

# Room Temperature Ferromagnetism in Partially Hydrogenated Epitaxial Graphene

L.F. Xie<sup>1,2</sup>, X. Wang<sup>1,2</sup>, J. Lu<sup>3</sup>, S. Chen<sup>1</sup>, H. Huang<sup>1</sup>, Z.H. Ni<sup>4</sup>, Z.Q. Luo<sup>4</sup>, Y.Y. Wang<sup>4</sup>, T. Yu<sup>4</sup>, Z.X. Shen<sup>4</sup>, K.P. Loh<sup>3</sup>, Ariando<sup>1,2</sup>, Andrew T. S. Wee, and W. Chen<sup>\*1,3</sup>

<sup>1</sup> Department of Physics, National University of Singapore

<sup>2</sup> NanoCore, 4 Engineering Drive 3, National University of Singapore

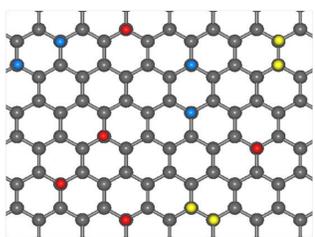
<sup>3</sup> Department of Chemistry, National University of Singapore

<sup>4</sup> School of Physical and Mathematical Sciences, Nanyang Technological University

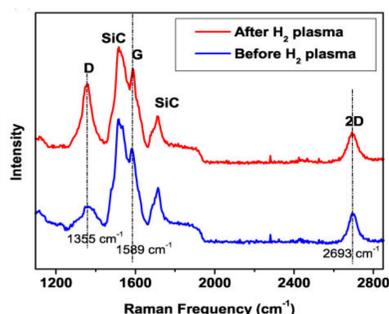
## Introduction

We report room temperature ferromagnetism in partially hydrogenated epitaxial graphene (HEG) grown on 4H-SiC(0001)<sup>1</sup>. The presence of ferromagnetism was confirmed by superconducting quantum interference devices (SQUID) measurements. Synchrotron-based near-edge x-ray absorption fine structure (NEXAFS) and high resolution electron energy loss spectroscopy (HREELS) measurements have been used to investigate the hydrogenation mechanism on the epitaxial graphene (EG) and the origin of room temperature ferromagnetism. The partial hydrogenation induces the formation of unpaired electrons in graphene, which together with the remnant delocalized  $\pi$  bonding network, can explain the observed ferromagnetism in partially HEG.

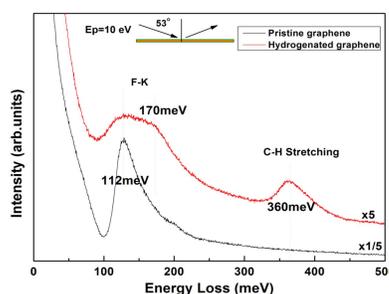
## Hydrogenation of graphene



➤ Radio frequency hydrogen plasma treatment @ RT: different configurations of H atoms are indicated in different colors, i.e., ortho-dimers in yellow, para-dimers in blue and monomers in red

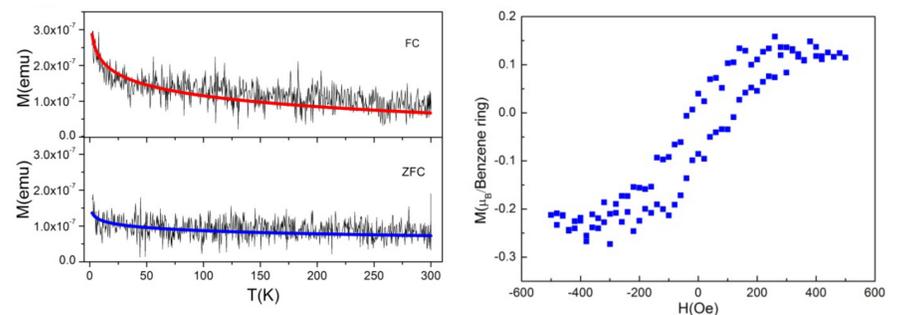


➤ Raman D peak strengthens after hydrogenation



➤ C-H  $sp^3$  stretching peak in HEG HREELS-Direct proof for H attachment on graphene

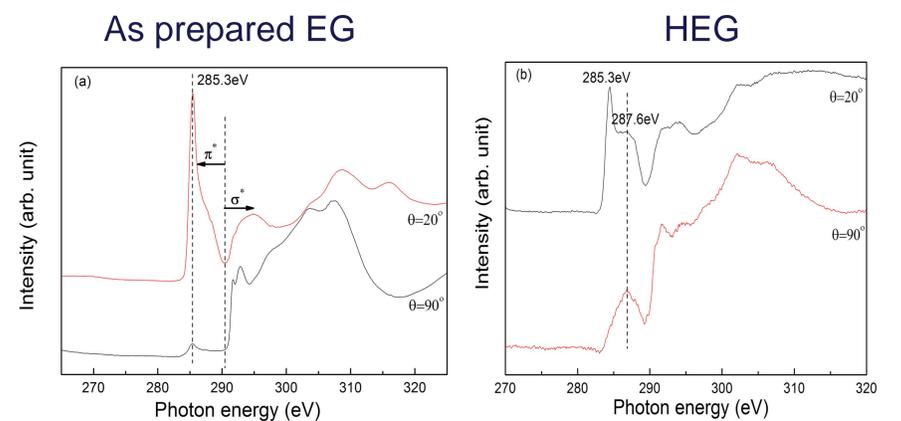
## SQUID measurements



➤ Temperature vibration of magnetization of HEG at 500 Oe showing the diverging zero field cooling (ZFC) and field cooling (FC) data

➤ Magnetic hysteresis of HEG @ 300K after subtracting the diamagnetic background

## NEXAFS investigation



➤ Remnant  $\pi^*$  band after hydrogenation supplies long range coupling between local spin units

➤ Attenuation of this  $\pi^*$  resonance peak compared with pure graphene: disrupted C=C  $sp^2$  bonding by C-H interaction

**Conclusion:** Room temperature ferromagnetism was observed in partially HEG. Hydrogenation was confirmed by HREELS experiments with the appearance of a C-H stretching peak. The mechanism of the observed ferromagnetism is explained by the formation of unpaired electrons during the hydrogenation process, together with the remnant delocalized  $\pi$  bonding network existing in the partially HEG. By this controllable hydrogenation method, we can easily turn graphene into a robust room-temperature ferromagnetic semiconductor and open up the possibility of making highly tunable graphene-based spintronic nanodevices.