Electronic Supplementary Material (ESI) for ChemComm.

# Supporting Information

## 2D Self-assembly and Electronic Characterization of Oxygen-Boron-Oxygen-Doped Chiral Graphene Nanoribbons

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## Section 1. General Information

**Raman spectra** were measured with laser scanning confocal microscope (Senterra, Bruck) excited at 532 nm. Scanning Raman maps were measured with a WITec alpha 300 R Raman spectrometer.

**UV-Vis-NIR spectroscopy** was performed on a Perkin-Elmer Lambda 900 spectrophotometer.

**Optical images** were taken with laser scanning confocal microscope (Senterra, Bruck). The OBO41-GNRs films were transferred on $SiO_2$/Si substrates for the measurements.

**Scanning electron microscopy** (SEM) was performed on Hitachi S-4800 after transfering the OBO41-GNRs films onto $SiO_2$/Si substrates.

**Ultraviolet photoelectron spectroscopy (UPS)** was performed in the chamber with a HeI UV source based on Einstein photoelectric effect, using an instrument from Kratos Axis supra. $E_b$ (binding energy) + $E_k$ (kinetic energy) = 21.22 eV; the Fermi level energy is 0 eV.

**Atomic Force Microscopy (AFM) measurements** were performed in intermittent contact mode using an Asylum Research Cypher ES AFM instrument. Olympus silicon cantilevers (OMCL-AC160TS; drive frequency 200–400 kHz and OMCL-AC240TS; drive frequency 50–90 kHz) were used.

**Scanning Tunneling Microscope (STM) experiments** were performed directly on the dry samples at room temperature (20–22 °C) using a PicoLE (Keysight) or Molecular Imaging STM system in constant-current mode. STM tips were prepared by mechanical cutting from Pt/Ir wire (80/20, diameter 0.25 mm, Advent Research Materials). AFM and STM images were processed using Scanning Probe Image Processor (SPIP, Image Metrology ApS) and WSxM (Nanotec Electronica, Spain) software.[S1] Molecular models were built using Hyperchem Professional 7.5.

## Section 2. Materials and Methods

### Materials

6,16-Dibromo-9,10,19,20-tetraoxa-9a,19a-diboratetrabenzo[*a*,*f*,*j*,*o*]perylene (DBTD) monomers were synthesized following previously reported protocol.[S2] Gold grains (99.999%) were purchased from VNANO Vacuum Technology Co., Ltd. Mica sheets were pruchased from Yasheng Electronic Technology Co., Ltd. Silicon wafers were purchased from Heifei kejing material technology Co., Ltd. Au etchant and polymethyl methacrylate (PMMA) were purchaesd from Simga-Aldrich.

### Methods

Before the synthesis of OBO41-GNRs, approximately 150 nm thick gold films were prepared via evaporation on spectific substrates. The vacuum evaporators is from VNANO Vacuum Technology Co., Ltd. Then the gold films were annealed by flamer for about 1 min. The CVD system includes tube furnace, vacuum pump and gas supply system (OTF-1200x, Heifei kejing material technology Co,. Ltd.) as well as a heating belt (Thermocoax Isopad S20). During the growth of OBO41-GNRs, Au (111)/mica or Au (111)/$SiO_2$/Si were loaded into tube fornace and heated to 250 °C under 125 sccm of Ar and 25 sccm of $H_2$. The pressure of the tube fornace was kept at ～1.0 torr. Meanwhile DBTD monomers were sublimed at 305-320 ℃ for 30 min via the heating belt, and deposited on the substrates to induce dehalogenation and radical addition. Finally, the samples were heated to 450 °C for 15 min to induce surface-assisted intramolecular cyclodehydrogenation, resulting in the atomically precise OBO41-GNRs.[S3]

The transfer of the CVD-grown OBO41-GNRs was operated by the following steps. Firstly, PMMA (4 wt%, Simga-Aldrich ) films were spin-coated on the as-grown OBO41-GNRs and dried at 125 °C. Secondly, OBO41-GNRs/mica or OBO41-GNRs/$SiO_2$/Si were placed on the surface of hydrofluoric acid solution (HF, 40%) or concentrated hydrochloric acids to etch mica or oxide layer. After the gold films were seperatedly from the underlying substrates, the samples were washed by deionized water for three times. Thirdly, the gold films were etched away by Au etchant (Simga-Aldrich), and the GNR films were transferred on target substrates. Finally, the PMMA films were dissolved by hot acetone at about 50 °C.[S4]

# Section 3. Atomic Force Microscopy Characterization of CVD-grown OBO41-GNRs

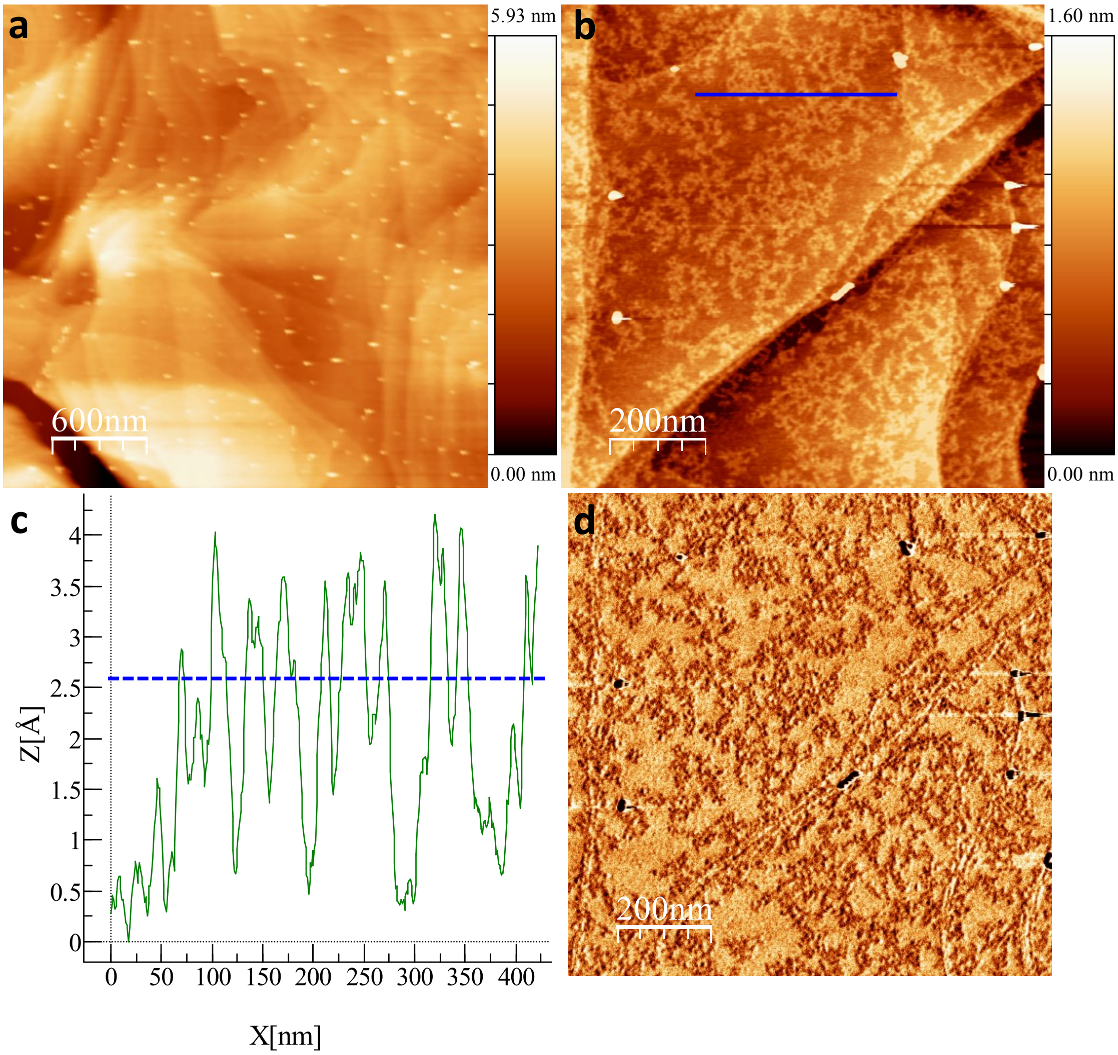

**Figure S1.** Atomic Force Microscopy (AFM) characterization of the CVD-grown OBO41-GNRs films supported on Au (111)/mica. (a) Large 3 × 3 $\mu m^2$ AFM topography image with a RMS roughness of 0.8 nm. (b) Smaller 1 × 1 $\mu m^2$ AFM topography image with a RMS roughness value of 0.2 nm; along with (d) phase image. (c) Line profile along the blue line in (b).

# Section 4. Additonal Scanning Tunneling Microscope Images of CVD-grown OBO41-GNRs

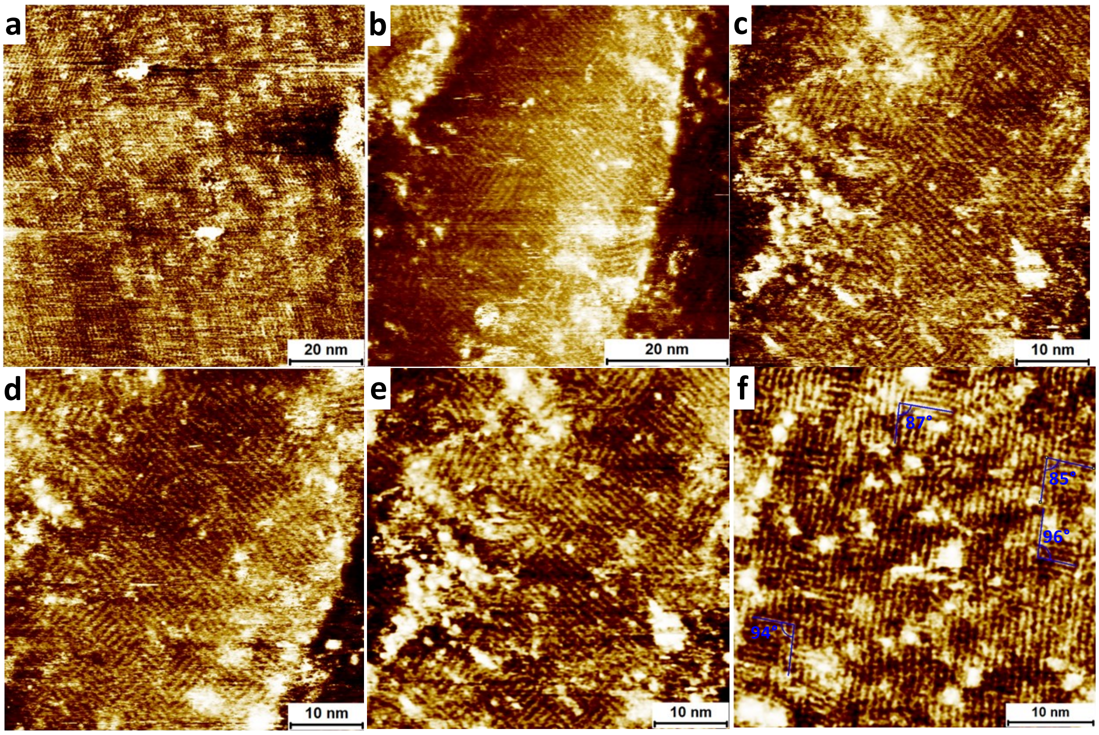

**Figure S2.** Additional STM images revealing a unique OBO41-GNRs network (~90 ± 6° angles).

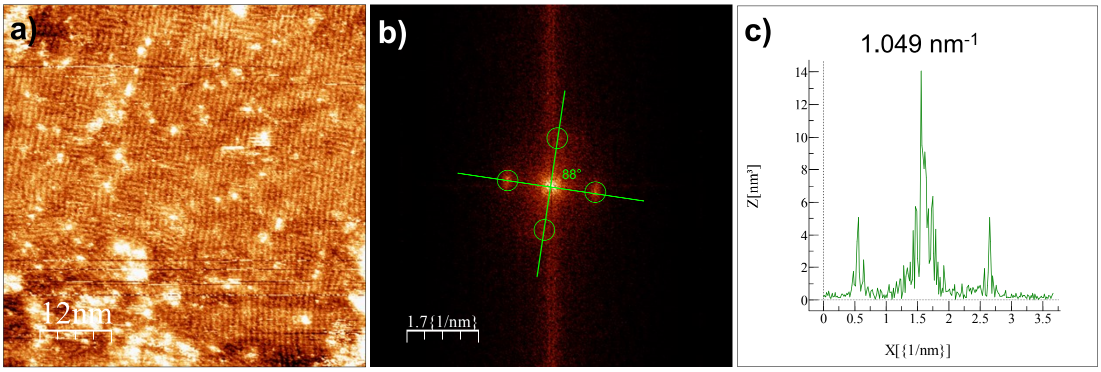

**Figure S3.** 2D-FFT analysis of the OBO41-GNRs. (a) Large scale STM image. (b) 2D-FFT spectrum showing the approximate orthogonal orientation of the GNRs. (c) Periodicity of the GNRs from the 2D-FFT of the STM image.

## Section 5. Structural Model for the 2D Self-assembly of OBO41-GNRs

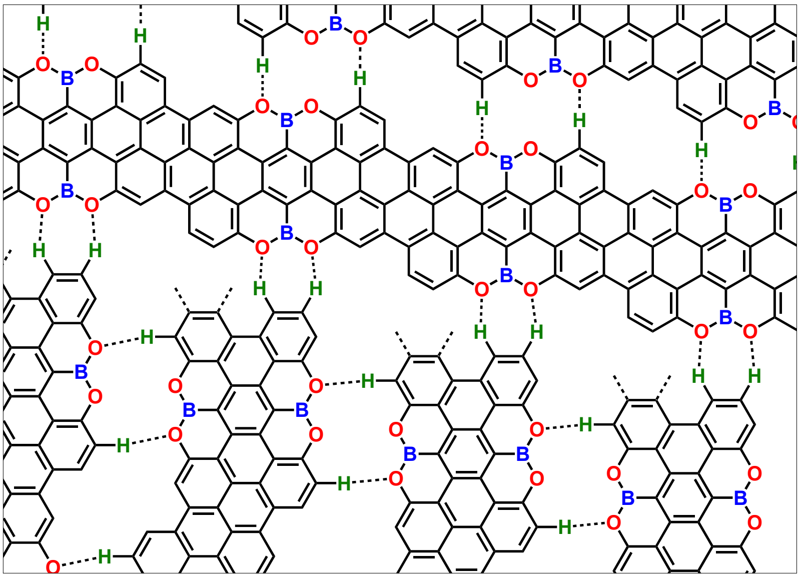

**Figure S4.** Structure model for the 2D self-assembly of OBO41-GNRs, showing the possible edge-to-edge and edge-to-end O···H hydrogen bonds.

## Section 6. Scanning Raman Mapping of CVD-grown OBO41-GNRs

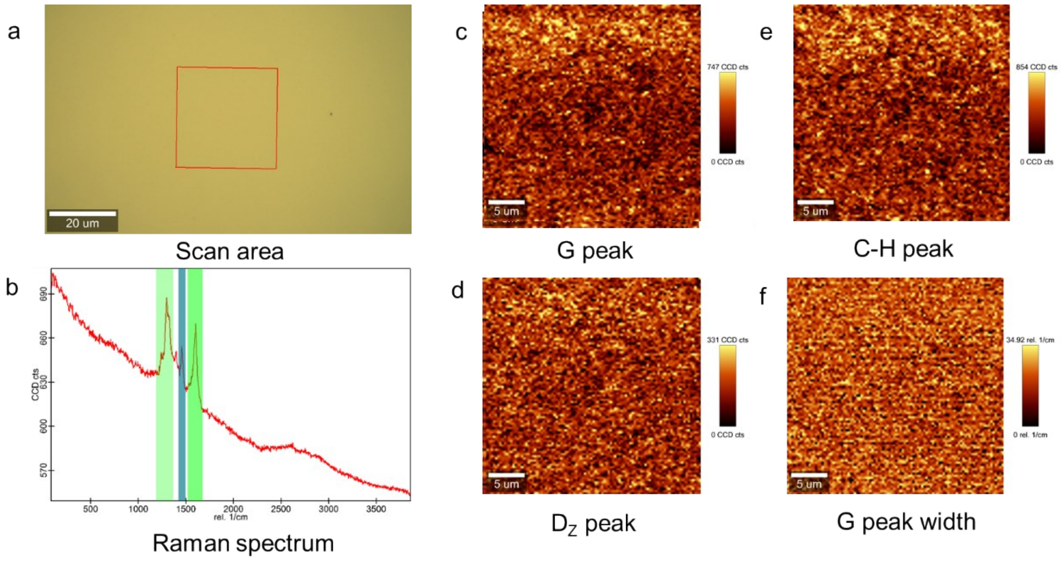

**Figure S5.** The quality and the uniformity of CVD-grown OBO41-GNRs were further characterized via Raman mapping. All the mapping graphs showed almost homogenization and uniform distribution, verifying the quality and homogeneity of the OBO41-GNRs.

## Section 7. Raman Spectra of OBO41-GNRs Transferred by Using Concentrate Hydrochloric Acid

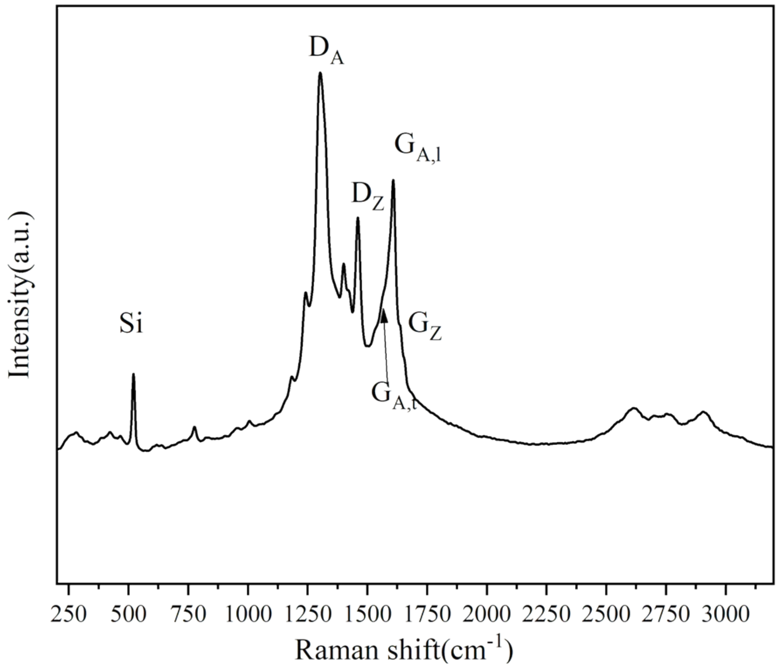

**Figure S6.** Raman spectra of OBO41-GNRs transferred by using concentrate hydrochloric acid (excited at 532 nm laser).

## Section 8. Supporting References

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