

Supplementary information

Engineering the magnetism of nanographene via the co-deposition method

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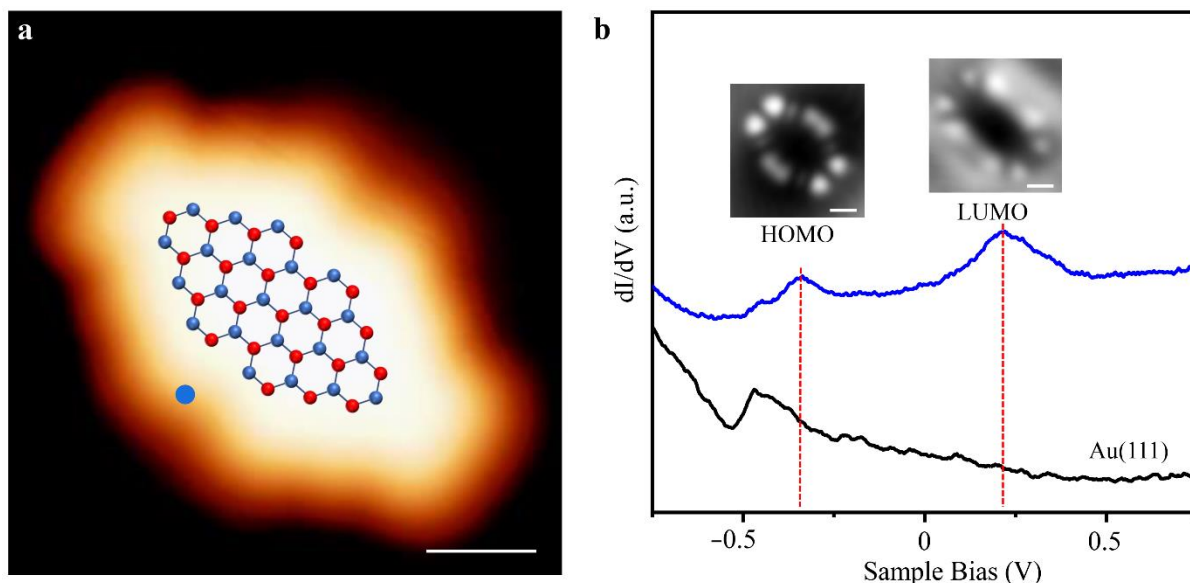
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Supplementary Figures S1-S6 describing supporting scanning tunneling microscopy and spectroscopy data:

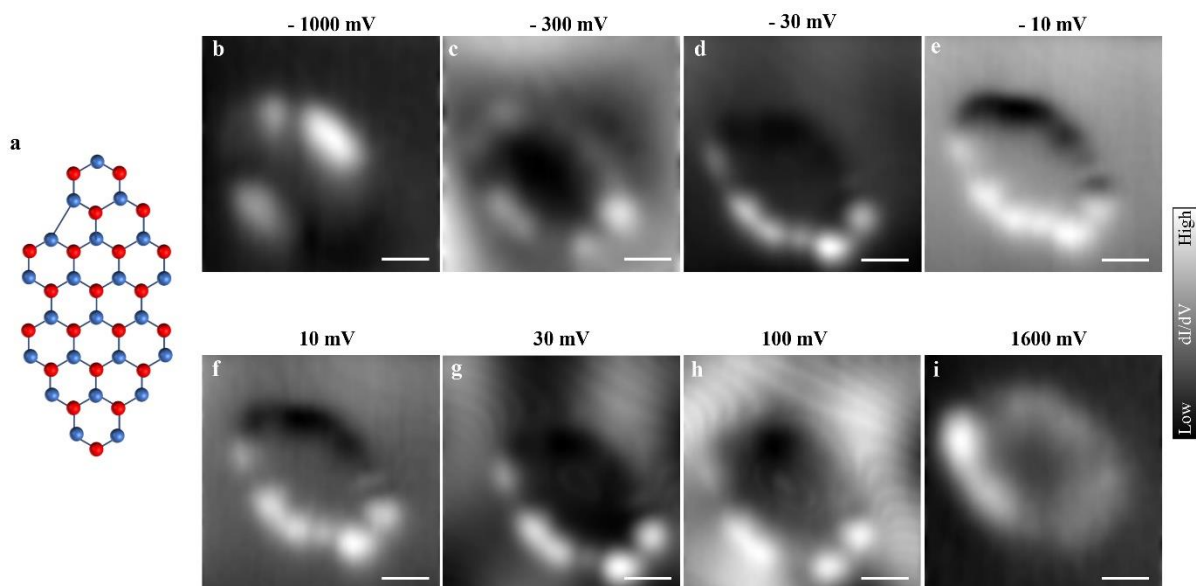
1. Characterizing the frontier molecular orbital of NG 1a
2. Additional dI/dV maps of NG 1b
3. Characterizing the Kondo resonance of NG 1b
4. Additional dI/dV maps of NG 1b
5. The radical positions of NG 2a of 6 Clar sextets
6. A comparison of BR-STM images of NG 1a and 1b

Supplementary Table S1 describing supporting comparison of close and NG 1a and altered NG 2a

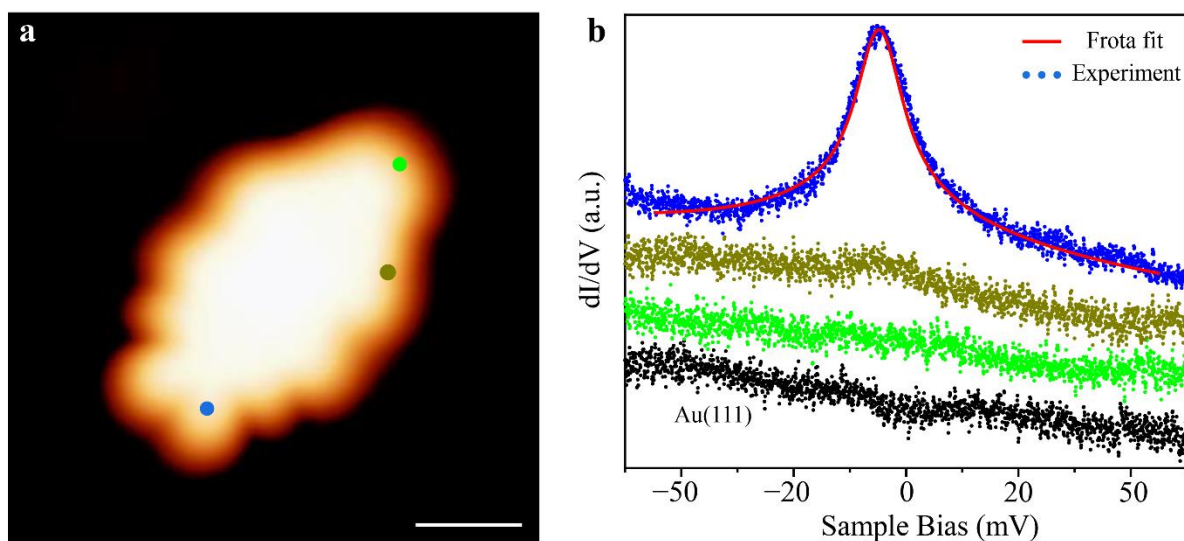
Supplementary Figures



Supplementary Figure 1 | Characterizing the frontier molecular orbital of NG 1a. (a) STM image and overlaid chemical structure of NG 1a. The red and blue balls represent the carbons from two different sublattices. Tunneling parameters: $V = -500$ mV, $I = 200$ pA. Scale bar: 6 Å. (b) The dI/dV spectra with zoomed-in energy resolution (from -0.75 V to $+0.75$ V) and the frontier molecular orbital of HOMO (-0.3 V) and LUMO ($+0.25$ V) are marked with red dotted lines. The corresponding dI/dV maps of HOMO and LUMO are attached and the node-like state distribution can be recognized. Tunneling parameters: $V = -1000$ mV, $I = 300$ pA, $V_{rms} = 10$ mV.

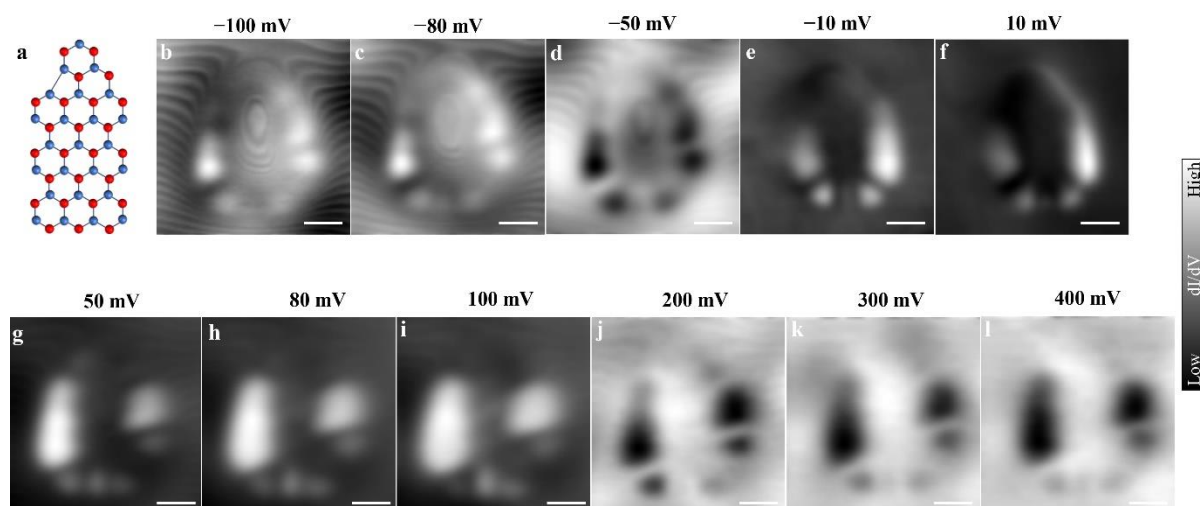


Supplementary Figure 2 | Additional dI/dV maps of NG 1b. (a) The chemical structure of NG 1b. The red and blue balls represent the carbons from two different sublattice. (b-j) dI/dV maps of various bias voltage. The dI/dV maps under the bias voltage of -30 mV and $+30$ mV show the similar appearance which indicate the frontier molecular orbital of SOMO and SUMO. Tunneling parameters: $I = 350$ pA. Scale bar: 6 \AA .



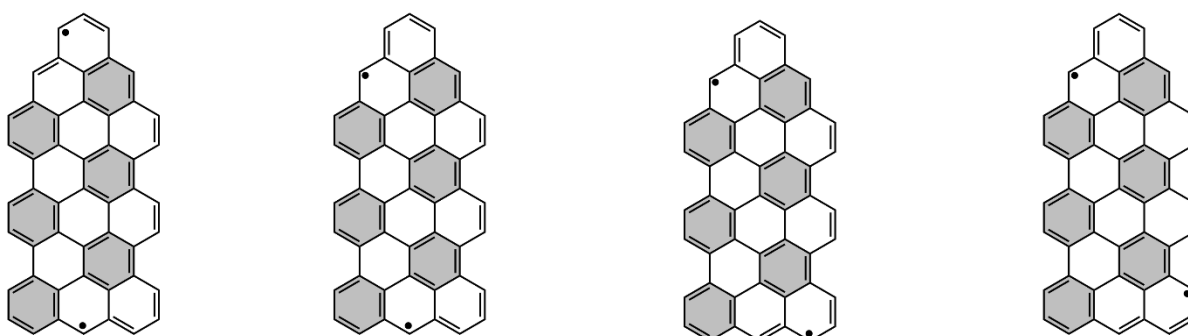
Supplementary Figure 3 | Characterizing the Kondo resonance of NG 1b. (a) The STM image of NG 1b. Tunneling parameters: $V = -200$ mV, $I = 350$ pA. Scale bar: 6 pm . (b) Low energy range dI/dV spectra of NG 1b and the signal acquisition position are marked in (a). The Kondo resonance appears when the tip position on the defect free terminus (bule circle), with

corresponding fit using the Frota function¹. Tunneling parameters: $V = -200$ mV, $I = 350$ pA, $V_{rms} = 0.4$ mV.

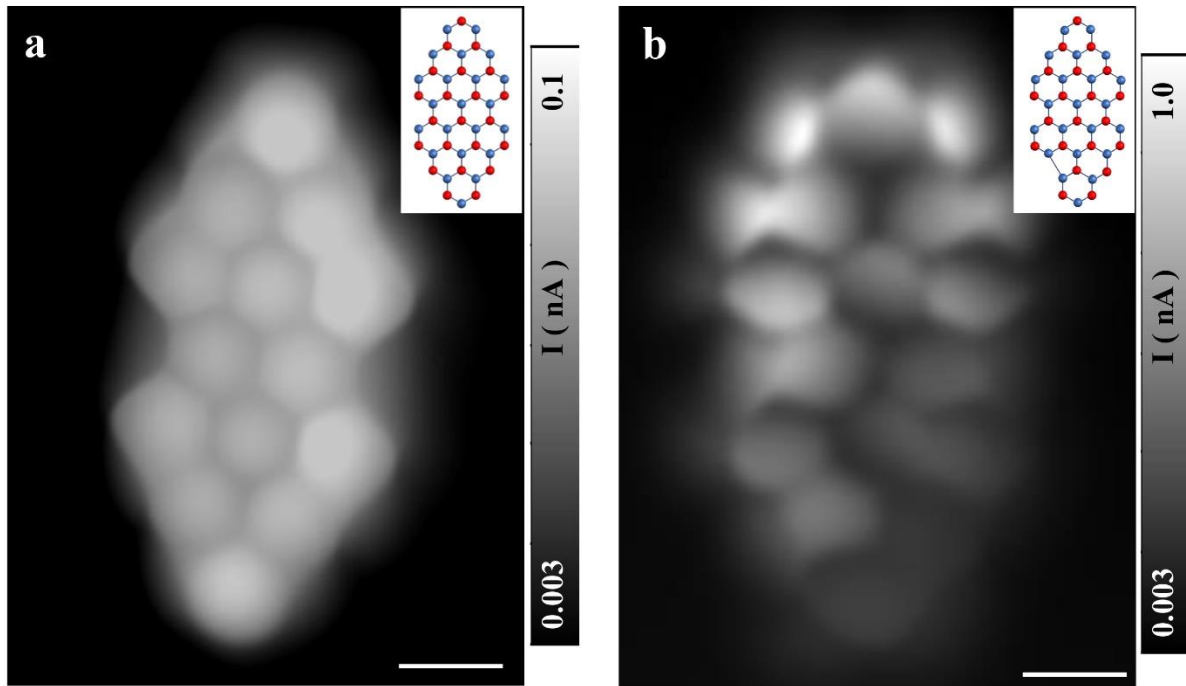


Supplementary Figure 4 | Additional dI/dV maps of NG 1b. (a) The chemical structure of NG 1b. The red and blue balls represent the carbons from two different sublattice. (b-j) dI/dV maps of a set of bias voltages. The dI/dV maps under the bias of -50 mV and $+200$ mV show the similar appearance which may indicate the frontier molecular orbital of SOMO and SUMO. Tunneling parameters: $I = 350$ pA. Scale bar: 6 \AA .

Radicals distribution of 6 Clar sextets



Supplementary Figure 5 | The radical positions of NG 2a of 6 Clar sextets.



Supplementary Figure 6 | A comparison of BR-STM images of NG 1a and 1b. (a,b) Constant height STM images of NG **1a** and **2a**. Constant height BR-STM in (a) perform as smooth feather while the enhanced states distribution at the top termini of NG **1b** which arises from Kondo resonance. Tunneling parameters: $V = +2$ mV, $z = 130$ pm (a); $V = +2$ mV, $z = 170$ pm (b). Scale bar: 3 Å.

Supplementary Table 1. A comparison of close and NG 1a and altered NG 2a

A summary of the comparison of NG **1a** and **2a**

	Chemical structure	The number C atoms in A sublattice (red)	The number C atoms in B sublattice (blue)	The total spin quantum number (S)	Spin order	Spin density distribution	BR-STM (scale bar: 4 pm)
NG 1a		22	22	0	-	-	
NG 2a		24	26	1	ferromagnetic		

1. Frota, H. O., Shape of the Kondo resonance. *Phys. Rev. B* **1992**, 45 (3), 1096-1099.