Electronic Supplementary Material (ESI) for Nanoscale

Supporting Information

Large Networks of Vertical Multi-Layer Graphenes with Morphology-Tunable Magnetoresistance

Z. J. Yue,[1,2] I. Levchenko[2,3], S. Kumar,[2,3] D. H. Seo, [2,3] X. L. Wang,[1] S. X. Dou,[1]

and K. Ostrikov,[2,3,1]

[1] Institute for Superconducting and Electronic Materials (ISEM), Faculty of Engineering, University of Wollongong, NSW 2522, Australia

[2] Plasma Nanoscience Centre Australia (PNCA), CSIRO Materials Science and Engineering, P. O. Box 218, Lindfield, NSW 2070, Australia

[3] Plasma Nanoscience, School of Physics, The University of Sydney, Sydney, NSW 2006, Australia.

Table of contents

1. Schematic of the inductively coupled plasma-enhanced CVD reactor

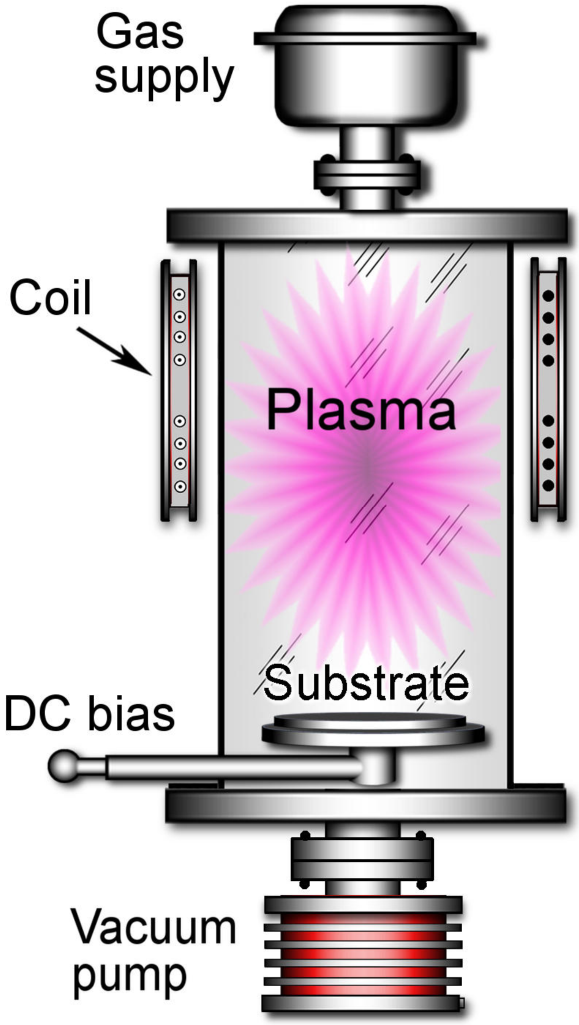

Fig. S1 Schematic of the inductively coupled plasma-enhanced CVD reactor.

Working frequency 13.56 MHz, input power up to 1.0 kW.

2. SEM and TEM images of the petal-like and tree-like graphene networks

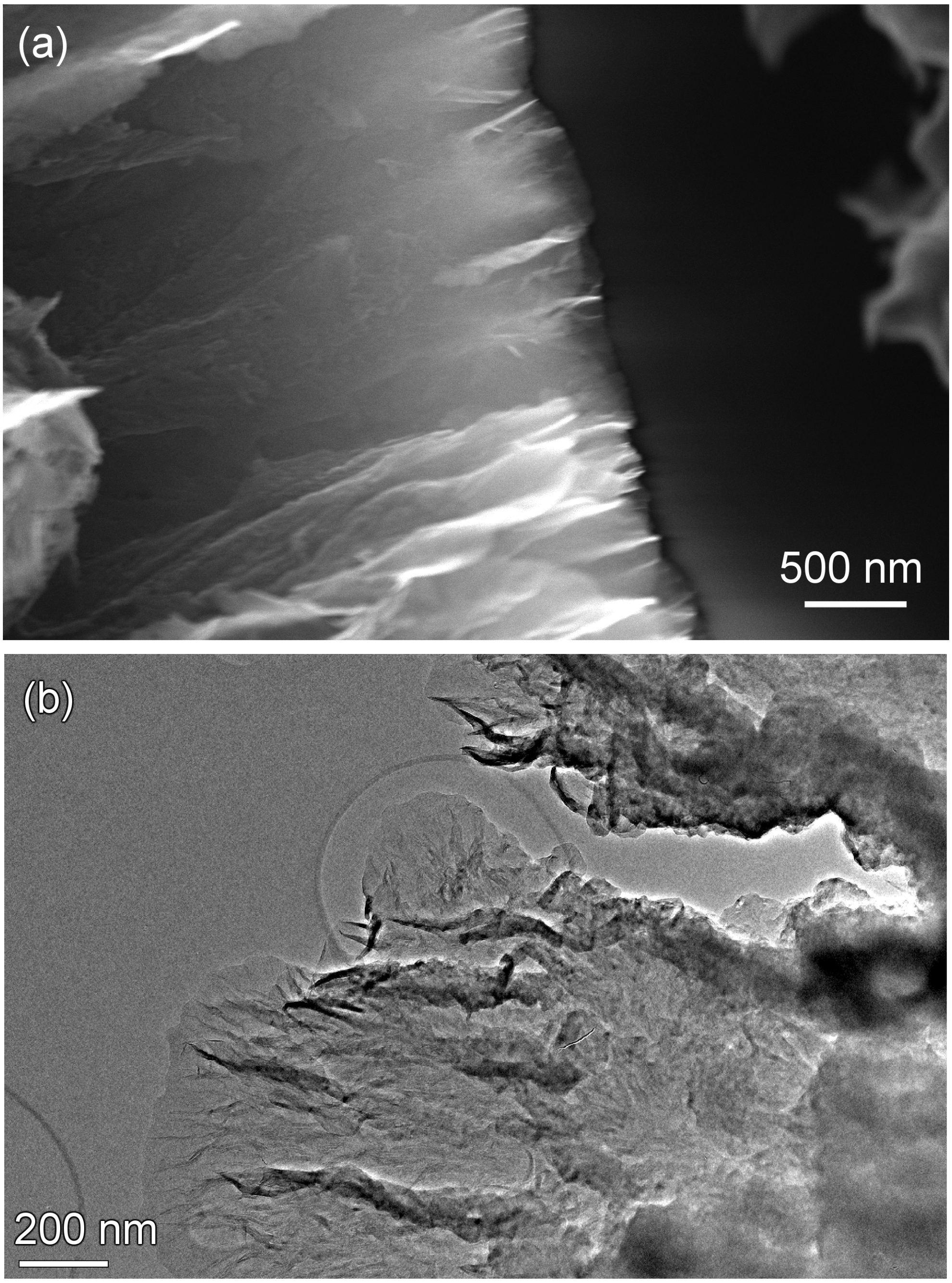

Fig. S2 SEM (a) and TEM (b) images of the petal-like graphene networks

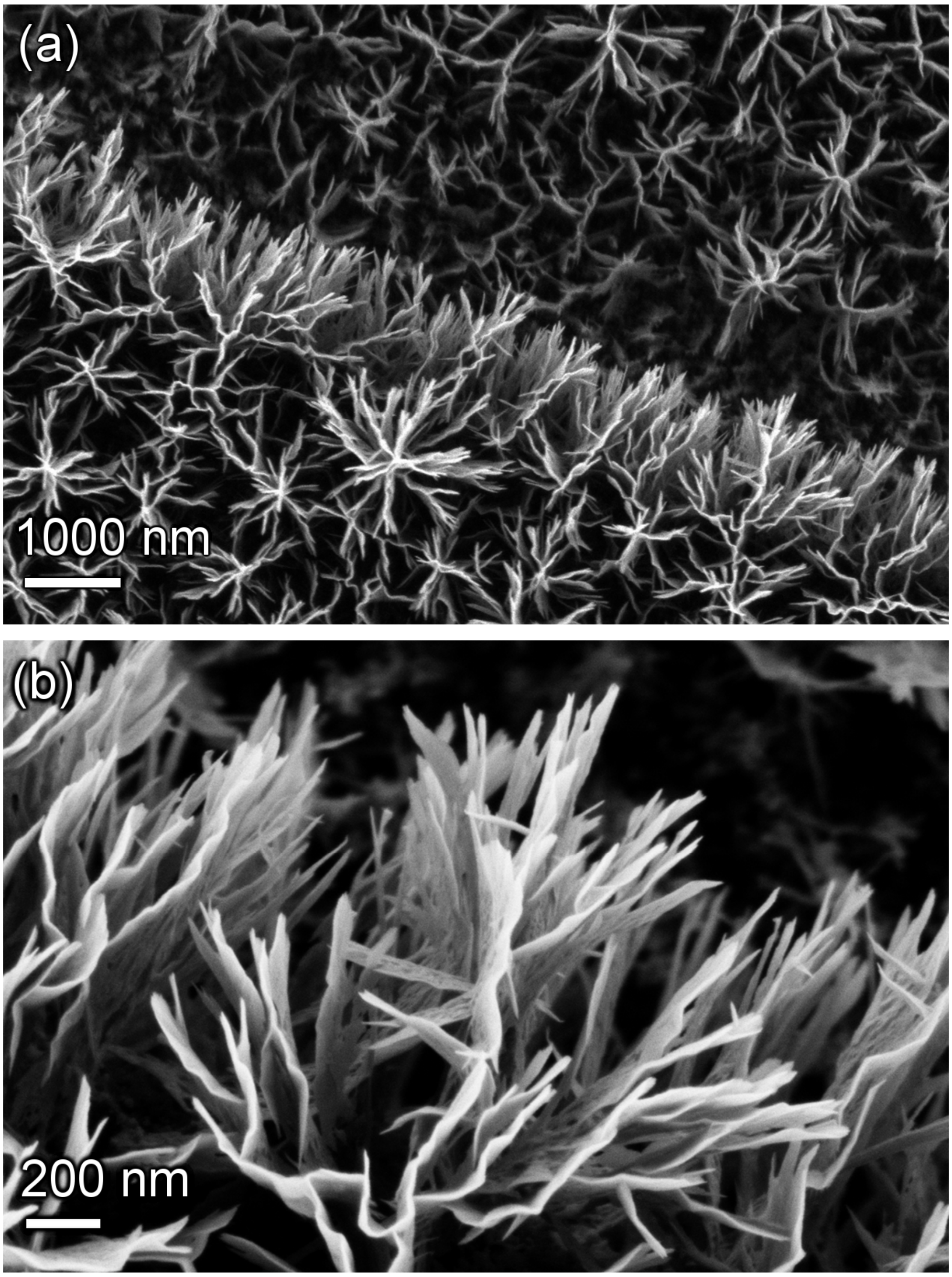

Fig. S3 Low (a) and high (b) resolution SEM images of the tree-like graphene networks

3. 3D representation of the petal-like and tree-like graphene networks

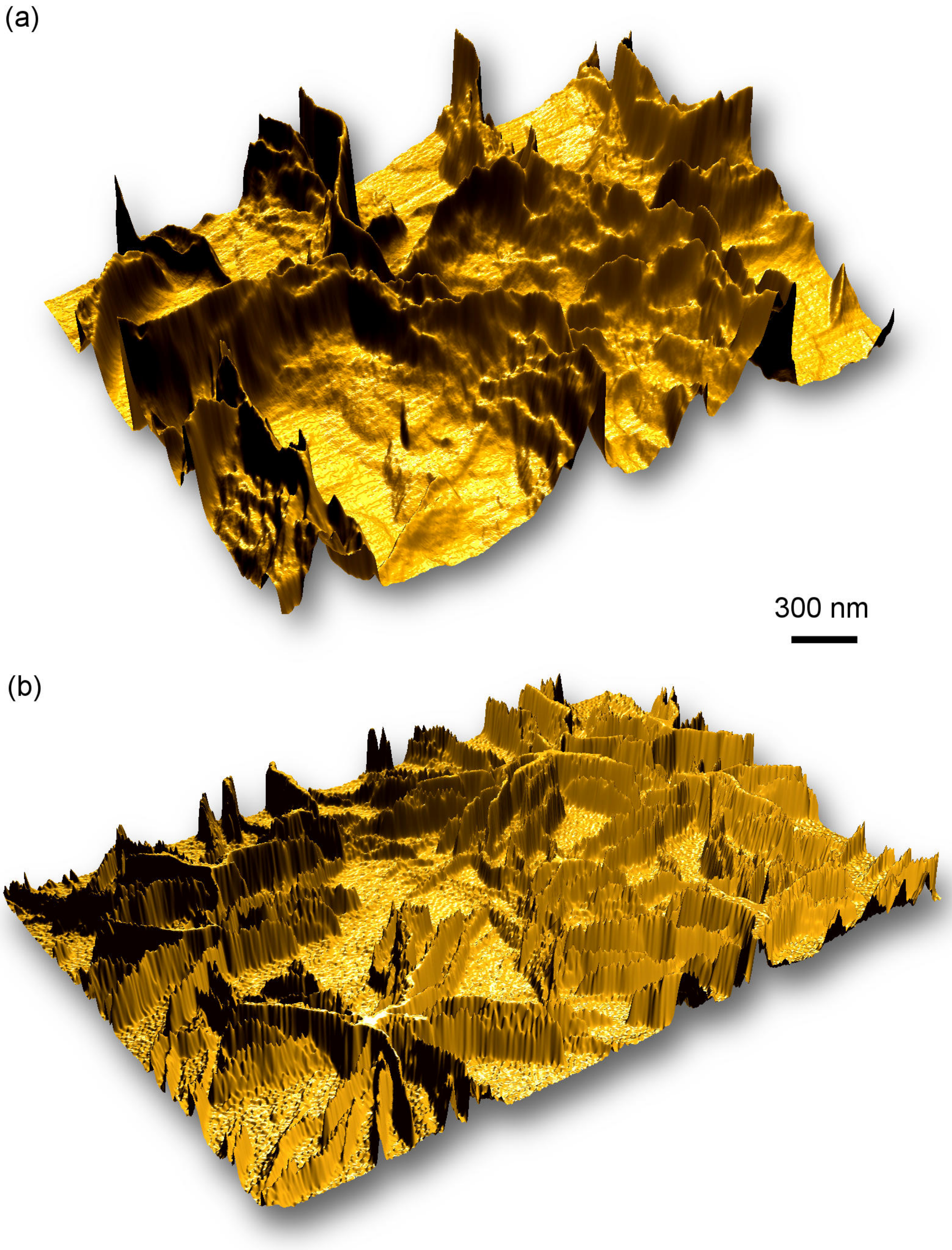

Fig. S4 3D representation of the petal-like (a) and tree-like (b) graphene networks

4. Details on the electrical transport measurements

Electrical transport properties of the vertical graphene networks were investigated using a 14 T Physical Properties Measurement System (PPMS). The resistances were obtained by applying a fixed electric current of 10 μA through the two outer silver contacts (see schematic of measurements in Fig. 1 in the paper text) and monitoring the voltage drop between the two inner contacts. The temperature dependences of the networks resistivity were measured at a zero magnetic field.

Figure S5 shows that the resistance of the petal-like graphene network decreases with temperature, thus demonstrating a semiconductor-like behaviour. This network have a relatively low order of graphitization with quite significant defects at ultra-long edges and boundaries, where the π-electrons could be trapped and get to the localized states. Charge carriers can easily move through the variable range hopping based on the localized states.

The Mott variable range hopping model describes conduction in strongly disordered systems with localized states and is expressed as

$$R = R_0 \exp\left(\frac{T_0}{T}\right)^{\frac{1}{d+1}}, \tag{1}$$

where d is the dimensionality, R_0 is a constant, and T_0 is the Mott characteristic temperature depending on the electronic structure[1]. The temperature dependence of the resistance of graphene network has been analyzed using equation (1).

As shown in the inset in Fig. S5, the dependence of $ln(R)$ versus $T^{-1/4}$ (d = 3) shows a quite linear fitting, thus indicating that the 3D VRH conduction controls the electron transport in petal-like graphene networks.

The $R(T)$ dependence for the tree-like graphene network is shown in Fig. S6. This dependence also displays a semiconductor-like electrical behaviour at low temperatures. Compared with petal-like network, the tree-like network has nanoribbon-like boundaries and this can lead to the formation of necks at the contact points of vertical graphene flakes. Irregularities on the boundaries also may induce formation of the constrictions and quantum dots, which lead to a remarkable electron-electron correlation.[2] These nanoribbon-like necks cause an abrupt reduction of conducting channels and thus result in quantum confinement effects and Coulomb blockade.[3] The connected edge points act as bottlenecks which eventually determine the conductance of the tree-like network. The *R(T)* data for the tree-like networks can be well fitted with a percolation model, as shown in the inset of Fig. S6.

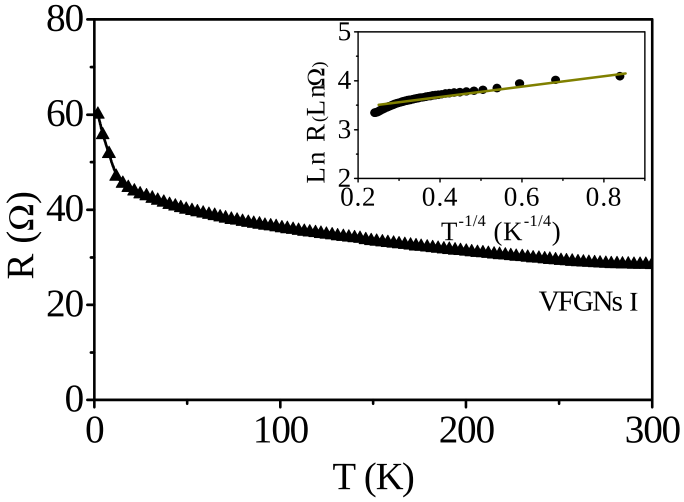

Fig. S5 Resistance as a function of temperatures in petal-like graphene network at a zero magnetic field. The inset shows the *R(T)* fitting with the VRH model.

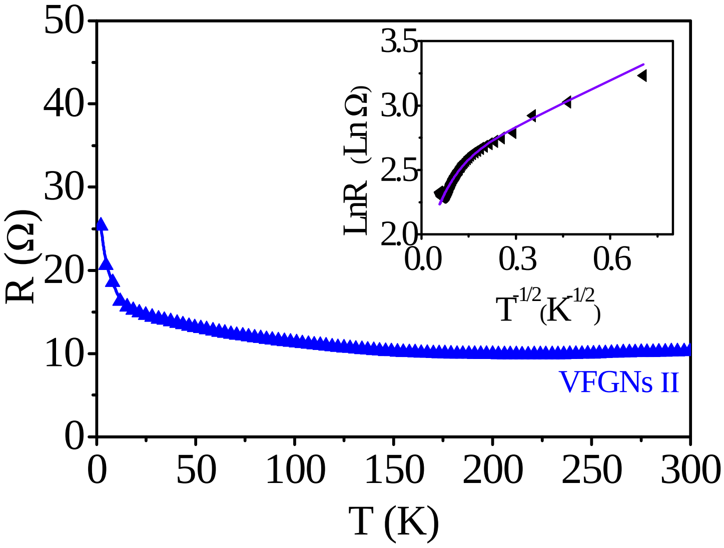

Fig. S6 Resistances as a function of temperatures in tree-like graphene network at zero magnetic field. The inset shows the R(T) fitting with percolation model.

References

1. N. F. Mott, *Philos. Mag.,* 1969, **19**, 835-852
2. V. N. Kotov, B. Uchoa, V. M. Pereira, F. Guinea, and A. H. Castro Neto, *Rev. Mod. Phys.,* 2012, **84**, 1067–1125.
3. F. Sols, F. Guinea, A. H. C. Neto, *Phys. Rev. Lett.,* 2007, **99**, (16), 166803.