Direct Patterning and Imaging of Graphene Structures with a Helium Ion Microscope

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The discovery of graphene in 2004[1] has triggered enormous experimental and theoretical efforts. As a gapless semiconductor, charge carriers in graphene can be tuned continuously from electrons to holes crossing the charge neutral Dirac point using an external electric field. Many exciting device concepts have been proposed when graphene is patterned into narrow ribbons[2]. Lithographically patterned graphene ribbons in the sub-100 nm range allow the engineering of energy band gaps, and such structures have been fabricated and characterized previously[3,4]. The size of these energy gaps depends inversely on the ribbon width. However, standard e-beam lithography gives only access to the 10 nm and above size range, and then only with specialized processing. To fully explore the potential of graphene nanoribbons for device applications with novel functionalities, the fabrication of ribbons in the sub-10 nm size range is necessary. Among the many predictions for such ribbons perhaps the most exciting are half-metallicity and ferromagnetism (provided that the edges are prepared such that they have well defined chirality). Equally important is to reach this size range by means of resist free patterning. In the context of graphene, one of the main challenges in resist based patterning is residual resist, Figure 1. On the sub-10 nm scale, even the smallest amount of residue will be detrimental on electronic transport properties of graphene nanoribbons and is likely to mask their fascinating intrinsic properties. We report a new method based on focused He+ ions which enables resist-free, direct patterning of graphene structures with sub-10 nm resolution.

Recent technological breakthroughs with the Gas Field Ion Source (GFIS), have enabled researchers to push imaging technology into the deep sub-nanometer regime with focused helium ion beams[5]. The Helium Ion Microscope (HIM), able to resolve nanoscale features on solid samples with an edge resolution of a mere 0.25nm[6], has a number of attributes which make it attractive for the imaging of graphene structures. For example, in addition to the high spatial resolution, the shallow escape depth (~1 nm) of the He+ excited secondary electrons provides images with tremendous surface contrast, revealing monolayer sheets of graphene and step variations on a variety of substrates, Figure 2. Even more compelling is the ability to directly modify graphene, through surface sputtering, enabling direct pattern transfer for the fabrication of graphene devices, Figure 3. This ability to directly pattern graphene structures promises great utility in the fabrication of sub-10 nm graphene devices with minimal contamination on conventional substrates. It also provides a mechanism for high resolution patterning on non-conventional substrates (such as suspended graphene membranes), where resist based lithographic techniques are not feasible. We have observed sub-10 nm pattern transfer on both supported (Si bulk, 300 nm $Si0_2$) and suspended structures, with graphene nano-ribbons of 7 nm width and individual holes of 5 nm being demonstrated.

Nevertheless, this technique is not without issues. Under typical conditions, the balance between ion beam induced deposition from residual hydrocarbons and surface sputtering weighs in favor of deposition. Techniques to eliminate the hydrocarbons prior to beam exposure are necessary to enable surface sputtering at the fluences required for the finest features. Additionally, the spatial dimensions of our finest patterns are still substantially larger than the expected beam probe size (<1 nm). The sources of this discrepancy are currently under investigation and we are exploring the fundamental limits of direct patterning with 30 keV helium ion beams. Finally, possible damage to the graphene bonds and its influence on edge effects is a concern. We are investigating the invasiveness of this process on graphene properties and ways to mitigate potential detrimental effects.

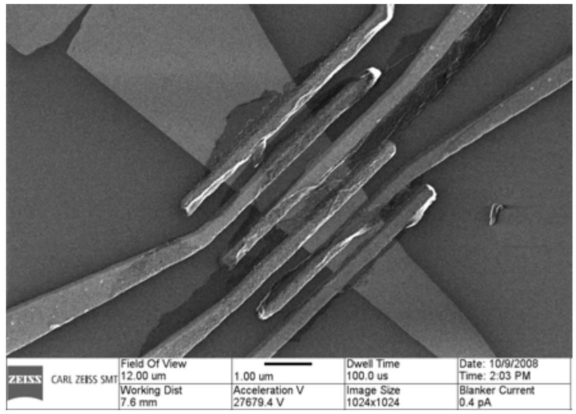

Figure 1 Residual PMMA on a patterned graphene device.

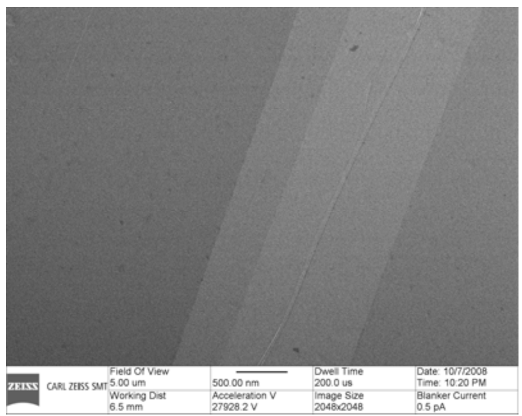

Figure 2 Imaging of graphene: A monolayer graphene sheet with a Z-fold on a Si substrate with 300 nm of $Si0_2$. Contrast between the single monolayer and the thicker layers of the fold is clearly evident.

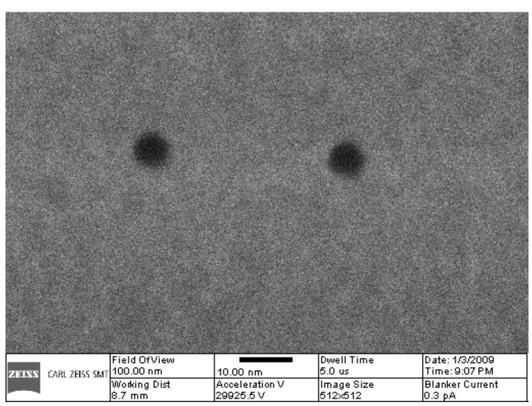

Figure 3 Direct sub-10 nm patterning of Graphene. The holes shown here, between 6-7 nm in diameter, were He+ milled in suspended graphene sheets a few monolayers thick. Smaller holes have been observed (<5 nm), but imaging at high magnification with sufficient signal to noise broadens the features.

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