

Electronic Supporting Information

## Direct exfoliation of graphene in organic solvents with addition of NaOH

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### Experiment

#### Materials

Natural graphite flakes with a particle size of about 500  $\mu\text{m}$  were purchased from Qinghao Haoyuan Graphite Company (China). This material was dried to remove the possible adsorption of water. Benzylamine (BA), N-methyl-2-pyrrolidone (NMP), N,N-Dimethylacetamide (DMA), Cyclohexanone (CYC), Benzyl benzoate (BNBZ) were purchased from Sinopharm Chemical Reagent Co., Ltd and used as received.

#### Sample preparation

A set of identical graphene dispersions were prepared by dispersing graphite flakes in organic solvents (10 mL) at an initial concentration of 5 mg mL<sup>-1</sup>. NaOH flakes (~50 mg) were added. These dispersions were sonicated for 1.5 h, followed by centrifugation at 3000 rpm for 60 min. Finally, the supernatant was carefully decanted and retained for further use.

#### Characterization

UV-visible spectrum was recorded on a Nicolet Evolution 300 UV-Visible Spectrophotometer (Thermo Fisher Scientific Inc., USA) using a quartz cell with a 1 cm optical path. **By measuring the absorbance of graphene dispersion at 660 nm, the concentration of exfoliated graphene dispersion was calculated based on Lambert-Beer law.<sup>1</sup>** Transmission electron microscopy (TEM) images and selected area electron diffraction patterns were taken from JEM-2010F (JEOL Ltd., Japan) with an accelerating voltage of 200 kV. Samples for TEM were prepared by drop casting the graphene dispersion onto a holey carbon-coated copper grid. Atomic force microscope (AFM) images of graphene were taken on a commercial scanning probe microscope with a Nanoscope IIIa controller under tapping mode (Digital Instruments, Multimode 3000, USA). Commercial silicon cantilevers were used (Nanosensors, Type NCL-100) with a resonance vibration frequency of ~ 330 kHz. AFM measurements were made by spraying the graphene dispersions onto silicon dioxide substrates, and the solvent was then removed by annealing under gas protection at 400°C for 4 h. Raman spectroscopy were collected on a Renishaw inVia plus laser Raman spectrometer (Renishaw, England) with an excitation laser wavelength of 514.5 nm (5 mW) at room temperature. The obtained materials were placed on a silicon substrate. Powder XRD patterns were measured with a powder X-ray diffractometer (model D/MAX 220/PC, Rigaku, Japan).

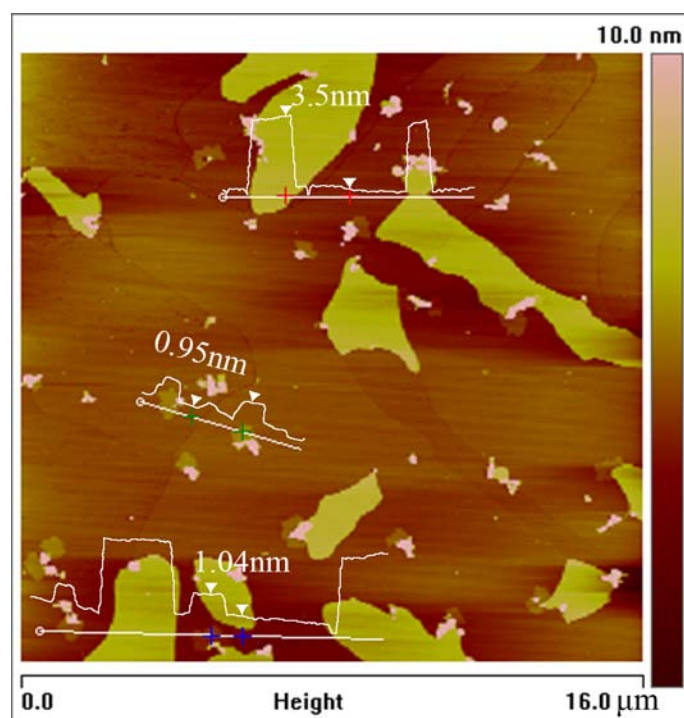


Fig. S1. AFM image of a few graphene flakes and their thickness profiles.

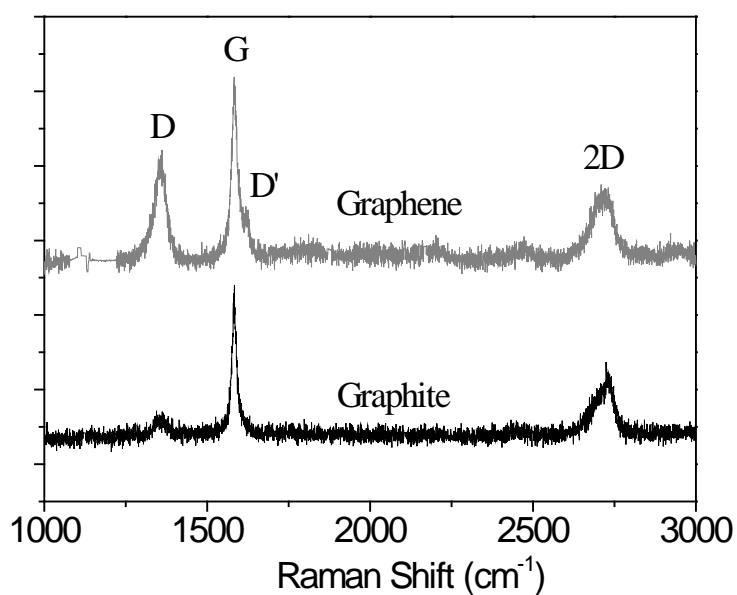


Fig. S2. Raman spectra for bulk graphite and graphene.

Raman spectra for the starting graphite material and the as prepared graphene are shown in Fig. S2. Raman features at approximately 1355 and 1620 cm<sup>-1</sup> are disorder-induced bands, called D and D' bands, respectively. They are usually observed when there is a symmetry-breaking perturbation on the hexagonal sp<sup>2</sup> bonded lattices for graphite, such as structure defects or flake edges.<sup>2</sup> Since the Raman excitation beam has a spot size of ~2 μm and this size is comparable to the sizes of the most graphene flakes, the beam can always “see” a large quantity of graphene

flake edges. As a result, the D band in Fig. S2 appears a little bit broader on graphene than that on graphite, and the splitting of the D' band from the G band can be observed. We suggest that the D band as well as the D' band split from the G band result mainly from the flake edges. The lack of broadening of the G band supports that the D band comes from edges, but not from structural defects inside graphene planes.

The shape of 2D peak varies with the number of layers in graphene flakes,<sup>3</sup> as recognized as a single narrower and sharper peak for monolayer, a band with four component peaks for bilayer, a broadened band for less than five layers, and finally a graphite-type band with two component peaks for more than 5 layers. For more than 5 layers the Raman spectrum becomes hardly distinguishable from that for bulk graphite. The slightly broader 2D peak observed in Fig. S2 for graphene than that for graphite indicates the presence of graphene flakes of a few layers in the present graphene samples.<sup>4</sup>

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