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Graphene and doped graphene from adsorbed molecules

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Summary

Graphene, the first 2D material to be recognized as such, possesses very special mechanical, electronic and thermal properties that surpass those of 3D solids, especially when multiple functions are required. Graphene can shape the future world if it can be understood completely and implemented carefully in relevant fields. As different applications have different requirements concerning the crystalline quality of the material, there have been extensive attempts during last decade to grow graphene via chemical exfoliation of graphite, by reduction of graphite oxide, by thermal graphitization of silicon carbide or by CVD on metallic substrates. For electronic device applications, a real challenge till date is to produce graphene with a low number of defects. Hence there is still room to search for new methods to grow graphene. Starting out from self-assembled monolayers is a promising route that has not received full attention yet. This approach is simple, easily upscalable and can be employed to produce doped graphene as well.

In this study our main goal was to synthesize large area graphene/doped graphene sheets from adsorbed molecules. One objective was to explore whether by this growth method graphene of equivalent or even better quality than CVD-grown graphene can be produced at temperatures lower than those required for CVD. A second objective was to inspect if the same

method can yield graphene on insulating substrates, and the third objective was to clarify if the doped graphene is achievable by this molecular route.

In Chapter 3 we demonstrated the molecular route for synthesizing graphene from self-assembled monolayers (SAMs) of 1, 1′-biphenyl-4-thiol on electropolished and oxidized copper substrates. The SAMs were first polymerized by light and then annealed in a vacuum furnace to generate graphene. The quality of the grown material was verified with a variety of techniques including contact angle, X-ray photoelectron and Raman spectroscopies, scanning electron and transmission electron microscopy. The graphene growth on oxidized copper foils yielded better quality than that on electropolished copper substrates. We also found that on electropolished Cu foil the quality of the obtained graphene strongly depends on the crystallographic orientation of the grains. With respect to our objectives, we identified an easy method for growing graphene on an insulating substrate but the temperatures necessary to obtain a quality comparable to CVD graphene was not lower than that necessary in CVD.

In Chapter 4 we presented the synthesis and characterization of B- and N-doped graphene from self-assembled monolayers (SAMs) of thiol borazine ($C_{78}H_{69}B_3N_{12}S_3$) on electropolished polycrystalline copper foils polymerized by UV-light and then annealed in a vacuum furnace to transform them into doped graphene. In particular we determined the ratio of hetero- to carbon atoms that can be obtained in the final product via this route. A multi technique characterization was employed to investigate the quality of the doped graphene. The resulting graphene was multilayer but the presence of doping atoms is an encouraging result that nearly satisfies our third objective. Future studies will have to find a way to lower the amount of carbon in the initial layer of adsorbed molecules, either by starting with mixed SAMs where thiol borazine is co-assembled with small molecules or by choosing a smaller molecule with heteroatoms as building block for the SAM.

Chapter 5 illustrated the growth of graphene on Cu(111) starting from the thermal decomposition of chemisorbed C_{60} molecules. Low energy electron diffraction, Raman spectroscopy and X-ray photoelectron spectroscopy were used to characterize the various intermediate products and the obtained graphene. As the temperature required to grow graphene from decomposition of C_{60} was still very high, the main aim to grow graphene at lower temperatures (less energy costly) than required for CVD growth still needs more work. The grown graphene was multidomain, with a preference for alignment along the substrate orientation, and Raman spectra showed that it is a single layer but with a high defect density. It might be worthwhile to control whether a sub-monolayer coverage could give a better result and to try to grow graphene *in situ* at higher annealing rates.

Finally in Chapter 6 we explored an alternative synthesis route of doped graphene, *i.e.* doped graphene from thermal decomposition of adsorbed $C_{59}N$ on Cu(111). Low energy electron diffraction showed that graphene grown in this fashion is multidomain, but growth along the substrate orientation is preferred. However the N concentration in the product was below the XPS detection limit in our experiments. Further experiments are planned to verify whether doped graphene can be produced this way and determine the dopant concentration.

Given our results detailed in Chapter 3 and 4, we may speculate that the synthesis from adsorbed molecules may also be helpful for the growth of doped graphene directly on insulating substrates. In fact, a precursor containing both carbon as well as nitrogen and/or boron allows to reduce the number of steps and the time to grow doped graphene with respect to currently employed methods.