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Exploring new regimes of plasma-graphene interactions by space- and time-resolved diagnostics

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Incorporation of foreign atoms in low-dimensional materials such as graphene are interesting for many applications, including biosensing, super-capacitors, and electronic device fabrication. In such processes, controlling the nature of the foreign atom incorporation is a key challenge, as different moieties can contribute differently to doping and present different reactivities. With plasma processing increasingly requiring atomic level precision, a detailed understanding of the mechanisms by which ions, electrons, reactive neutrals, excited species, and photons interact simultaneously with materials such as graphene has become more important than ever.

In recent years, we studied the interaction of low-pressure argon plasmas with polycrystalline graphene films grown by chemical vapor deposition. Spatially-resolved Raman spectroscopy conducted before and after each plasma treatment showed defect generation following a 0D defect curve, while the domain boundaries developed as 1D defects. Surprisingly and contrary to common expectations of plasma-surface interactions, damage generation was slower at the grain boundaries than within the graphene grains, a behavior ascribed to a new preferential self-healing mechanism. Through a judicious control of the properties of the flowing afterglow of a microwave N2 plasma obtained by space- resolved optical emission spectroscopy, we further demonstrated an aromatic incorporation of nitrogen groups in graphene with minimal ion-induced damage. The use of both reactive neutral atoms and N2 excited states (mostly metastable states) was a radical departure from what was the state of the art in atomic manipulation, mainly because excited species can provide sufficient energy for the activation of adatom covalent incorporation while leaving the translational energy of both the impinging species and the low-dimensional materials undisturbed. A selective nitrogen doping due to preferential healing of plasma-generated defects near grain boundaries was also highlighted.

Very recently, a new setup was specifically designed to examine plasma-graphene interactions. In-plasma Raman spectrometry is used to monitor the evolution of selected Raman peaks over nine points of the graphene surface. On one hand, for high-energy ions, defect generation progressively rises with the ion dose, with no significant variations after ion irradiation. On the other hand, for very-low-energy ions, defect generation increases at a lower rate and then decreases over a very long time scale after ion irradiation. Such self-healing dynamics cannot be explained by a simple carbon adatom-vacancy annihilation. Using a 0D model, it is demonstrated that that various mechanisms are in play, including carbon adatom trapping by Stone-Wales defects and dimerization. These mechanisms compete with the self-healing of graphene at room temperature, and they slow down the healing process. Such features are not observed at higher energies for which carbon atoms are sputtered from the graphene surface, with no significant populations of carbon adatoms. We believe that these experiments can be used as building blocks to examine the formation of chemically doped graphene film in reactive plasmas using, for example, argon mixed with either traces of N- or B-bearing gases.

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Keyword-2

Plasma-surface interactions

Keyword-3

Low-dimensional materials

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