

Supporting Information

In situ Measurement of Contact Angle and Surface Tension of Interfacial Nanobubbles in Ethanol Aqueous Solutions

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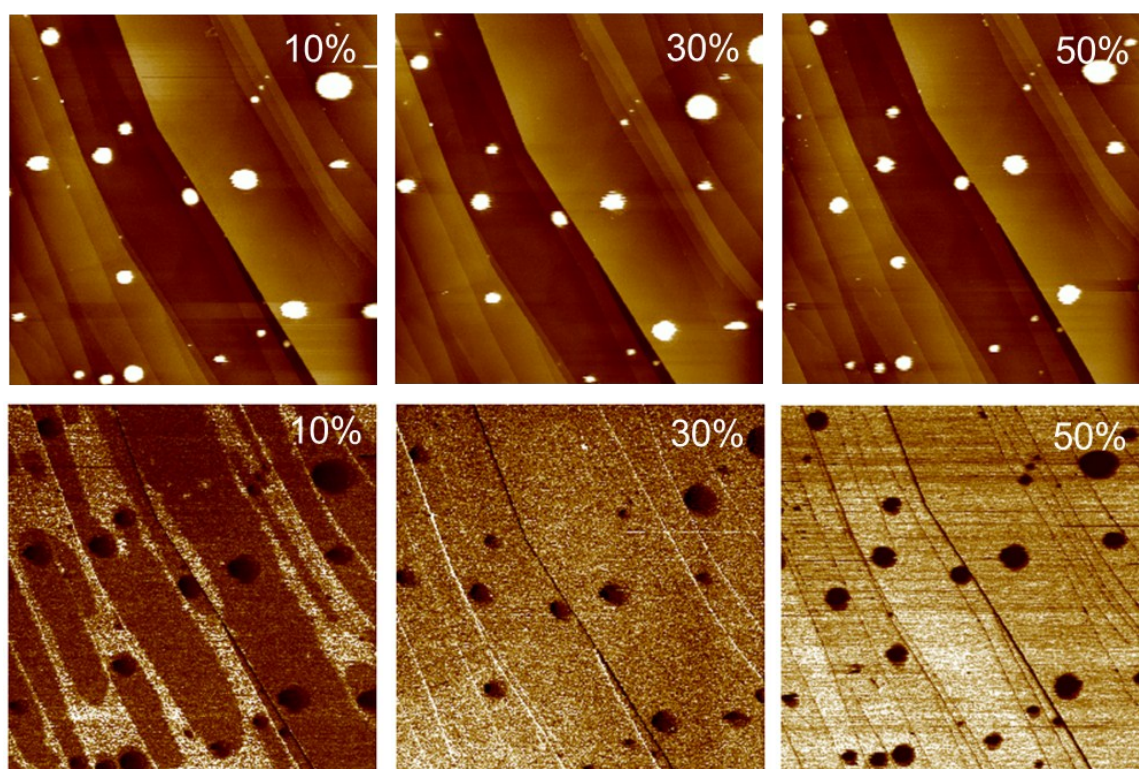


Fig. S1 *In situ* PF-QNM height and stiffness images of nanobubbles in ethanol aqueous solutions with different concentrations (10%, 30%, 50%). Scan size 5 μm \times 5 μm .

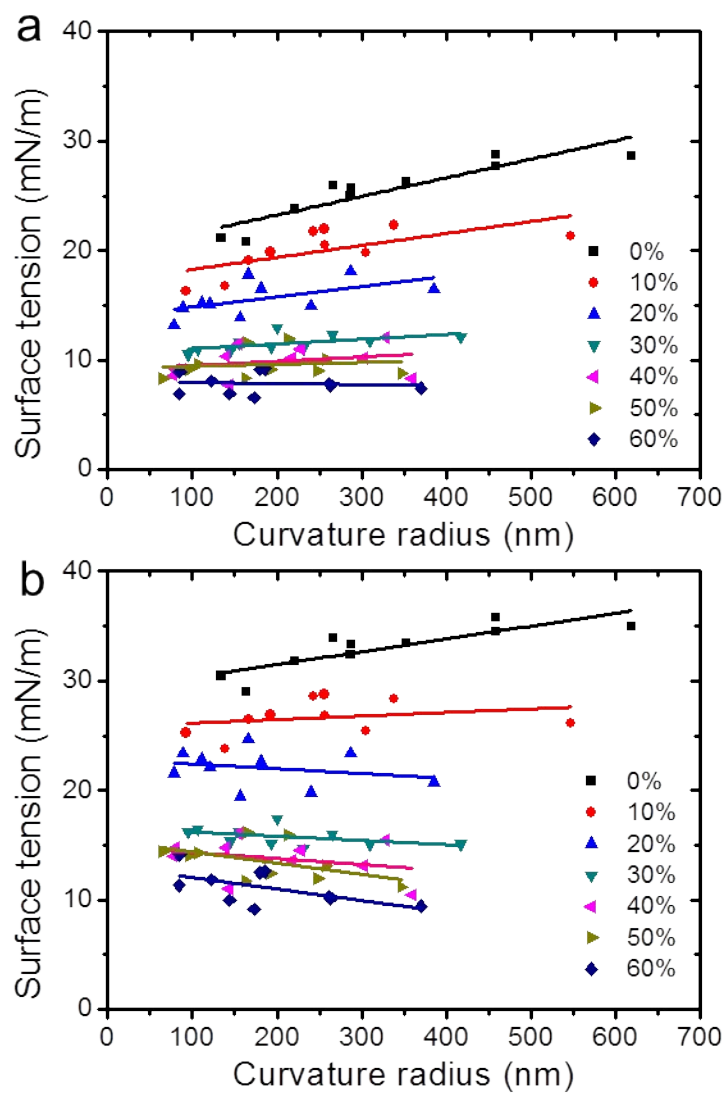


Fig. S2 Size dependence of the surface tensions of nanobubbles in different ethanol aqueous solutions. The surface tensions were determined by method 1 (a) and by method 2 (b), respectively.

Berkelaar *et al.* reported that in some cases the AFM imaged nanobubble-like objects might be induced by PDMS contaminations.¹ However, when we imaged the surface first in water and then in ethanol, no objects were observed on the surface in our experiments (Fig. S3). Another proposed simple method to test whether the objects are nanobubbles or PDMS contaminants is to remove the liquid from the surface and image the same area. Gaseous nanobubbles would disappear when the surface is dry, while PDMS contaminants would leave residues.² Although how reliable this method can be is yet to be confirmed, we checked our samples anyway by following the proposed method. We found that no residues were observed after removing the liquid and then imaging the same area in water again (Fig. S4). The above tests may exclude the possibility of PDMS contamination in our experiments. Furthermore, we can discriminate nanobubbles from the deliberately produced PDMS nanodroplets in another experiments.³

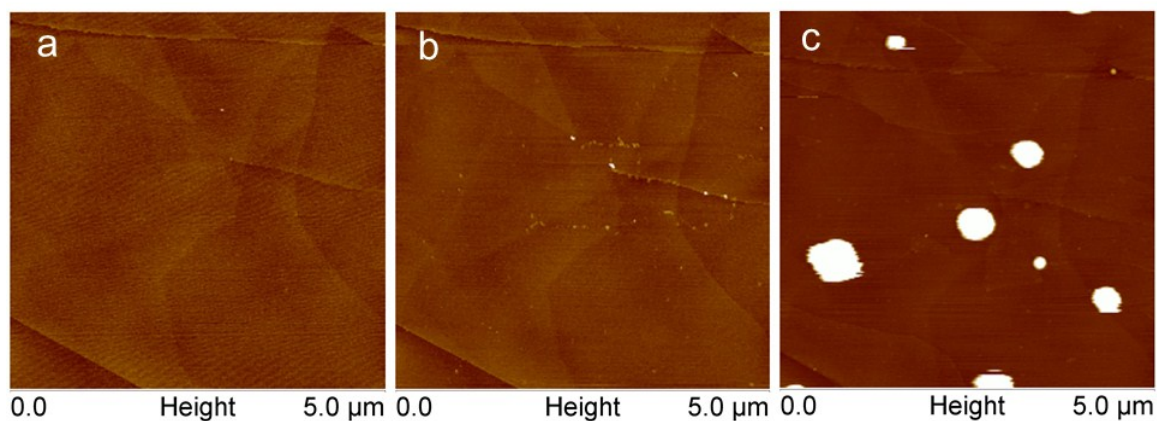


Fig. S3 Height images of HOPG surface during the ethanol-water exchange procedure: (a) in water, (b) in ethanol, and (c) in water again. Scan size $5\mu\text{m}\times 5\mu\text{m}$.

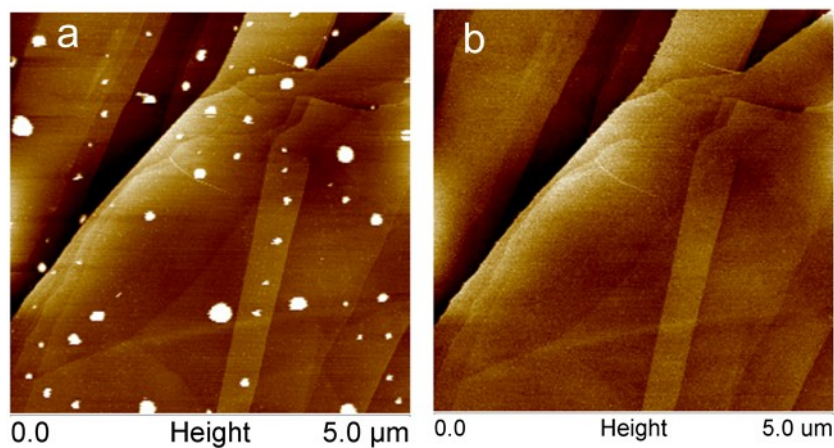


Fig. S4 Height images of HOPG surface: (a) with nanobubbles after ethanol-water exchange, (b) without residues after removing the water and was imaged in air. Scan size $5\mu\text{m}\times 5\mu\text{m}$.

References

1. Berkelaar, R.P. et al. Exposing nanobubble-like objects to a degassed environment. *Soft Matter* **10**, 4947-4955 (2014).
2. An, H.J., Liu, G.M. & Craig, V.S.J. Wetting of nanophases: Nanobubbles, nanodroplets and micropancakes on hydrophobic surfaces. *Advances in Colloid and Interface Science* **222**, 9-17 (2015).
3. Wang, X. Y., et al. Discrimination of nanobubbles from PDMS nanodroplets by Force Volume mode Atomic Force Microscopy. *To be submitted to Langmuir*.