

Supplementary Information to:

**Elucidation on the wettability of graphene throughout a multi-length-scale  
investigation approach**

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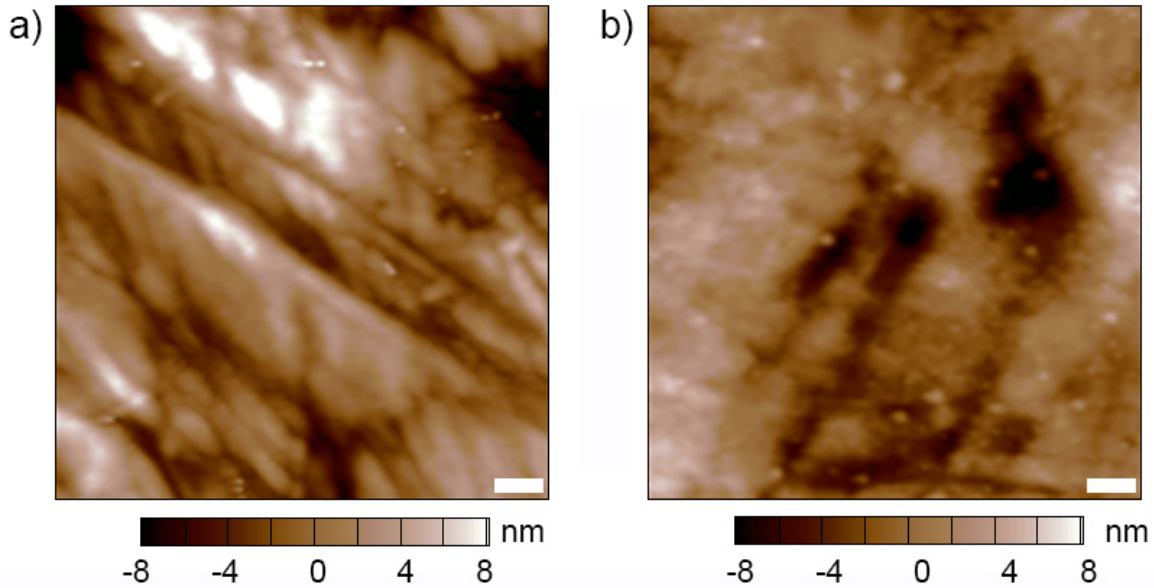
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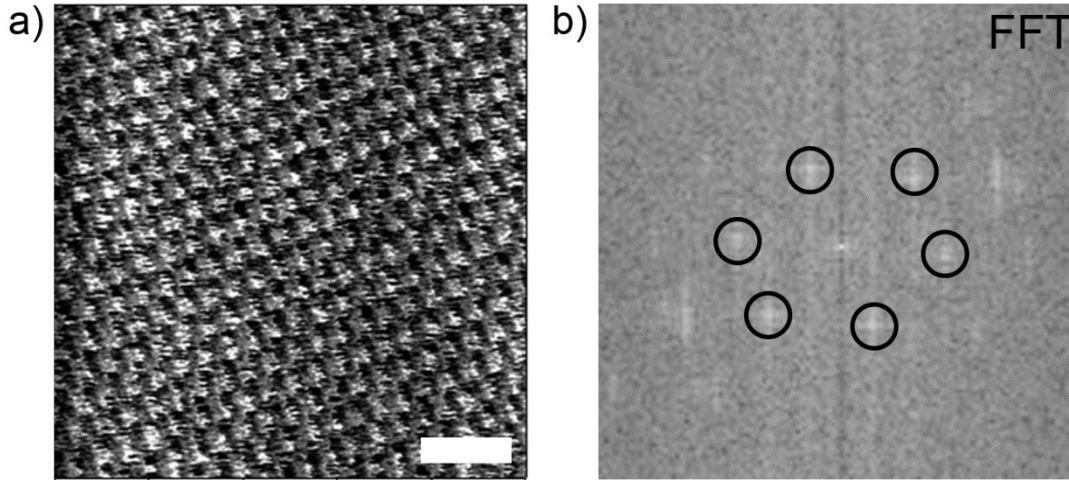
### AFM study on the bare metal



**Figure S1: bare metal surface characterization by means of AFM. AM AFM topographical images of Pt (a) and Au (b). The scale bars are 100 nm.**

Figure S1 represents images of the bare metals (Au and Pt) acquired in AM AFM. During the image acquisition, the tip is not in mechanical contact with the sample and the topography is recovered by means of long range interaction. Thus, the tip is oscillating in attractive regime.<sup>6</sup> It is possible to observe that the topographical image of the bare Pt (Figure S1a) is comparable to the one obtained for GML/Pt surface presented in the main manuscript (Figure 1a). Moreover, the RMS of the two surfaces (Pt and GML/Pt) is comparable (3.3 nm and 2.4 nm). This is due to the fact that the graphene growth is driven by the crystallographic orientation of the Pt. The roughness of the Au sample is qualitatively different compared to the roughness of the GML/Au surface. This was expected, since the GML was growth on Cu sample and then mechanically transferred on the Au surface. However, the RMS of the Au surface is 2.8 nm which is a value close to the one obtained for GML/Au.

### Crystallographic structure of GML



**Figure S2: high resolution image of GML. (a) lateral force AFM image on GML/Au and its FFT (b). Scale bar is 1 nm.**

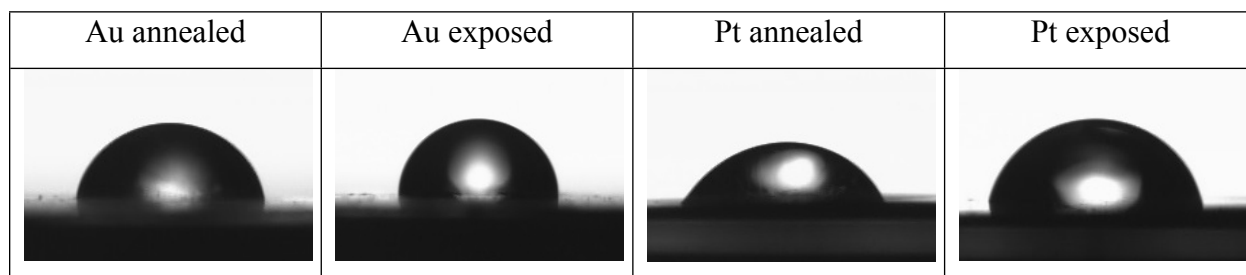
Figure S2a represent 5x5 nm image acquired by means of lateral force AFM (see main manuscript). The presence of the hexagonal patten is corroborated by a FFT (Figure S2b) image which exhibits hexagonal spot patterns, highlighted with black circles. This pattern is typical of graphitic surfaces as it was demonstrated with other microscopy tools.<sup>8,9</sup>

### Analysis of metal surfaces after annealing

Table S1 shows the influence of the annealing (12h at 230 °C in vacuum) on the SCA of metal surfaces. It is possible to observe that the annealing lower the SCA of the bare metal (Figure S3), and this is probably due to adsorption removal. The following ambient exposure, lead the contact angels to a value identical to the one before the annealing.

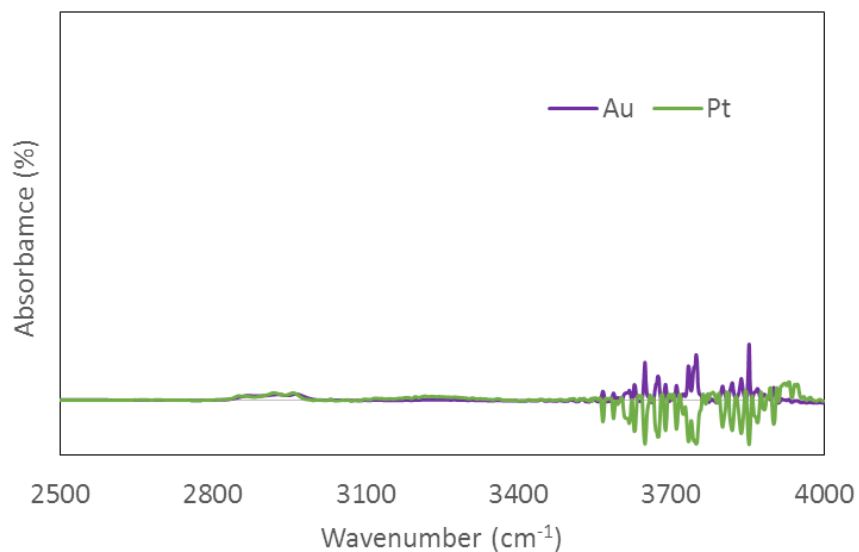
**Table.S1 SCA measurements for annealed and exposed samples.**

Sample	Before annealing (°)	After annealing (°)	Exposed (°)
Au	80.4±2	74.7±1.5	80.5±1.8
Pt	65.3±3	57.9±3.9	66.2±1.8



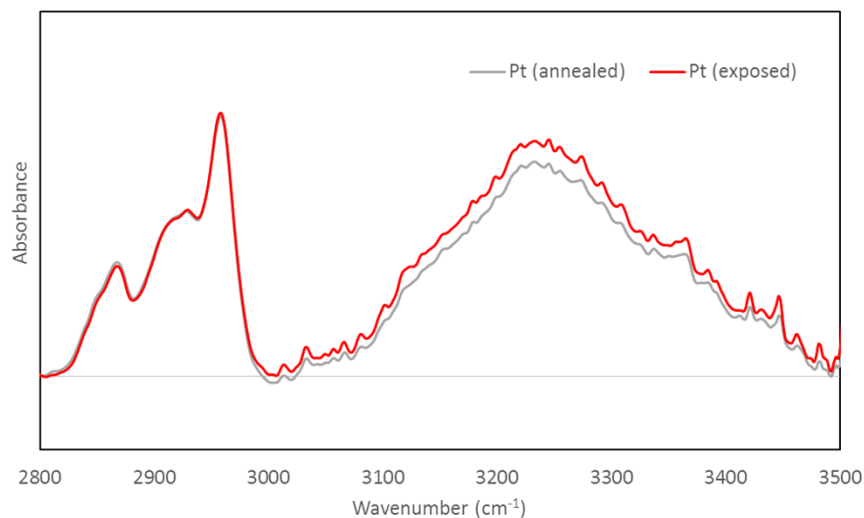
**Figure S3: SCA of bare metal surfaces.**

In order to study the adsorption on the bare metals we carried out Fourier transform Infrared spectroscopy (FTIR). The FTIR measurements (Figure S4) for both annealed samples are conducted with absorbance unit and right after the annealing. From the spectra, we can see that the surfaces of both samples are clean without obvious peaks of C-H and O-H bonds. Note that there are vapor signals detected due to the measurement were done in the ambient conditions.



**Figure S4: FTIR spectra on metal surfaces.**

We then exposed the Pt surface exposed to the ambient air for 12 hours and we record the evolution of the FTIR spectra (Figure S5). By focusing, on the wavenumber window between 2800-3500  $\text{cm}^{-1}$ , we could see that the only varying signals belong to O-H stretching bonds (14% of increase), indicating possible water adsorption.



**Figure S5: evolution of FTIR spectra on Pt exposed to 12 hours to environmental conditions.**

### **Dynamic contact angle analysis**

Table S2 summarizes the dynamic contact angle (DCA) analysis for the Pt and GML/Pt sample. Advancing and receding contact angles (ACA and RCA) were measured by recording the advancing and receding of the contact line, while increasing or decreasing the droplet volume by using the well-known sessile drop method.

Table S2: Dynamic contact angle analysis for Pt and GML/Pt.

System	ACA (°)	RCA (°)
Pt	70.2±4.9	50.5±3.2
GML/Pt	100.4±2.6	66.7±5.1

Pt displays a different behaviour compared to the graphene covered one (GML/Pt). In, particular, the contact angle hysteresis (i.e. the difference between the ACA and the RCA) of Pt is nearly 15° less compared to GML/Pt one. Moreover DCA obtained for GML/Pt are similar to the one obtained for graphite.<sup>1</sup> This two observations support the minor role played by the substrate, indicating the non-wetting transparency for GML.

## References

1. Katan, A. J.; Van Es, M. H.; Oosterkamp, T. H. Quantitative force versus distance measurements in amplitude modulation AFM: a novel force inversion technique. *Nanotechnology* 2009, 20 (16), 165703.
2. Giessibl, F. J. Forces and frequency shifts in atomic-resolution dynamic-force microscopy. *Physical Review B* 1997, 56 (24), 16010-16015.
3. Sader, J. E.; Jarvis, S. P. Accurate formulas for interaction force and energy in frequency modulation force spectroscopy. *Appl. Phys. Lett.* 2004, 84 (10), 1801-1803.
4. Santos, S.; Guang, L.; Souier, T.; Gadelrab, K.; Chiesa, M.; Thomson, N. H. A method to provide rapid in situ determination of tip radius in dynamic atomic force microscopy. *Review of Scientific Instruments* 2012, 83 (4), 043707.
5. Santos, S.; Amadei, C. A.; Verdaguer, A.; Chiesa, M. Size Dependent Transitions in Nanoscale Dissipation. *The Journal of Physical Chemistry C* 2013, 117 (20), 10615-10622.
6. García, R.; San Paulo, A. Attractive and repulsive tip-sample interaction regimes in tapping-mode atomic force microscopy. *Physical Review B* 1999, 60 (7), 4961-4967.
7. Cleveland, J. P.; Anczykowski, B.; Schmid, A. E.; Elings, V. B. Energy dissipation in tapping-mode atomic force microscopy. *Appl. Phys. Lett.* 1998, 72 (20), 2613-2615.
8. Gass, M. H.; Bangert, U.; Bleloch, A. L.; Wang, P.; Nair, R. R.; Geim, A. K. Free-standing graphene at atomic resolution. *Nat Nano* 2008, 3 (11), 676-681.
9. Chatterjee, A.; Zhang, L.; Leung, K. T. Self-Directed Growth of Aligned Adenine Molecular Chains on Si(111)7×7: Direct Imaging of Hydrogen-Bond Mediated Dimers and Clusters at Room Temperature by Scanning Tunneling Microscopy. *Langmuir* 2013.