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Sub-10 nm patterning by focused He-ion beam milling for fabrication of downscaled graphene nano devices

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ABSTRACT

In this work, a novel hybrid fabrication method for graphene quantum dot devices with minimum feature sizes of ~3 nm and high yield is described. It is a combination of e-beam lithography and direct milling with the sub-nm focused helium ion beam generated by a helium ion microscope. The method is used to fabricate graphene quantum dot devices contacted with metal to allow electrical characterization. An annealing step is described that reduces hydrocarbon contamination on the sample surface and allows complete removal of graphene by the helium ion beam and therefore successful isolation of side gates. The electrical characterization of the final device demonstrates the successful fabrication of the first electrically characterized He-ion beam patterned graphene device. The highly controllable, fine scale fabrication capabilities offered by this approach could lead to a more detailed understanding of the electrical characteristics of graphene quantum devices and pave the way towards room-temperature operable graphene quantum dot devices.

1. Introduction

Carbon-based materials play a major role in present science and technology. Graphene, a two-dimensional (2D) single atomic layer of graphite, is a recent discovery in the continuous advances in the science of carbon. Despite its short history, its unique material features have ensured a rapid growth of interest in several areas of science and technology. Graphene is considered an excellent candidate for fabricating high mobility nanoscale electronic devices due to its remarkable electron-transport characteristics that include e^2/h minimum conductivity even at nominally zero densities [1] and the highest known intrinsic mobility, in excess of 200,000 $cm^2\ V^{-1}\ s^{-1}$ at low temperatures [2]. In addition, spin–orbit coupling and hyperfine interaction with carbon nuclei are both small in graphene, and a very long spin relaxation length has been demonstrated [3,4], which make graphene a promising candidate material for Quantum Information Technologies (QIT) and spin qubit embodiment [5]. These superior transport properties encourage the downscaling of graphene devices further to the regime where the coherent nature of electronic and spin states can be fully exploited. However, this requires the development of ultrafine patterning technologies which enables accurate nanoscale fabrication beyond the present electron-beam lithography technique.

Single Electron Transistors (SETs) with Double Quantum Dot (DQD) are considered as promising candidates for building blocks of future quantum computer circuits [6] and beyond CMOS technology. Recently, a few studies have demonstrated the behavior of Quantum Dot (QD) devices in monolayer graphene [7–10] and bi-layer graphene [11] flakes.

Currently, e-beam lithography followed by reactive ion etching (RIE) is the most established method for fabricating graphene devices. However, e-beam spot size, proximity effect and undercutting of the resist layer during etching can limit the resolution of this technique. Furthermore, uneven thickness of the resist layer, which may be caused by the roughness of the SiO_2 surface layer and the presence of graphite pieces and metallic alignment marks, can also limit resolution and affect the symmetry of the patterns fabricated by this method [9–12]. Fig. 1 shows a Graphene DQD (GDQD) device, fabricated by e-beam lithography and RIE. The asymmetry in the size of the fabricated features, i.e. DQD, and width-variations along the channel, are clearly evident.

Helium ion microscopy (HIM) is a new surface imaging technique that involves scanning a focused beam of helium ions across a surface to generate an image from the resulting secondary electron (SE) emission. An atomically sharp and extremely bright source, combined with the larger momentum (and so smaller de Broglie wavelength) of helium ions compared to electrons, enables a sub-nanometer probe size at the sample surface and high resolution imaging, below 0.35 nm in some systems [13–15]. Direct

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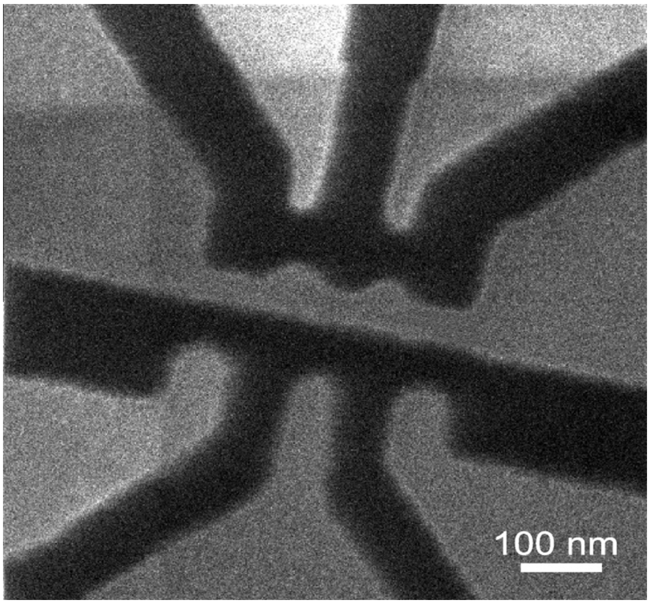

Fig. 1. HIM SE image of GDQD fabricated by e-beam lithography and RIE etch. The image clearly shows the asymmetry in the size of the fabricated features.

milling of graphene using a focused beam of helium ions generated in a helium ion microscope (HIM) has been demonstrated recently [13,16–18] as a high resolution, resist-free patterning technique. This includes HIM milling on a suspended graphene flake to form a 5 nm wide nano ribbon with a 60:1 aspect ratio [18] and the patterning of isolated graphene flakes on SiO_2/Si substrates with a variety of QD device designs, with minimum feature sizes of <10 nm [16]. Furthermore, the cutting of contacted, suspended graphene flakes with HIM has been demonstrated, with electrical measurements before and after confirming the complete milling through the flake, with a minimum feature size of 10 nm [17]. A contacted graphene flake on an SiO_2/Si substrate has also been electrically characterized *in situ* during HIM milling. Here, the residual current through the flake saturated at ~4 nA (from an original, pre-milling value of above 0.9 μA), which was attributed to hydrocarbon contamination on the SiO_2 substrate in the milled area [17].

In this work, we report a novel, hybrid fabrication process that combines conventional e-beam lithography and HIM milling to fabricate high resolution graphene DQD devices, reliably and reproducibly. E-beam lithography is used to form metal contacts onto graphene flakes. Helium ion milling is then used to pattern the flakes with intricate DQD devices, with sub-10 nm resolution and high fidelity, and to isolate the gate electrodes from the source and drain regions of the devices, enabling the fabricated nanoscale devices to be electrically characterized. To obtain the desired high resolution HIM milling, an annealing step is included between the two patterning steps to remove the contamination produced during the e-beam lithography steps on the surface of the graphene flakes. The electrical characterization of the final device demonstrates the successful fabrication of the first electrically characterized He-ion beam patterned graphene device. This approach could pave the way to a better understanding and more detailed study of graphene quantum devices.

2. Fabrication process

The processing steps for the fabrication of GDQD by HIM milling are illustrated in Fig. 2.

2.1. Graphene flake preparation

Before deposition of graphene, the substrates were patterned with reference alignment marks and bond pads by two e-beam lithography steps followed by the e-beam evaporation of Ti/Au (5 nm/45 nm for the first step and 15 nm/300 nm for the second step) and the subsequent lift-off. Graphene flakes were produced by mechanical exfoliation of Kish graphite and were transferred onto highly doped Si substrates with a 295 nm-thick SiO_2 top layer, as described by Novoselov et al. [19]. Monolayer flakes were identified by optical microscopy and Raman spectroscopy.

2.2. E-beam processing

The first e-beam lithography stage was employed to fabricate metal contacts onto the deposited flakes. The fine helium ion beam milled features would be damaged during resist coating and lift-off processes and so the e-beam processing had to be carried out prior to HIM milling. This also enabled more accurate alignment of the HIM and e-beam defined patterns than would have been possible if e-beam processing was carried out after HIM milling. E-beam lithography was performed using a JEOL 9300FS e-beam lithography system. To minimize e-beam induced defects in the graphene flakes [20], the samples were spin-coated with a ~460 nm-thick layer of *Methyl Methacrylate* (MMA) 8.5 resist. The high sensitivity of MMA resist allows the use of a low e-beam base dose (110 μC/cm^2), with the e-beam operating at an acceleration voltage of 100 kV. MMA resist is often used with a *Poly Methyl Methacrylate* (PMMA) resist layer to construct a bi-layer resist layer and ensure a successful lift-off process. However, by proper tuning of the e-beam lithography exposure and development conditions, i.e. baking conditions of the resist (90 s at 150 °C), developer mixture (1:1 mixture of MIBK:IPA) and the resist development time (75 s), a single MMA resist layer on its own can result in a successful lift-off. The e-beam exposed patterns were then metalized by electron beam evaporation of Ti/Au (5 nm/55 nm) at a pressure of 3×10^{-7} mbar in a Leybold LAB700 evaporator. The subsequent lift-off was performed in *N-Methyl-2-pyrrolidone* (NMP) at 50 °C followed by a rinse in IPA. Fig. 3 shows an optical image demonstrating the successful fabrication of the metal contacts onto a graphene flake using the single resist layer and the lift-off process.

For a working GQD device, in addition to patterning flakes with the desired design, the metal gate contacts need to be fully isolated from each other and the source and drain contacts. It is desirable to minimize total HIM milling time because drifting of the sample during the milling can jeopardize pattern fidelity for long exposure times. This was observable in previous work [16] by our group, where the milled QD patterns experienced noticeable distortions due to drift. Therefore, to minimize the milling time, a second e-beam lithography step was employed to introduce isolation lines on the flakes, separating the metal contacts and leaving an area of approximately 500 nm × 420 nm for the final HIM milling step. For the second lithography step, the samples were spin-coated with a ~40 nm-thick layer of PMMA 495K resist. The isolation patterns were exposed by a constant e-beam dose of 190 μC/cm^2. The exposed patterns were then transferred into the flakes by RIE etch in an Ar/O_2 (4:1) atmosphere using a RF power of 35 W, a 10 s etch duration and at a vacuum pressure of 25 mTorr. The resist was then removed in warm NMP followed by a rinse in IPA. Introducing the isolation lines on the flakes by this method greatly reduced the He-ion milling exposure time by reducing the amount of graphene that needed to be milled by the He beam to isolate the pattern. This allowed milling of complex patterns in graphene whilst avoiding the problem of distortion due to drift. Secondary electron images, taken with the helium ion microscope, of a graphene flake after e-beam processing, are shown in Fig. 4.