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Applications of graphene in nanotechnology : 1D diffusion, current drag and nanoelectrodes

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SUMMARY

The initial motivation and the main aim of our research was to investigate electromigration at the atomic scale (**Chapter 2**). Knowledge gained from such experiments would give clues to many unanswered questions about current-induced forces. Graphene on an insulating substrate was the first system to be investigated since the current would be confined within an atomic layer. Electromigration would be observable by the current-induced motion of individual adatoms deposited on the surface, which can be imaged by scanning tunneling microscopy (STM). We used graphene on Si/SiO₂ and that comes with a few limitations that prevented us from observing migration of adatoms with the current densities achievable in our STM. Graphene nanoribbons (GNR) proved to be a good alternative to graphene, although the underlying substrate is conducting and current is not confined within the GNR. We used armchair graphene nanoribbons grown on Au(111), and on top of this substrate we evaporated cobalt atoms. Using an STM tip to send a voltage pulse while in contact with a GNR, we induced one-dimensional motion of Co adatoms on the GNR. We did not observe directional motion of the adatoms which would indicate the presence of current-induced forces, most likely because the current density along the GNR was too small. These results suggest that migration of Co on GNR mainly comes from thermal activation. Temperature-dependent diffusion measurements gave us information on the relative energy barriers for Co adatoms on gold and on GNRs. We expect that current-induced motion can be observed and quantified by this method if most of the GNR's length can be separated from the substrate by an insulating layer.

The study of electromigration is also the subject of the second part of the research presented in this thesis (**Chapter 3**). The recent observation⁵ that dragging a droplet along a strip of graphene induces a voltage across the ends of this strip, inspired us to ask the following question: would it be possible to do the opposite and induce motion of the droplet by applying a voltage across the graphene strip? Our experiments have shown that it is, in fact, not possible to do this. In order to understand the role of the adsorption of ions, formation of an electric double layer and the formation of a pseudocapacitor, we investigated several different ionic liquids. Results for an aqueous solution of NaCl showed that the voltage signal is linearly dependent on the speed of the droplet, and that the sign of the voltage depends on the direction of the droplet motion. We also observed that the voltage signal, induced by moving a droplet of 0.01 M concentration, is larger than the signal for an ion concentration of

⁵ Yin *et al.*, Nature Nanotechnology, **9**, 378 (2014)

0.6 M. Furthermore, we utilized ionic liquids which we assumed will bond strongly to graphene and the underlying polyethylene terephthalate (PET) substrate: sodium benzenesulfonate and benzenesulfonic acid. This led to an interesting discovery: a droplet of distilled water, when moved along the graphene surface after having used these ionic liquids, also induces a voltage. This was an indication that certain types of ions adhere so strongly to graphene that they leave the droplet and form a charged layer on graphene. We concluded that the polarizability of water has a contribution to the voltage signal almost as large as ionic liquids, when moved over such a charged graphene surface.

As it often happens when doing scientific research, looking into certain problems sometimes leads to gaining knowledge (and interest) in concepts that are far from the initial objective. After doing many experiments with graphene, we were motivated to take it a step further and attempt to design atomically thin nanoelectrodes by means of a reproducible and reliable fabrication method. This project, described in **Chapter 4**, was particularly attractive, having a prospect for applications such as electron-transport measurements on single molecules and sequencing of biopolymers. We present a few possible methods for creating graphene electrodes on Si/SiO₂: the *broken-graphene* and the *etched-graphene* method. While the broken graphene method did not yield usable graphene nanoelectrodes with full graphene coverage up to the edge of the substrate, the etched graphene approach was very successful. All techniques used to characterize etched graphene edges (Raman spectroscopy, SEM, AFM and electrical measurements) unanimously confirmed that graphene reaches up to the edge within respective resolutions of each of the techniques. Using a modified STM and a twisted-sample approach, we have created the first tunable tunneling nanogap between graphene-edge electrodes. By pushing the electrodes closer together, we have been able to form an atomic sized junction between the two edges of graphene.