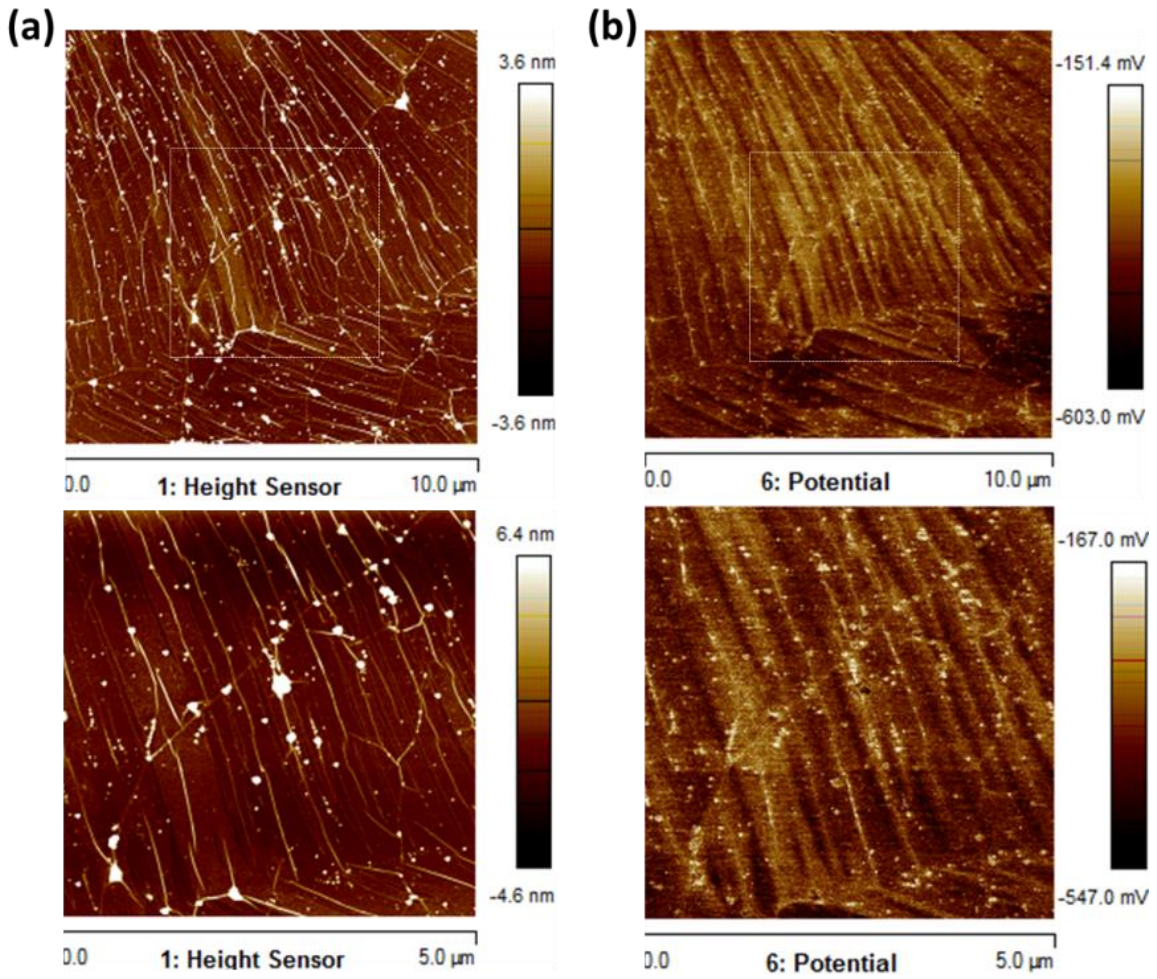

Figure S2: (a) Surface topography and (b) potential images of pre-treated CVD graphene transferred onto 5nm-SiO_2/Si, provided by AM-KPFM.

Surface topography and potential images of pre-treated CVD graphene transferred onto 5nm-SiO_2/Si, provided by AM-KPFM (Kelvin Probe Force Microscopy) and collected at different scales (10 x 10 µm²) and (5 x 5 µm²), are illustrated in Figure S2. Potential images display a variation in surface potential with respect to the surface composition. They reveal the presence of bright small particles associated to the presence of Si-based nanoparticles of approximatively 50-200 nm size. A few nanometers thick blocks are revealed in both topography and potential modes and associated to thick $PMMA^A$ layers. Large particles of hundreds nanometers size, evidenced only in topography mode, were attributed to $PMMA^B$ (bulk PMMA) left on the surface. These particles exhibit similar surface potential than $PMMA^A$ (dense PMMA layer). Here, we have not been able to report any distinguishable characteristics between $PMMA^G$ and thin $PMMA^A$ layers, associated with low potential area.

S3. Electrical transport properties of optimized plasma-treated CVD graphene

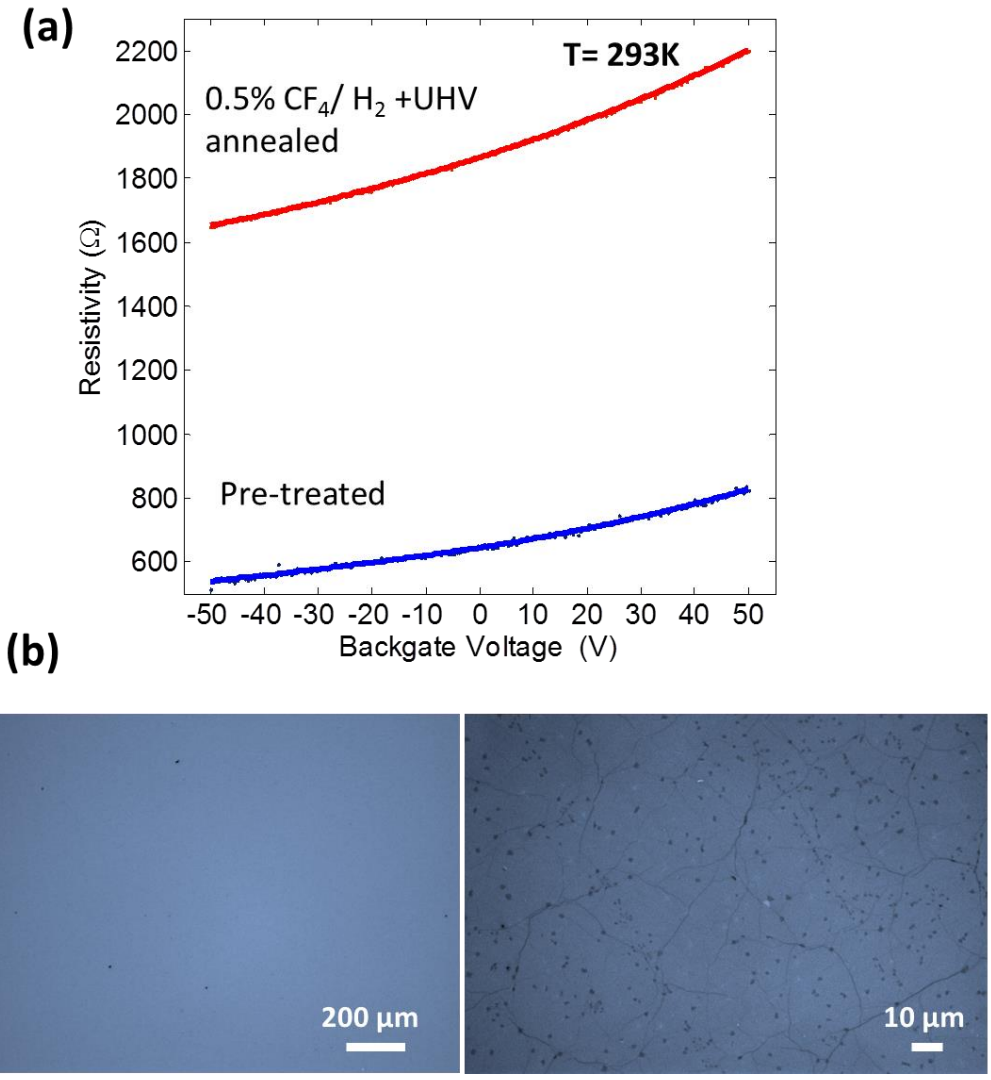

Figure S3: (a) Electrical transport measurements recorded from untreated and post-plasma treated and annealed CVD graphene transferred onto a SiO_2/Si substrate showing an increase in resistance after the surface treatment. (b) Large-scale optical microscopy images of sample obtained after plasma cleaning process.

Figure S3 (a) shows the electrical transport measurement in the CVD graphene on 300 nm-SiO_2/Si, measured between two Au electrodes (100 mm dots deposited 5 mm apart), and obtained before and after an optimized plasma treatment and annealing. The Au electrode deposition is done after the cleaning process. The high resistance measured on uncleaned graphene is explained by strong doping due to surface impurities (PMMA, Si- nanoparticles). Cleaning and annealing increase the resistance by, approximately, a factor of 3. This result suggests that the sample is p doped, even after the annealing, probably due to the discontinuity of the CVD graphene sheet at the grain boundaries.

S4. Work function measurement on pure H_2 plasma-treated CVD graphene

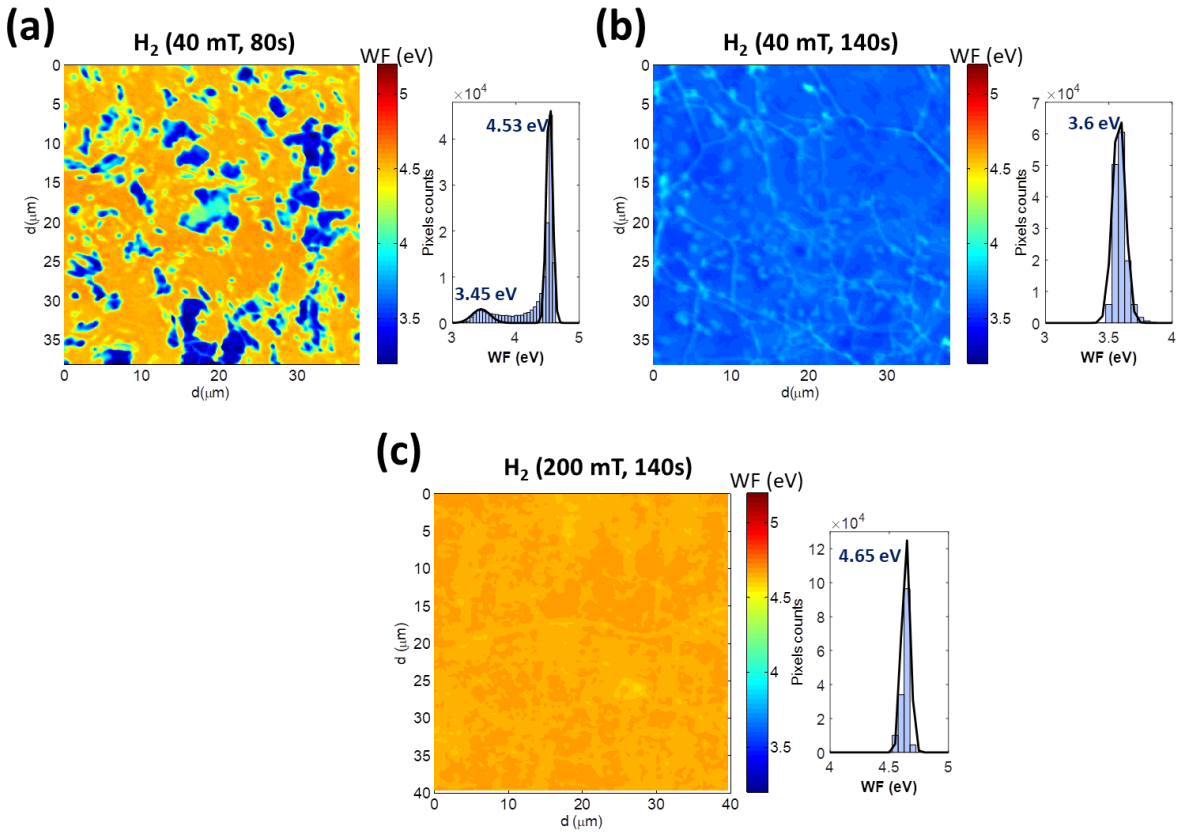

Figure S4: Work function maps (42 x 42mm) derived from the threshold PEEM analysis with their corresponding histogram, after plasma exposure for (a) 80 s and (b) 140 s at a working pressure of 40 mTorr (condition 1) and (c) exposure for 140 s at a working pressure of 200 mTorr (condition 2). Thermal annealing of the samples in vacuum for 1 hour at 400 ^{o}C was performed to desorb hydrogen and water from the surface.

Figure S4 (a) and (b) shows the work function maps (42 x 42 μm^2) and corresponding histogram of the resulting surface from reactive-ion H_2 plasma etching after 80 s and 140 s exposure, respectively. For the reduced-time plasma surface treatment (80 s), a work function value of 3.45 ± 0.2 eV (blue color area) and 4.53 ±0.2 eV (orange color area) are found while only a value of 3.6 ± 0.2 eV is found at the end of the process. The 4.53 work function is associated to undoped graphene, while the work function value of ≈ 3.5 eV is assigned to the damaged graphene. Although the energies of the ion bombardment are considered small enough, the etching is not selective which results in the deterioration of the graphene structure. Figure S4 (c) shows the work function map (42 x 42 μm2) and corresponding histogram of the resulting surface from radical H_2 plasma etching after 140 s time exposure. The surface is very homogeneous and displays a work function value of 4.65 ± 0.2 eV (orange color), assigned to p-doped graphene by remaining PMMA[G]. According to these results, the preparation of a high-quality monolayer graphene/SiO_2/Si is in contradiction with the use of H_2 plasma when a certain amount of ions energy is supplied to the etching of the residual contaminants.

S5. Pressure tuning for plasma cleaning process optimization

Table S1: Impact of pressure on the H_2 plasma cleaning processes

Ions energy	Pressure (mTorr)	Graphene/Cu	Graphene/SiO_2/Si
> 10 eV	10	Damage in the graphene.	Damage in the graphene.
~ 10 eV	20	Minimal pressure that not damage graphene.	Damage in the graphene.
< 10 eV	40	**Ideal pressure sets for cleaning**	Damage in the graphene.
	100		Minimal pressure that not damages graphene.
	200		**Ideal pressure sets for cleaning**
	300 and higher		Increase the amount of H_2 formed at the interface.

In our ICP reactor, a pressure range between 10- 500 mTorr enables a stable plasma operation. Beyond the pressure range, we face operational issues including the generator ignition, plasma stability, and lack in species concentration control. The optimal operating pressure is selected on the basis of the ion energy that didn't damage sp^2 graphene structure. Figure S5 (a) displays the ions energy distribution for H_2 plasma working at 20 mTorr and 800 W. The impact of pressure on graphene have been investigated and revealed a gap in optimal pressure that leads to undamaged graphene supported on SiO_2/Si and copper, as discussed in the paper and summarized above in Table S1. Figure S5 (b) shows optical microscopy image recorded after plasma treatment at 360 mTorr and 800 W. Basically, higher working pressure leads to an important accumulation of H_2 at the interface.

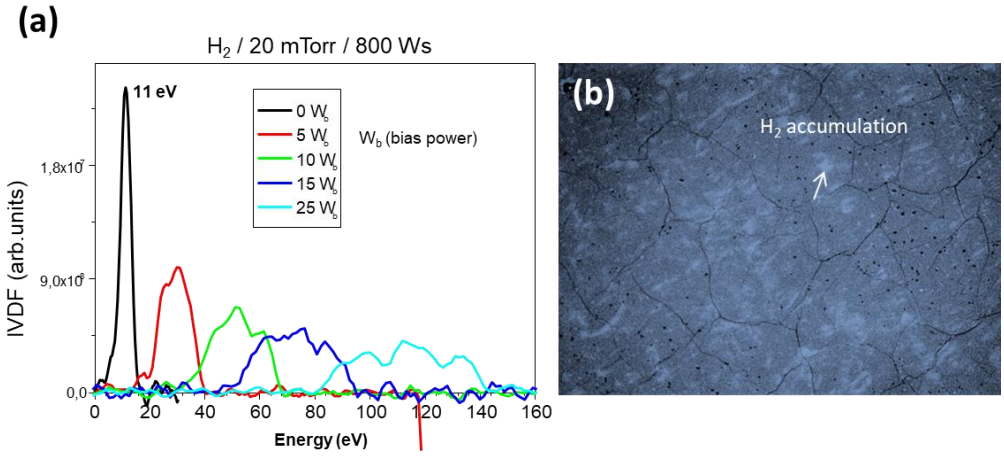

Figure S5: (a) Ions energy distribution for H_2 plasma at 20 mTorr and 800 W using ICP reactor, obtained at different bias power. (b) Optical microscopy image obtained after H_2 plasma working at 360 mTorr and 800 W.