SUPPORTING INFORMATION

Probing the Interactions in Graphene Oxide/MoS_2 and Reduced Graphene Oxide/MoS_2 Nanoarchitectures Using Multimodal Scanning Probe Microscopy

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1. Experimental

1.1 Synthesis of graphite oxide

For the synthesis of graphite oxide, a modification of the Hummers method was used, which consists of adding 46 mL of sulfuric acid to a 500 mL round flask containing 2.0 g of graphite and 1.0 g of $NaNO_3$ under magnetic stirring in an ice bath for 15 minutes. Then, 6 g of $KMnO_4$ was slowly added to the flask at 0 °C under stirring. After 75 minutes of stirring, 92 mL of distilled water was added to the mixture under stirring for 10 minutes, then 280 mL of distilled water at 100 °C were added. 10 mL of 30% H_2O_2 aqueous solution was sequentially added under stirring for 30 minutes. The mixture was washed with a 1 $mol.L^{-1}$ HCl solution and ultrapure water until neutral pH was obtained. The system was filtered, and the resulting solid was dried at 60 °C for 24 hours.

1.2 Synthesis and preparation of graphene oxide (GO) and reduced graphene oxide (rGO) films

GO was acquired by exfoliating a mixture of 1 mg of graphite oxide/ 1 mL of water for 2 hours in ultrasound bath. The dispersion was left to rest for decantation for 15 hours and then subjected to centrifugation at 3000 rpm for 90 minutes, separating the GO-containing supernatant water dispersion. To obtain the rGO, 4 mg of sodium borohydride was added to each mL of the supernatant, and this mixture was refluxed for 3 hours. The system was filtered, and the resulting solid (rGO) was washed with ultrapure water and dried at 50 °C for 24 hours. The dispersion (30 $\mu g\ mL^{-1}$) in 20 mL of toluene was obtained in an ultrasound bath for 90 min.

For the formation of thin films by the liquid-liquid interfacial route (LLIR), 20 mL of aqueous GO dispersion (0.02 $mg\ mL^{-1}$) and 20 mL of toluene were added to a 50 mL round-bottom flask under stirring at 2500 rpm for 12 h to form the GO film. For the rGO film, 20 mL of the rGO dispersion (30 $\mu g\ mL^{-1}$) in toluene and 20 mL of ultrapure water were add in a 50 mL round-bottom flask under stirring at 2500 rpm for 3 h. After this time the stirring was interrupted and a thin film was spontaneously organized at the liquid-liquid interface. The solvents were replaced by fresh ones 3 times, and the film was subsequently transferred to solid substrates by putting the substrate bellow the film and lifting it across the interface.

1.3 Synthesis and preparation of molybdenum disulfide (MoS_2)

10 mL of ammonium molybdate (1.0 mmol L^{-1}) and 0.5 mL of H_2SO_4 (1.0 mol L^{-1}) were added in a 50 mL round bottom flask. This system was stirred at 1500 rpm for 2 h. 10 mL of $(NH_4)_2S$ (0.1 mol L^{-1}) was added and stirring was changed to 2500 rpm for 3 hours. The system was centrifuged for 20 min at 4000 rpm. The supernatant was exchanged for ultrapure water to wash the solid, and this procedure was repeated three times. The final solid was dried in an oven at 70 ºC. The solid underwent heat treatment in a tubular furnace with an argon atmosphere at a temperature of 800 ºC for 1 h. After this period, 1.4 mg of the resulting solid was exfoliated in 6 mL of acetonitrile (0.23 mg mL^{-1}) using ultrasound for 6 h. After exfoliation, the dispersion was centrifuged at 3000 rpm for 5 min. The supernatant was used to form the MoS_2 thin film and nanocomposites.

In a 50 mL round bottom flask containing 6 mL of the MoS_2 dispersion, 20 mL of ultrapure water and 20 mL of toluene were added under stirring at 2500 rpm for 3 h. After this period, the system showed the formation of a MoS_2 thin film at the liquid-liquid interface, which was washed by exchanging both the aqueous phase and the organic phase for ultrapure water and toluene 3 times, respectively.

2. Analysis of dispersed films by Phase Contrast Microscopy

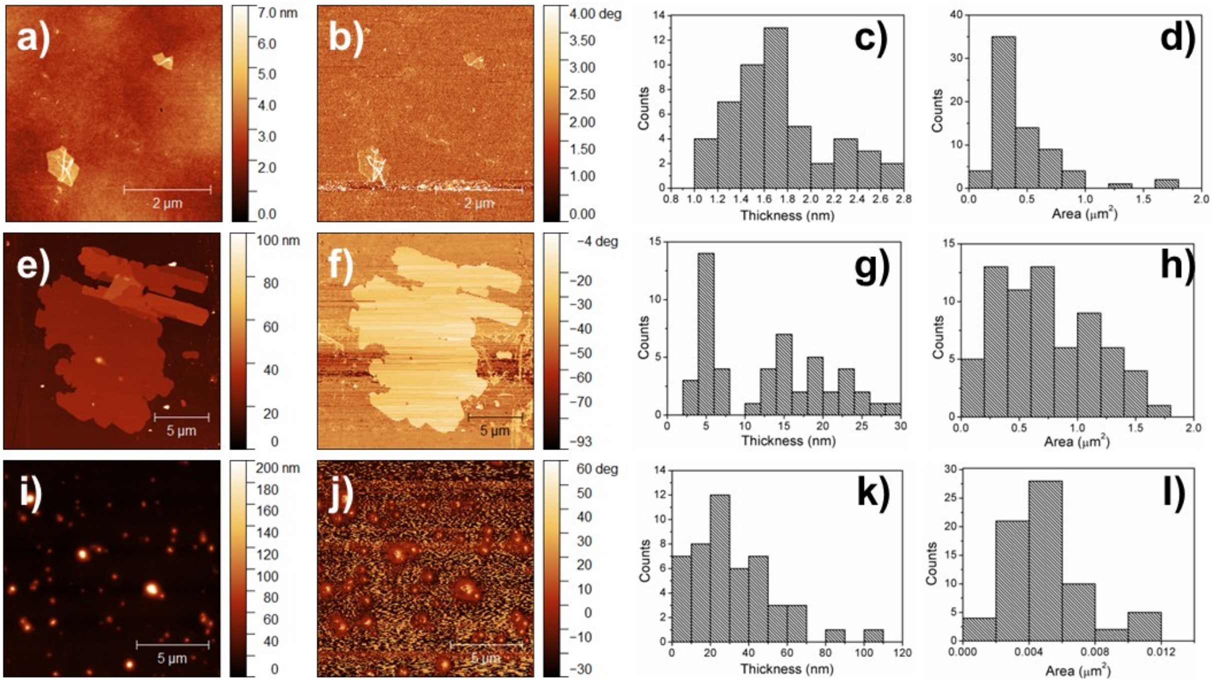

Figure S1. (a,e,i) Topographic and (b,f,j) phase contrast images and their respective (c,g,k) thickness and (d,h,l) area histograms of (a-d) GO-dispersed, (e-h) rGO-dispersed and (i-l) MoS_2-dispersed films deposited on Si/SiO_2 substrates. Several topographic images were used to measure the thickness and area of nanomaterial dispersions.

3. Analysis of dispersed films by Kelvin Probe Force Microscopy

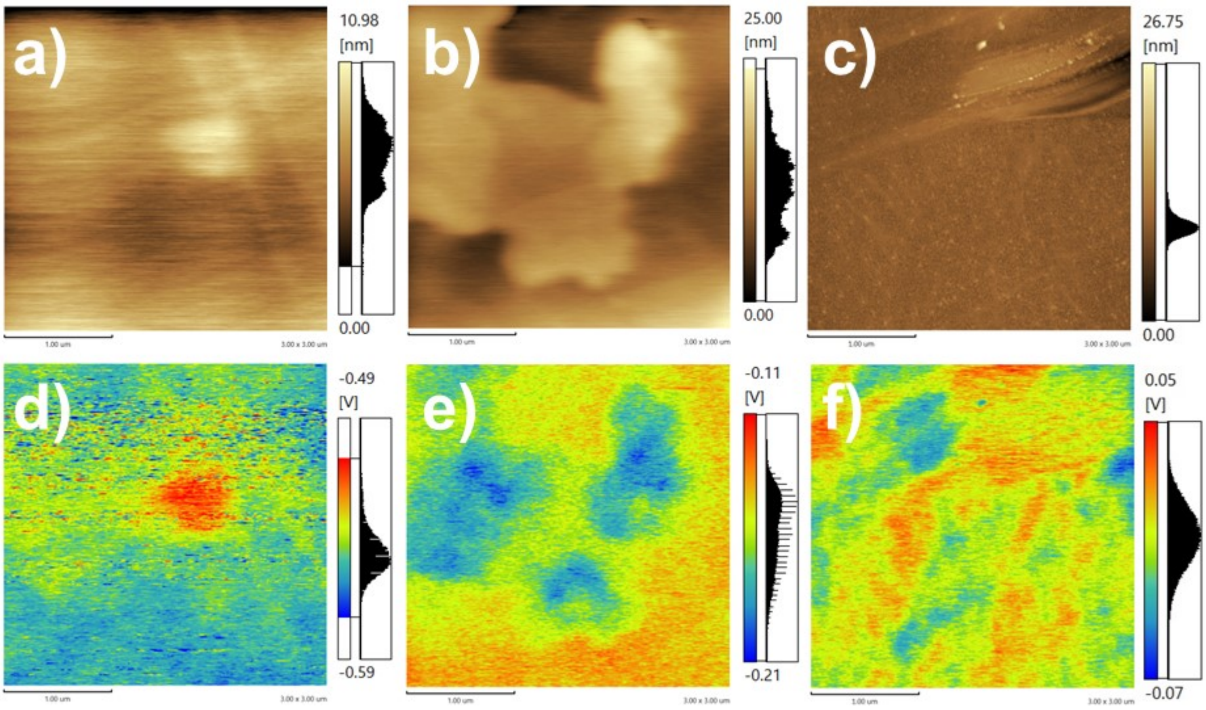

Figure S2. Topographic/KPFM images of (a/d) GO-dispersed, (b/e) rGO-dispersed and (c/f) MoS_2-dispersed films on Si/SiO_2 substrates.

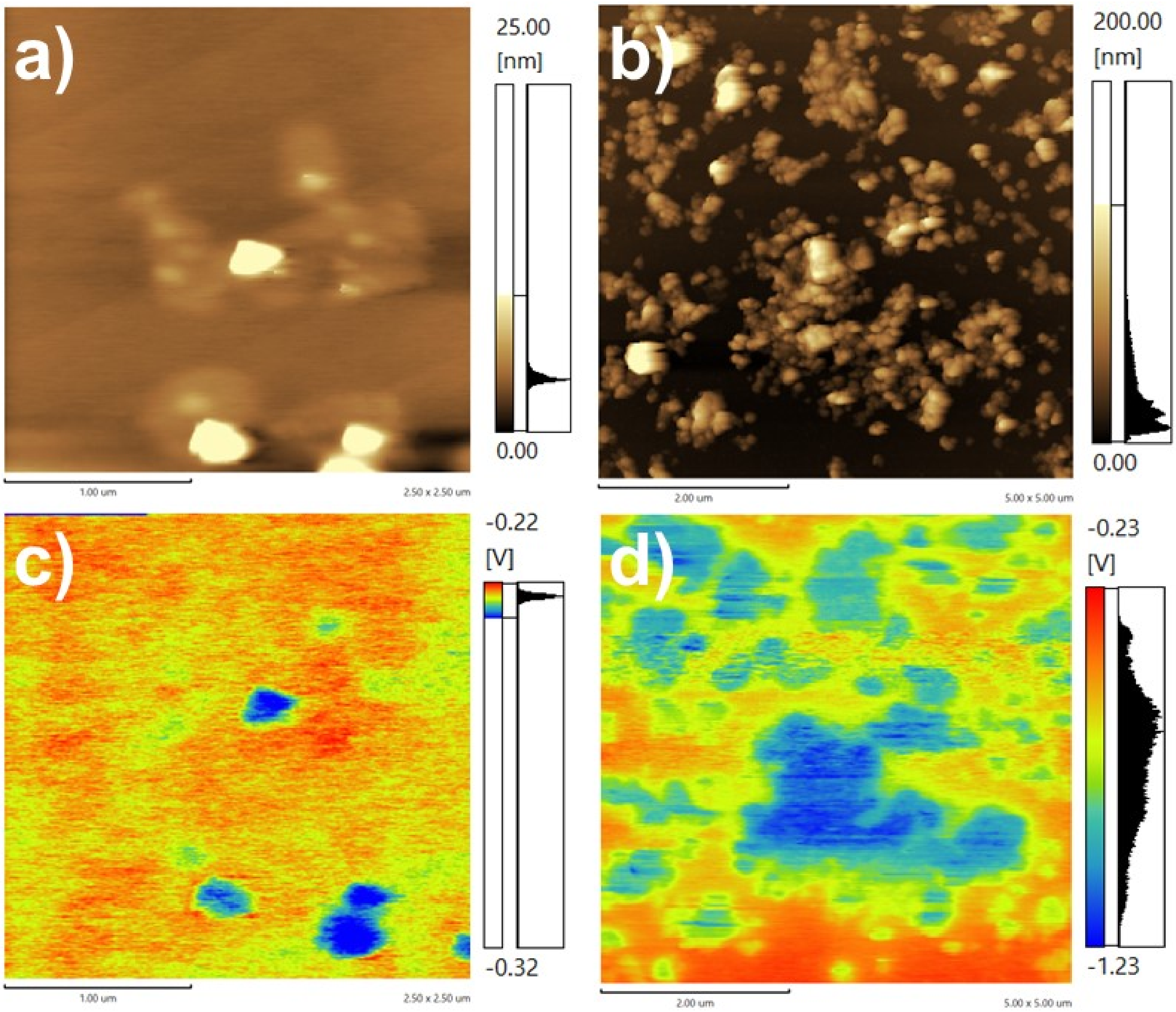

Figure S3. Topographic images of the (a) GO/MoS_2-dispersed and (b) rGO/MoS_2-dispersed films deposited on Si/SiO_2 substrates, and their respective (c,d) KPFM images.

4. Analysis of dispersed films by Lateral Force Microscopy

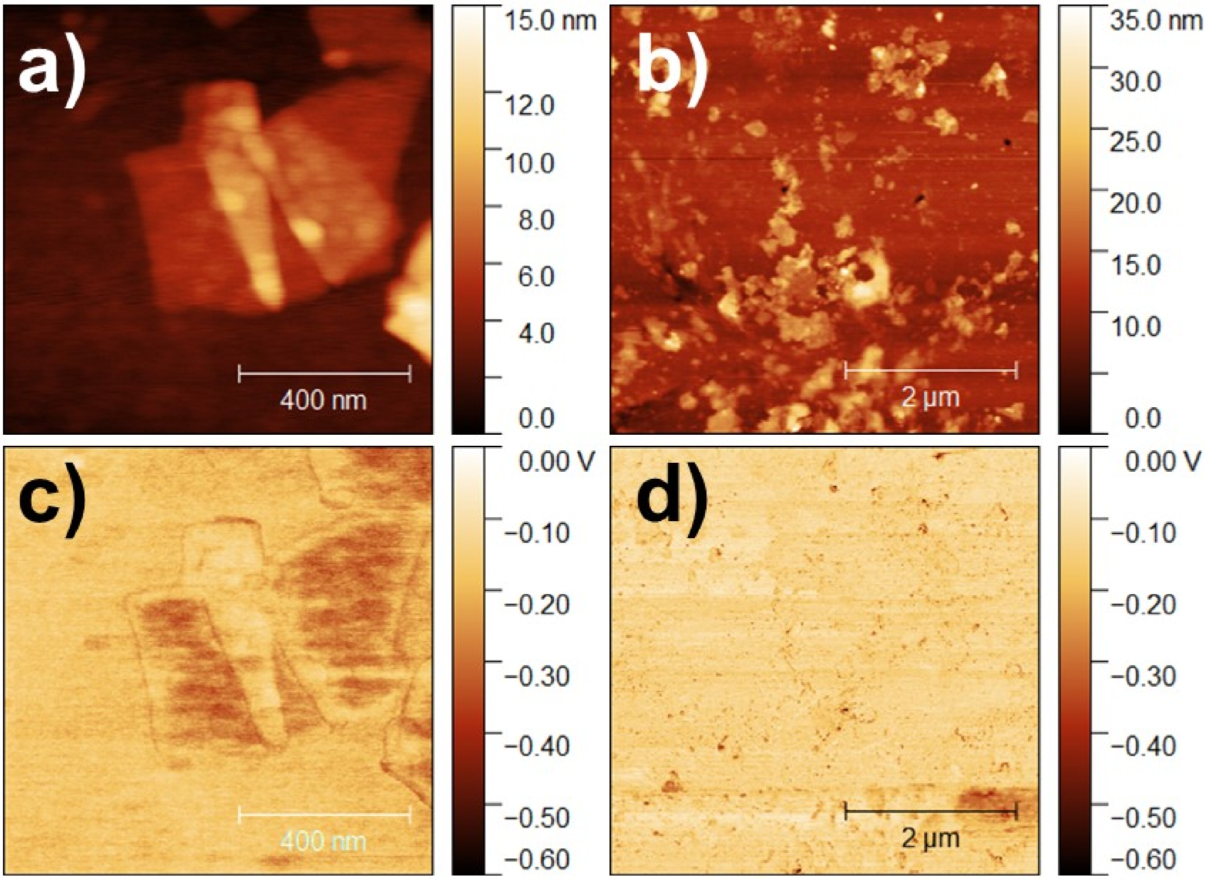

Figure S4. Topographic images of (a) GO/MoS_2-dispersed and (b) rGO/MoS_2-dispersed films deposited on Si/SiO_2 substrate, and their respective (c-d) LFM images. In the LFM images, darker regions are regions with greater friction and lighter regions with less friction. Topographic and LFM images were obtained simultaneously and correspond to the same region.

5. Analysis of dispersed films by PeakForce-QNM®

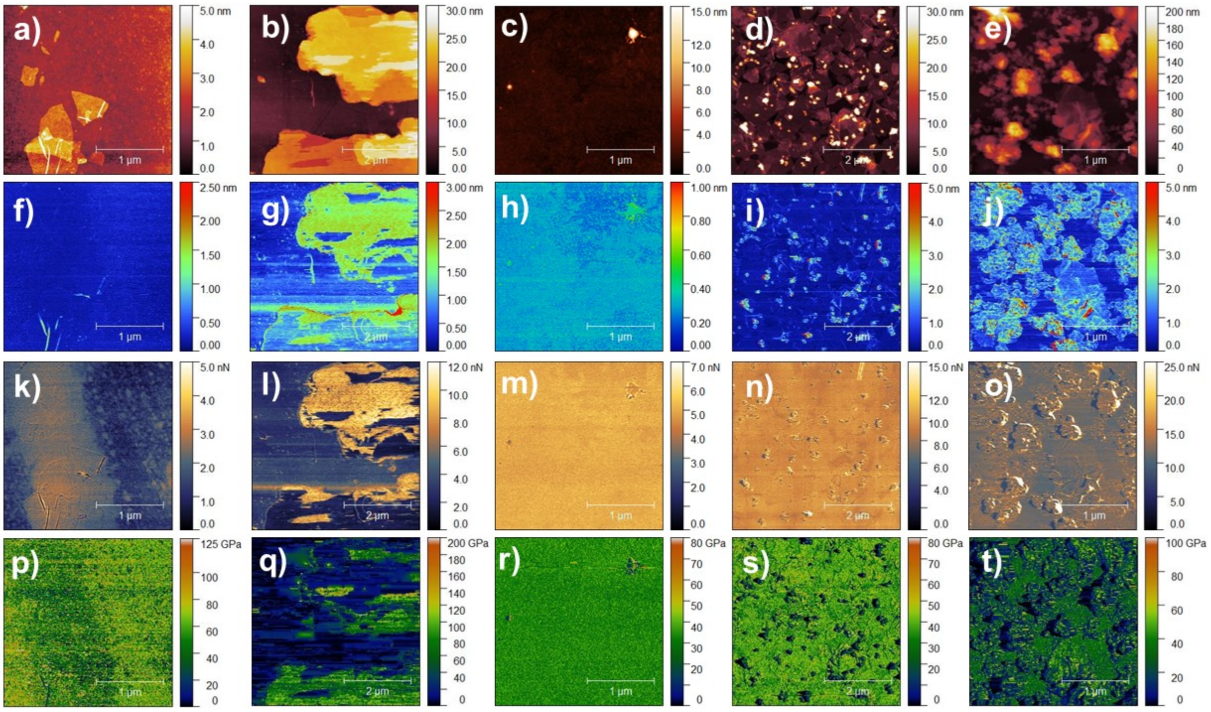

Figure S5. Topographic images of the (a) GO-dispersed, (b) rGO-dispersed, (c) MoS_2-dispersed, (d) GO/MoS_2-dispersed and (e) rGO/MoS_2-dispersed films deposited on Si/SiO_2 substrate, and their respective images of (f-j) deformation, (k-o) adhesion force and (p-t) DMT modulus.

6. Analysis of films by PeakForce-QNM® (force curves)

The operating principle of PeakForce-QNM® involves rapidly executing a force curve at each image pixel to gather data on adhesion force, deformation, and Young's modulus. **Figure S6** displays representative force curves for the analyzed films. In the approach force curve (**Figure S6a**), the deformation regions are identified by the indentation depth approximation. Meanwhile, the retraction force curve (**Figure S6b**) reveals the adhesion, characterized by a lower minimum on the probe withdraw.

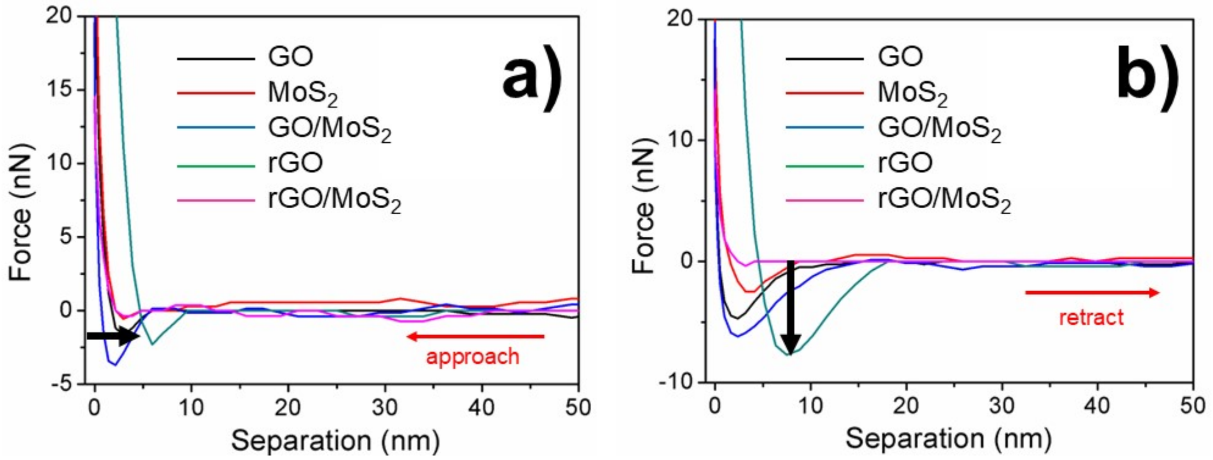

Figure S6. Probe approach (a) and retraction (b) force curves of GO, rGO, MoS_2, GO/MoS_2 and rGO/MoS_2 films.