Investigation of Low-Energy Ion-Implanted Multilayer Epitaxial Graphene

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There is considerable interest in integrating magnetism with graphene in the hope of creating a new class of spintronic materials. With the recent discovery of a large band-gap semiconducting form of graphene on SiC substrates [1], there are new possibilities for epitaxial graphene-based electronics and integrating magnetism could introduce a new technological dimension. One potential avenue is to make graphene magnetic using defects to generate p-orbital magnetism. Indeed, atomic hydrogen [2] attached to the graphene surface as well as vacancies [3] in graphene have been shown to induce magnetism, while high energy (MeV) proton irradiation can produce ferromagnetism at room-temperature in graphite [4]. However, detailed investigations of these systems are absent so that little is known about the density and configuration of defects, the role of interfaces, or how these relate to magnetism. Moreover, the p-orbital magnetism in these materials is unusual and of scientific interest.

In this talk, we will discuss our investigation [5] of atomic hydrogen implanted at low energies into multilayer epitaxial graphene grown on C-face SiC. The flat interface of this epitaxial system is conducive to x-ray and neutron reflectivity studies where the latter is sensitive to both the H density and magnetism. X-ray diffraction, measured *in situ* during 500 eV implantation, shows that the spacing between the graphene layers increases significantly with the ion dose. Most of the H ions remain within the sample after dosing as revealed by unpolarized neutron reflectivity. It is found from SQUID magnetometry that low-energy H implantation induces a ferromagnetic moment at room temperature. The implications of these results and others, such as ARPES and electron microscopy, will be discussed.

[1] M. Conrad, F. Wang, M. Nevius, K. Jinkins, A. Celis, M. Narayanan Nair, A. Taleb-Ibrahimi, A. Tejeda, Y. Garreau, A. Vlad, A. Coati, P. F. Miceli, and E. H. Conrad, Nano Lett. **17**, 341 (2017). M. S. Nevius, M. Conrad, F. Wang, A. Celis, M. N. Nair, A. Taleb-Ibrahimi, A. Tejeda, and E. H. Conrad, Phys. Rev. Lett. **115**, 136802 (2015).
[2] H. González-Herrero, J. M. Gómez-Rodríguez, P. Mallet, M. Moaied, J. J. Palacios, C. Salgado, M.M. Ugeda, J.-Y. Veuillen, F. Yndurain, I. Brihuega, Science **352**, 437 (2016).
[3] M. M. Ugeda, I. Brihuega, F. Guinea, and J. M. Gomez-Rodrıguez, Phys. Rev. Lett. **104**, 096804 (2010).
[4] P. Esquinazi, D. Spemann, R. Höhne, A. Setzer, K.-H. Han, T. Butz, Phys. Rev. Lett. **91**, 227201 (2003).
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Supplementary Information

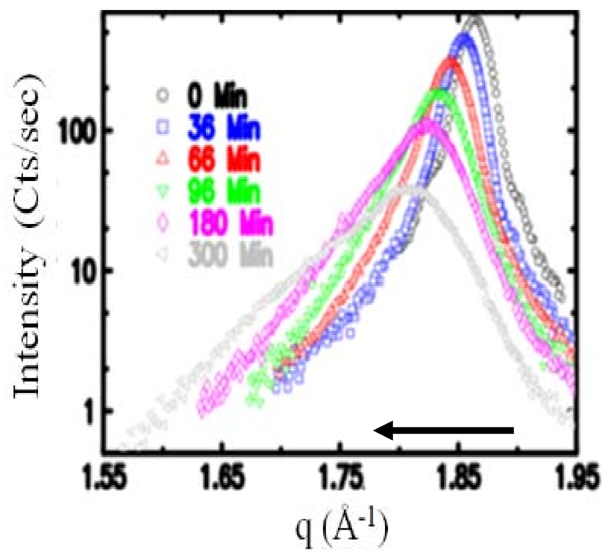

Fig. 1: X-ray diffraction measured *in situ* during 500 eV H-ion implantation of 20 layers of graphene on C-face SiC. The distance between graphene sheets is found to expand with dose, up to 5%.

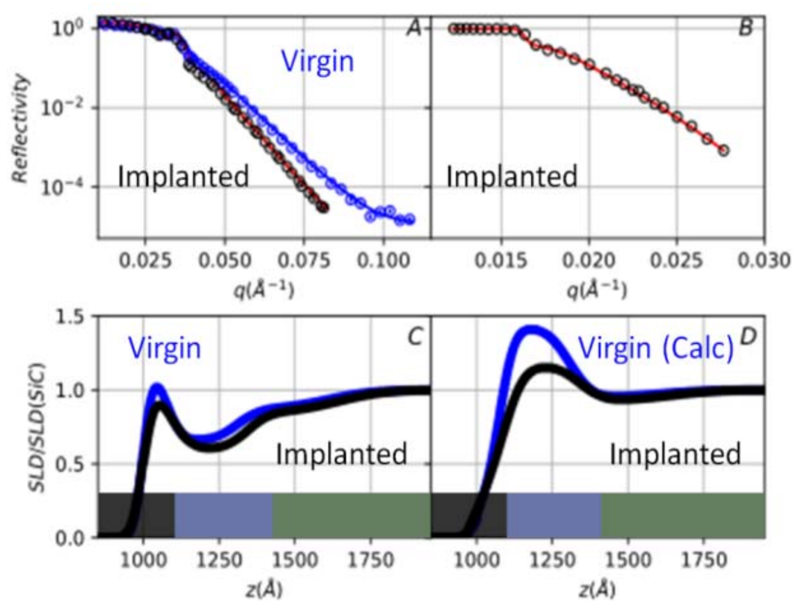

Fig. 2: Grazing angle x-ray reflectivity and neutron reflectivity measured for H-implanted epitaxial graphene on C-face SiC. (A) shows the x-ray reflectivity before and after H-implantation, while (B) shows the measured neutron reflectivity for the implanted sample. From a comparison of the scattering-length density (SLD) profiles obtained from x-rays (C) and neutrons (D), it was determined that roughly half of the implanted H remains in the sample. Note that the SLD has been normalized to the value for SiC.