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(I) Defect Engineering in Plasma-Treated Graphene Films

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Engineering of defects located in-grain or at grain boundary is central to the development of functional materials and nanomaterials. While there is a recent surge of interest in the formation, migration, and annihilation of defects during ion and plasma irradiation of bulk (3D) materials, the detailed behavior in low-dimensional materials remains most unexplored and especially difficult to assess experimentally. A new hyperspectral Raman imaging scheme providing high selectivity and diffraction-limited spatial resolution was adapted to examine plasma-induced damage in a polycrystalline graphene film grown by chemical vapor deposition on copper substrates and then transferred on silicon substrates. For experiments realized in nominally pure argon plasmas at low pressure, spatially resolved Raman conducted before and after each plasma treatment shows that the defect generation in graphene films exposed to very low-energy (11 eV) ion bombardment follows a 0D defect curve, while the domain boundaries tend to develop as 1D defects. Surprisingly and contrary to common expectations of plasma-surface interactions, damage generation at grain boundaries is slower than within the grains. Inspired by recent modeling studies, this behavior can be ascribed to a lattice reconstruction mechanism occurring preferentially at domain boundaries and induced by preferential atom migration and adatom-vacancy recombination. Further studies were realized to compare the impact of different plasma environments promoting either positive argon ions, metastable argon species, or VUV-photons on the damage formation dynamics. While most of the defect formation is due to knock-on collisions by 11-eV argon ions, the combination with VUV-photon or metastable atom irradiation is found to have a very different impact. In the former, the photons are mainly thought to clean the films from PMMA residues due to graphene transfer from copper to silicon substrates. On the other hand, the surface de-excitation of metastable species first impedes the defect generation and then promotes it for higher lattice disorder. While this impediment can be linked to an enhanced defect migration and self-healing at nanocrystallite boundaries in graphene, such effect vanishes in more heavily-damaged films. Finally, these experiments were used as building blocks to examine the formation of chemically doped graphene film in such plasmas using argon mixed with either traces of N- or B-bearing gases.

Primary author: STAFFORD, Luc (Universite de Montreal)

Presenter: STAFFORD, Luc (Universite de Montreal)

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