## Atomic force microscope nanolithography of graphene: cuts, pseudo cuts and tip current measurements.

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Scanning probe microscopy (SPM), not only a powerful tool for imaging is also an established method for the manipulation and patterning of materials on the nanometre scale[1]. The atomic force microscope (AFM), part of the SPM family, now routinely used as a lithographic tool for Si and GaAs nano-scale device fabrication[2,3], shows significant potential for device fabrication in graphene[4], a material of great current interest due to its exceptional mechanical and electronic properties[5].

AFM lithography (AFML) on graphene, performed under ambient conditions, utilises the dissociation of water molecules, due to a tip induced electric field, and the subsequent oxidation of the graphene substrate to fabricate desired nano-structures. Most commonly electron beam lithography and subsequent plasma etching is used for the fabrication of nano-scale graphene devices[6,7] however AFML offers several advantages over this technique: it has nm scale resolution, can be performed under ambient conditions, eliminates the need for e-beam resists and associated contamination, allows in situ device measurement during lithography and allows further device modifications to be carried out easily and at any time. Many of the key parameters required for successful AFML are not yet well established and device fabrication is not yet routine. In addition there has as yet there not been any systematic study of the tip current during AFML of graphene.

In this work, we investigate in detail the cutting of the graphene lattice with an AFM tip. In particular, we measure the tip current,  $I_{tip}$ , during the cutting process. From these measurements we identify two distinct regimes (see Fig 1): finite and zero tip current lithography. We find that we cut graphene only when  $I_{tip}$ , drops to zero (within our noise floor) and that pseudo cuts appear when  $I_{tip}$ , is non-zero. These pseudo cuts, in which the electron system of graphene remains uninterrupted, cannot be distinguished from real cuts by AFM height imaging but become apparent using transport experiments and scanning electron microscopy (see Fig 2). Our results provide new insight into the oxidation mechanism of graphene and identify the parameter range for true cuts which is crucial for device fabrication.

Finally, we demonstrate device fabrication. Fig.3 (a) & (c) show AFM images of a quantum wire and quantum dot (QD), respectively, formed in a bi-layer flake by AFML. Fig.3 (b) shows conductance *vs* back gate voltage  $V_{bg}$  of the quantum wire at 4.2 K. The flexibility of AFML is illustrated by Fig.3 (d) which shows measurements of the QD conductance *vs*  $V_{bg}$  at 4.2 K both for the quantum dot as shown in Fig.3 (c) (blue line) as well as that of the same device but with the entrance barriers of the quantum dot narrowed from ~ 150 nm to ~ 50 nm in a subsequent AFML step shown by the blue arrows in Fig.3 (c). As expected the conductance is significantly lower in the post modification device with an increase in the gap observed[8]

In conclusion, we have studied the local oxidation of graphene by an AFM tip. We demonstrate that at low bias voltages the graphene is typically not cut even though clear depressions are observed in AFM height images. Our work demonstrates that a high tip current is not necessary for local oxidation of the graphene lattice. Only when the tip current vanishes is the graphene lattice cut. These conclusions are supported by scanning electron microscopy and transport experiments. The ability to distinguish between pseudo cuts and cuts as demonstrated here is important for graphene device fabrication by AFM nanolithography.

## References

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## Figures



**Figure 1.** A series of cuts in a monolayer graphene flake, made using an AFM tip at increasingly negative voltages relative to the grounded flake. Upper panels show averaged height profile cross-sections of the cuts, central panels show the corresponding AFM micrographs and the bottom panels show the current,  $I_{tip}$ , through the AFM tip as a function of time t, recorded during the cutting process where t = 0 is the start of tip contact. The series is performed using a single, non coated, doped silicon sensor tip.



Figure 2. (a) & (b) show AFM (left) and SEM images of two triangles cut with  $V_{tip} < V_{thresh}$  such that the (right) current, I<sub>tip</sub>, through the tip during cutting is approximately zero. In the topographic AFM images the central regions are clearly visible, however in the SEM images the central regions are no longer seen, these regions must be up in the electron beam and must therefore be charging isolating. (c) & (d) show AFM (left) and SEM electrically images (right) of two triangles cut with V<sub>tip</sub> > V<sub>thresh</sub> such that the current,  $I_{tip}$ , t through the tip during cutting is ~ 100 uA. The AFM images are qualitatively similar to those in (a) & (b) however no contrast is seen in the SEM images and the central region in this case remain electrically connected to the rest of the flake. (e) & (f) show SEM images of the areas of the flake on which the triangles were indicate the locations of the cut with blue arrows to triangles shown in (a)-(d).



(a) AFM image of a ~ 65nm constriction Figure 3. bilayer flake using AFML and (c), its formed in а conductance, G, vs V<sub>bg</sub> at 4.2 K. (c) QD formed in a bilayer the entrances are initially ~ 150 nm, flake using AFML, as indicated by the blue arrows. these are then modified. using AFML to be  $\sim$ 50 nm. (d) G vs  $V_{bg}$  at 4.2 K of the dot pre-modification (blue curve) and post-modification (black curve).