Investigation of Ions and Induced Magnetism in Epitaxial Multi-layer Graphene via Polarized Neutron Reflectivity

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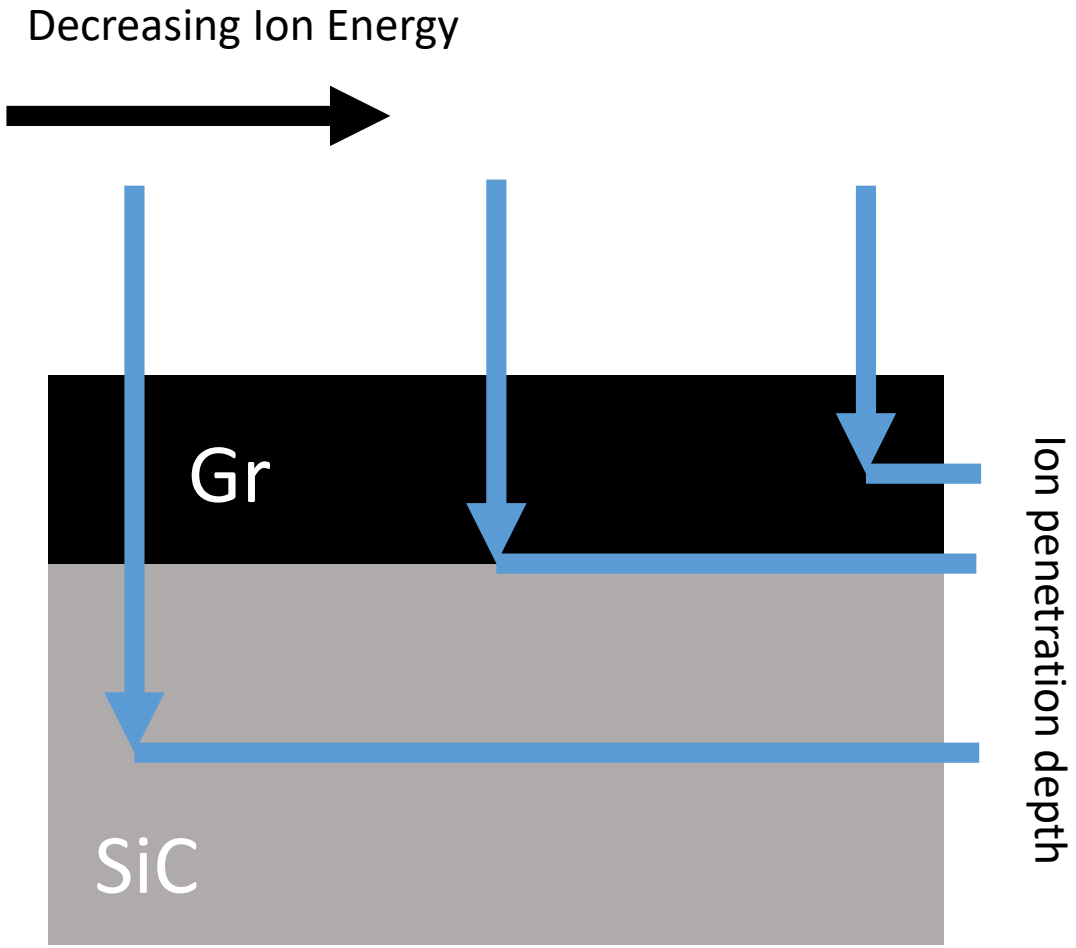
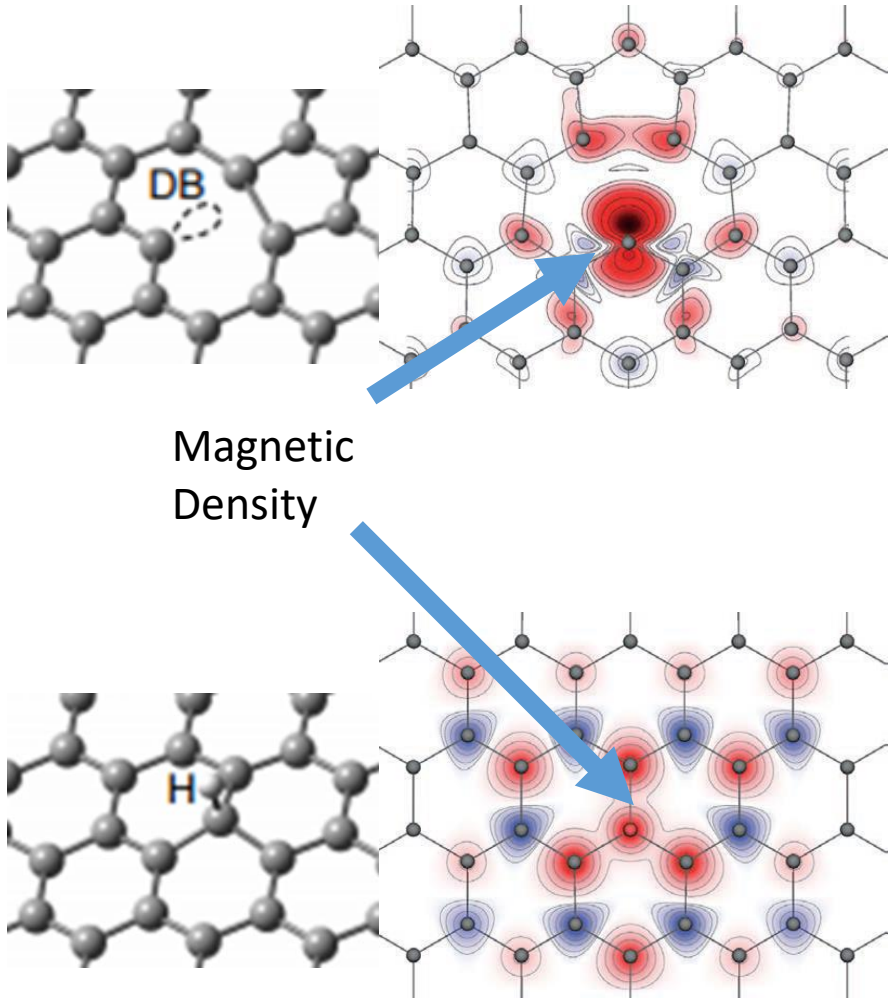
Ion Implantation

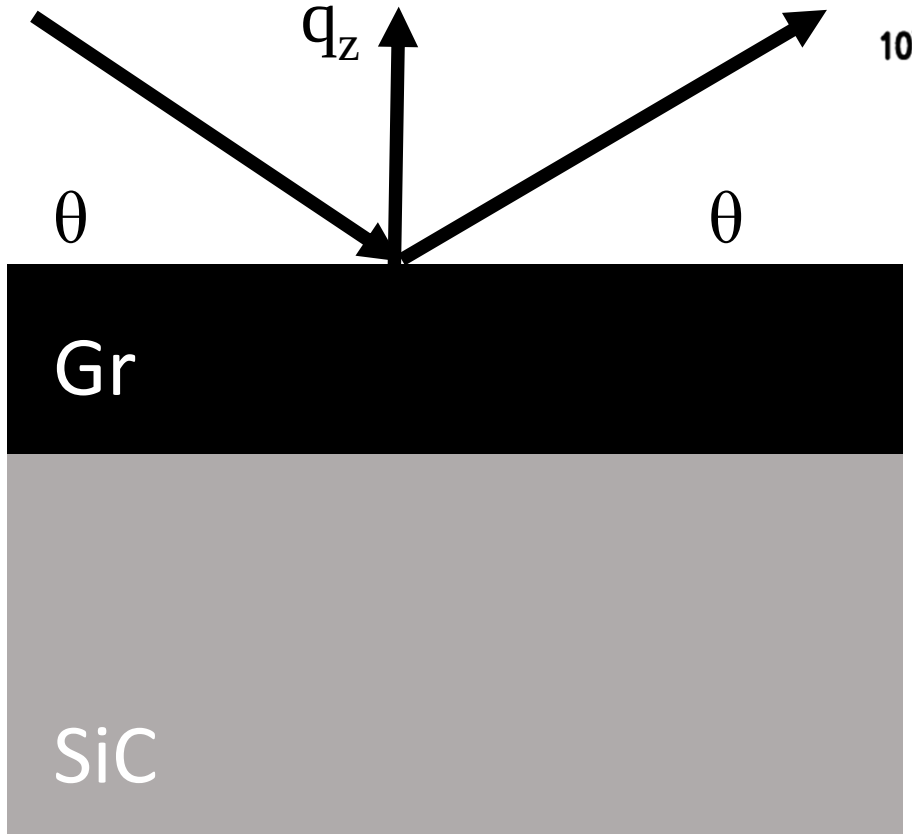


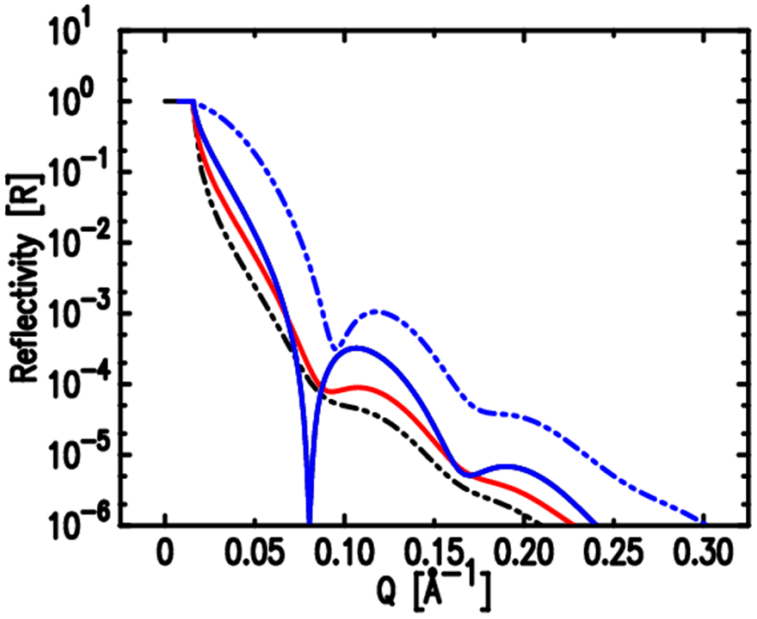
Defects lead to Magnetism

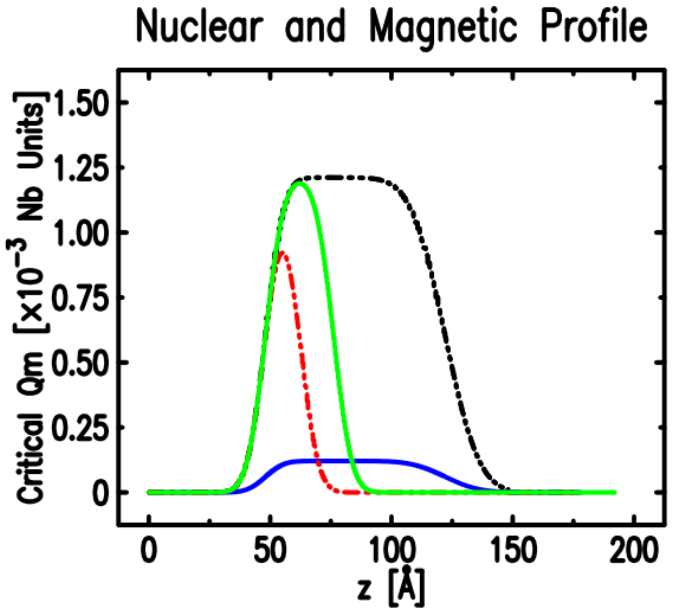


O. Yazyev and L. Helm, Phys. Rev. B 75, 125408 (2007).
W. Han, R. Kawakami, M. Gmitra and J. Fabian, Nature Nanotech 9, 794-807, (2014).

PNR to simultaneously obtain a profile for magnetism, defects







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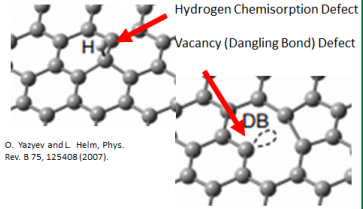
Importance of Structure in Understanding Altered Graphene

- Defects due to hydrogen can be vacancies, displacements, and chemisorbed atoms
- Lieb's theorem: materials consisting of bipartite lattices are candidates for a ferromagnetic ground state
- Defects in graphene exhibit novel p-orbital type magnetism from a purely organic material



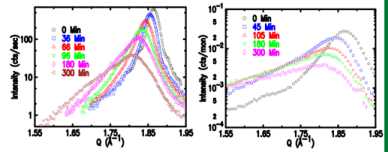
O. Yazyev and L. Helm, Phys. Rev. B 75, 125408 (2007).

What we did:

- Implanted many-layered graphene on C-face 4-H SiC with hydrogen/deuterium ions
- Used in-situ x-ray extended-range reflectivity (EXRR) and cross sectional high resolution transition election microscopy (HRTEM) to study changes in treated samples
- Studied magnetism and ion profile using polarized neutron reflectivity (PNR) and magnetometry

Extended-Range X-ray Reflectivity

- Samples studied were treated with two ion energies 2kV and 500V to vary the ion penetration depth
- Bragg shift indicating expansion of graphene layers
- Linear strain distribution observed increasing as a function of dose



Motivation – Why Alter Graphene?

- Goal: to tune the properties of graphene to have a semiconducting band gap for use in applications such as spintronic devices
- Hydrogen has been experimentally shown to produce a bandgap in graphene which is gapless in the virgin state

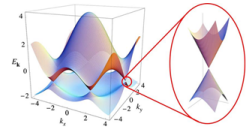
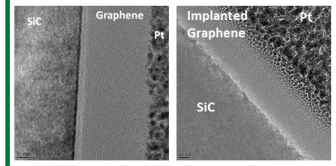


Image from: A. Castro Neto, F. Guinea, N. Peres, K. Novoselov and A. Giem, Reviews Of Modern Physics 81, 110-155, (2009).

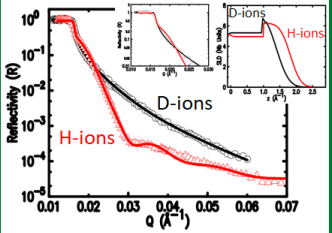
High Resolution TEM

- Prior to ion implantation the crystalline planes are clear
- After ion implantation crystalline domains are smaller, the interface is damaged and the film becomes more porous



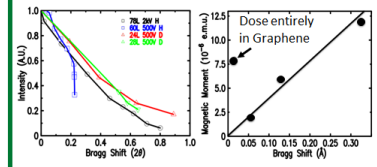
Polarized Neutron Reflectivity

- PNR measured (shown below) on BL-4A at SNS suggested sensitivity to ion type and density in graphene
- No measurable magnetic signal but have made samples which are within the limits of the instrument since

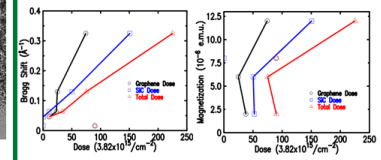


Structural Relationships to Magnetism and Ion Dose

- Shift in the Bragg from EXRR depends on the ion penetration depth
- Samples having the center of the ion distribution at the interface exhibited the largest shift while for samples with the dose primarily in graphene the shift saturated early in the treatment process



- The dose presented a more complicated story – the total dose (in graphene and SiC) scales with magnetization
- SiC has been shown to become magnetic with N defects, suggesting possible magnetic signal from the substrate
- Shift in the graphene Bragg peak interestingly scales with dose in SiC/total dose rather than dose in graphene



Drawing Conclusions

- Ion implantation leads to dramatic structural changes in graphene and in SiC
- The nature of the structural changes in the film are described by an expansion of and linear strain in the layers
- Magnetic signal scales as a function of ion dose and graphene expansion
- The nature of the magnetism relating to the profile in SiC and graphene is currently being studied at ORNL
- PNR measurement are planned for samples with increased magnetization
- An in-situ ion implantation chamber is in planning for BL-4A at the SNS

Oak Ridge National Laboratory | Spallation Neutron Source
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