

## Supplemental materials:

# Identification of Structural Defects in Graphitic materials by Gas-phase Anisotropic Etching

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### 1. Surface morphology of graphite samples.

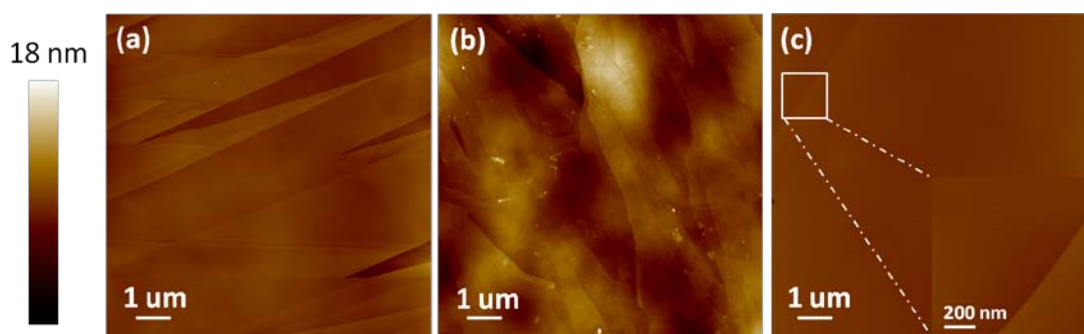


Fig. S1 AFM images of three pristine graphite samples. (a) HOPG-A, (b) HOPG-B, (c) Kish graphite.

### 2. More Raman data on O- and Ar- plasma treated samples.

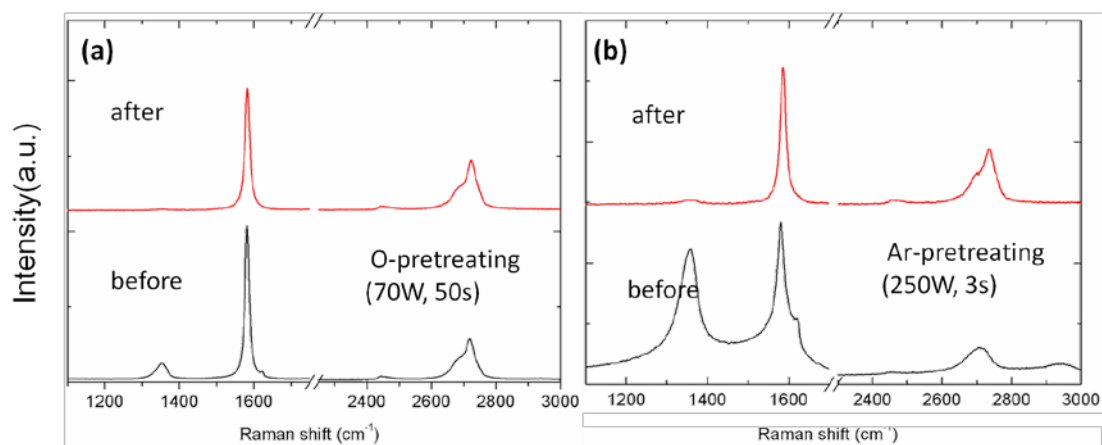


Fig. S2 Raman spectra of graphite with O- and Ar-pretreatment respectively before and after H-plasma etching.

### 3. Further discussions on Figure 5

The magnification of the three dimensional structure of the multilayer hexagonal pits emerging from non-basal dislocations parallel or inclined at some angle to the c-axis is shown in Fig. 5 (c). The stairs-like steps are contained in the pits as well as the walls taper towards the center-bottom. Two different sized pits with one flat and one point bottom are embedded at the flat bottom of a larger multilayer hexagonal area, which is similar in shape with the computation model of shallow hexagonal pits formation on the graphite surface.<sup>1</sup> With the aid of explanation previously reported,<sup>2</sup> flat-bottomed pits shows that their non-basal dislocation ends at voids and grain boundaries lying in or close to the basal plane. Another one non-basal dislocation can be identified from the bottom right inset image of Fig. 5 (c): A multilayer hexagonal pit with a flat bottom is observed, but its inner asymmetric shape contrasting with the common symmetric hexagonal ones suggests that dislocation sites at the inner-bottomed layers shift along the a-axis on the prolong etching. The inference can be made that non-basal dislocation bends at an angle deviating from the c-axis direction within the graphitic crystal and then assume a random direction lying close to the basal plane, combined with the study on similar layer structures.<sup>3-4</sup> Despite the fact that screw dislocation commonly exists in pyrolytic graphite whose density varies from 0.01 to 5  $\mu\text{m}^{-2}$ ,<sup>5</sup> these depression pits aren't either like ones with screw patterns in pyro-graphite,<sup>5</sup> or hexagonal cone-shaped vortex microholes with the spiral cleavage steps produced by electron beam irradiation emerging from a screw dislocation in HOPG<sup>6</sup> or a left handed growth spiral proving the existence of screw dislocation in oxidized Ticonderoga graphite crystal<sup>7</sup> but resemble the pits constituting intersecting arrays forming L-shaped boundary of edge dislocation,<sup>2</sup> so it is reasonable to infer that the multilayer etched pits in the image are more likely the termini of non-basal edge dislocation than that of non-basal screw dislocation.

### References

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