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## Ultraviolet/ozone treatment to reduce metal-graphene contact resistance

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We report reduced contact resistance of single-layer graphene devices by using ultraviolet ozone treatment to modify the metal/graphene contact interface. The devices were fabricated from mechanically transferred, chemical vapor deposition grown single layer graphene. Ultraviolet ozone treatment of graphene in the contact regions as defined by photolithography and prior to metal deposition was found to reduce interface contamination originating from incomplete removal of poly(methyl-methacrylate) and photoresist. Our control experiment shows that exposure times up to 10 min did not introduce significant disorder in the graphene as characterized by Raman spectroscopy. By using the described approach, contact resistance of less than  $200 \,\Omega \,\mu$ m was achieved for 25 min ultraviolet ozone treatment, while not significantly altering the electrical properties of the graphene channel region of devices. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4804643]

Graphene is considered a candidate material for postsilicon electronics,<sup>1</sup> and graphene-based electronic and optoelectronic devices have developed rapidly in recent years.<sup>2</sup> In graphene-based devices, the metal/graphene contact is viewed as a limiting factor in its performance.<sup>3–7</sup> Ohmic metal/graphene contacts with low contact resistance  $(R_c)$  are necessary for graphene FET performance to approach its expected high intrinsic speed. To date, the best reported R<sub>c</sub> for lithographically defined contacts deposited onto exfoliated graphene flakes ranges from  $200 \Omega \mu m$  to  $500 \Omega \mu m$ .<sup>3,4,8</sup> R<sub>C</sub> for contacts formed to epitaxial graphene on SiC have been reported to be less than  $100 \Omega \mu m$  (Ref. 9) and with specific contact resistivity ( $\rho_c$ ) of order  $10^{-7} \Omega \text{ cm}^{2.9,10}$  The best reported values of R<sub>c</sub> for chemical vapor deposited (CVD) graphene typically range from  $500 \Omega \mu m$  to several thousand  $\Omega \mu m.^{3,6,11}$  Despite the technological attractiveness of CVDgrown graphene, these contact resistances remain too large for most applications and are far from that reported for contacts to epitaxial graphene on SiC.

Possible contributors to the large, experimentally determined metal/graphene contact resistance include dipole formation at the interface due to charge transfer, perturbation of the graphene beneath the metal,<sup>12</sup> and contamination of the metal/graphene interface.<sup>10,13</sup> Interface contamination during the device fabrication, especially when a photolithography process is employed, is known to be problematic and efforts are made to clean the contact interface in conventional semiconductor processing. Using the same or similar photolithographic processes to pattern contacts onto graphene make it reasonable to expect photoresist residue contamination on graphene surface.<sup>13</sup> Several researchers have introduced methods to reduce the contact resistance. For example, specific contact resistivity as low as  $10^{-7} \Omega \text{ cm}^2$  was obtained from short channel length (L <  $3 \mu m$ ) transfer length method (TLM) on epitaxial graphene by a low power plasma treatment.<sup>10</sup> Unfortunately, the plasma treatment is aggressive and after tens of seconds of treatment, the graphene can be seriously degraded, leading to a high variance in device to device contact resistance.<sup>14</sup> R<sub>c</sub> less than 100  $\Omega$   $\mu$ m has been reported for contacts formed to epitaxial graphene on SiC by other researchers, but details about device processing, importantly, contact formation are absent in the report.<sup>9</sup> Using a double contact device geometry  $R_c$  of  $200 \Omega \ \mu m$  to  $500 \Omega$  $\mu$ m on CVD-grown graphene was reported.<sup>7</sup> Similar contact resistance was reported for metal contacts to CVD-grown graphene by introducing an Al sacrificial layer.<sup>6</sup> Thermal annealing in vacuum at 300 °C to 400 °C has been shown to be helpful for cleaning the graphene channel region and improving the device performance by other groups,<sup>10,13,15,16</sup> but it is not very effective for cleaning the metal-graphene interface since it is already covered by metal. Unfortunately, thermal annealing cannot be employed just after the contact window opening in the resist layer and before the metal deposition, because the thermal budget is not suitable for post resist processing (resist flow, loss of features, resist crosslinking, etc.). Such strategies complicate the device fabrication process and make it necessary to develop a simple and robust process for reducing the metal/graphene contact resistance.

In this work, we report significantly reduced contact resistance to CVD-grown, single-layer graphene obtained by using a simple graphene surface cleaning method: ultraviolet ozone (UVO) treatment. UVO is a common cleaning process used in semiconductor device research and manufacturing, and in applications requiring critically clean interfaces such

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as those involving the assembly of molecules on metal or oxide surfaces for which aggressive plasma and ion bombardment processes cannot be tolerated.<sup>17</sup> By using UVO, we are able to reduce  $R_c$  to mechanically transferred, CVD-grown single-layer graphene to less than  $200 \,\Omega \,\mu$ m while preserving the electrical properties of the graphene device.

In graphene devices as well as other semiconductor devices, the current flows between the graphene and the metal contact non-uniformly. The current is highest at the metal contact edge and decreases exponentially with distance under the contact. The distance over which 1/e of the current is transferred to the metal contact is defined as the transfer length, L<sub>T</sub>. The macroscopic contact resistance is therefore  $R_c = \rho_c/(WL_T) \operatorname{coth}(L_C/L_T)$ , where  $\rho_c$  is the interface resistivity and W and L<sub>C</sub> are the width and length of the metal electrode contact.<sup>18,19</sup> For L<sub>C</sub> > 1.5L<sub>T</sub>,  $R_c \approx \rho_c/(WL_T)$ , where WL<sub>T</sub> is the effective contact area. For most reported graphene devices, the transfer length ranges between tens and several hundred nanometers,<sup>8,18</sup> which are usually much less than the metal contact length. For the case of  $L \gg L_T$ , contact resistance per unit width ( $\Omega \mu m$ ) is widely used to characterize the contact and compare findings.<sup>4,6–8</sup> In this work, we report on contact resistance for devices fabricated with L<sub>C</sub> much greater than the extracted and calculated values for L<sub>T</sub>, and thus we also report contact resistance per unit width ( $\Omega \mu m$ ).

In this study, we fabricated TLM test structures from single layer graphene that was grown on Cu foil by CVD method and then mechanically transferred onto a heavily doped Si substrate with 300 nm SiO<sub>2</sub> using a "modified RCA clean method."<sup>20</sup> Following the graphene transfer onto the SiO<sub>2</sub> surface, the test structures were fabricated by using conventional contact photolithography and metal deposition. The process flow is shown schematically in Fig. 1. After opening the windows for the metal contacts in the photoresist layer, the substrate was placed into a commercial UVO system to remove resist residue prior to metallization. Ti (20 nm)/Au (80 nm) was evaporated and patterned by lift-off process. A second photolithography step and oxygen plasma etching were used to pattern the graphene channel. The sacrificial photoresist mask used to protect the graphene channel region during the etch process was removed by using solvents.

We first evaluate the aggressiveness and effectiveness of the UVO cleaning step by using atomic force microscopy (AFM) (Fig. 2) and Raman spectroscopy (Fig. 3). A poly (methyl-methacrylate) (PMMA) layer was used as a polymer support layer during the mechanical transfer process of the graphene and needed to be removed at the end of the transfer process before proceeding with the first photolithographic step. However, PMMA was not thoroughly removed with solvents for overnight immersion and a thin residue layer still remained on the graphene surface. This is visible in Fig. 2(a), which shows the AFM topography image for a transferred, single-layer graphene domain after the solvent removal of PMMA. Next, 1  $\mu$ m thick commercial photoresist (AR-p 5350 from Allresist GmbH) was spin coated onto the transferred graphene surface and the substrate went through the same exposure and developing steps used in the fabrication of the TLM test structures. Thus, Fig. 2(b) shows representative surface topography of the contact regions after developing the resist openings and just prior to the metal deposition. The rough surface features on the single layer graphene indicates substantial resist residue remains on the graphene surface and we expect, in the absence of additional cleaning processes, that this residual resist will prevent the formation of an intimate metal/ graphene contact interface. Figs. 2(c)-2(f) sequentially show the results of accumulative 5, 10, 16, 22 min UVO treatments. The surface appears smooth after about 16 min and no further change of the surface topography was observed for UVO exposure up to 22 min. The root mean square surface roughness increases from 2.20 nm (Fig. 2(a)) to 7.89 nm (Fig. 2(b)) and then decreases to 1.49 nm (Fig. 2(e)) (see supplementary material, Fig. S1).<sup>27</sup> These results indicate the resist residue on graphene surface which comes from both the transfer process and the photolithography process was effectively removed.



FIG. 1. Schematic illustration of the graphene device fabrication process after mechanical transfer of the CVD-grown single layer graphene and the solvent removal of the sacrificial PMMA layer.

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FIG. 2. AFM images of the graphene surface topography throughout the UVO treatment process. (a) After transfer and solvent removal of PMMA, (b) after photolithography, and (c)–(f) UVO treatment for 5, 10, 16, 22 min, respectively. Scale bar:1  $\mu$ m, Color scale: 10 nm.

Fig. 3 shows the Raman spectra taken at nominally the same position on the graphene domain shown in Fig. 2 and in parallel with the AFM topography scans during the series of accumulative UVO treatment times. Raman spectroscopy has become a widely used characterization method for evaluating the quality of graphene.<sup>21,22</sup> Usually three Raman peaks near  $1580 \text{ cm}^{-1}$  (G peak),  $2650 \text{ cm}^{-1}$  (G' peak), and  $1350 \text{ cm}^{-1}$  (D peak) are observed in the spectra of graphene.<sup>21,22</sup> A high D-to-G peak intensity ratio correlates to a greater degree of disorder in the graphene structure and increased charge carrier scattering. In our experiments during the UVO treatment process, the Lorentzian fitted D-to-G peak area ratio<sup>23</sup> remains to a low value, and a marked increase was not observed until 22 min treatment (see supplementary material, Fig. S2).

It is important to acknowledge here that UVO processes can vary greatly among UVO systems and depend on the specific configuration and use in individual laboratories (e.g., exhaust rate, feed gas, exposure time, sample-to-grid lamp distance, sample temperature, and lamp intensity). In fact, one



FIG. 3. Raman spectra series for the transferred CVD-grown single layer graphene during the UVO treatment process. All spectra were taken from roughly the same position from the sample shown in Fig. 2 for comparison.

early study using aggressive UVO processing conditions reports significant damage to pristine graphene at short time scales.<sup>24</sup> We have collected additional Raman spectra (not shown) on mechanically transferred, CVD-grown single-layer graphene post photoresist processing and after UVO treatment in a different UVO systems and obtained D-to-G peak intensity ratios similar to that shown in Fig. 3 for the first 10 min of UVO treatment. However, a pronounced increase in D peak intensity (increase in D-to-G peak intensity ratio) is observed after 16 min of UVO treatment. Additionally, results from preliminary X-ray photoelectron spectroscopy (XPS) studies (not shown) on these same samples reveal changes in the C 1s and Si 2p peak intensities with UVO exposure time that indicate organic contamination and removal with UVO exposure. XPS data indicated the eventual degradation of the graphene when exposed to longer UVO treatment times entirely consistent and coincident with the pronounced emergence of the D peak in the Raman spectral.

Contact resistance was extracted from TLM test structures that were fabricated by using the process flow depicted in Fig. 1. Fig. 4(a) shows the optical micrograph (contrast enhanced) of a TLM test structure. The width (W) of the photolithographically defined graphene strip (device channel) is 10  $\mu$ m and length (L<sub>C</sub>) of the metal contacting the graphene strip is  $6 \,\mu m$ . Fig. 4(b) shows a plot of representative width normalized current-voltage (I-V) characteristics of three devices with the same inter-electrode separation  $(L = 22.5 \,\mu m)$ , but with different UVO treatment time, as well as the I-V characteristics for a device with identical L but processed without UVO treatment. All I-V characteristics are linear over a large applied voltage range and indicate the contacts are ohmic. The measurements were taken at room temperature in air with the back-gate electrode grounded. Fig. 4(c) shows gate modulation of the current according to Fig. 4(b). For our Ti/Au contacted test structures, the neutrality point is shifted positive by many 10's of volts in air. Therefore, the reported contact resistance here is for the hole. However, the neutrality point is restored to near  $0 V (V_{\sigma})$  when measured in vacuum (Fig. 4(c), inset). It is



FIG. 4. (a) Optical micrograph of a completed graphene TLM test structure. (b) and (c) Width normalized output (b) and transfer (c) characteristics of  $22.5 \,\mu$ m channel length devices with and without UVO treatment which were measured in air. Inset in (c): transfer characteristics measured in vacuum.

known that  $R_c$  measured far from the Dirac point is almost independent of the gate bias.<sup>8</sup> Our measurements confirm this trend, and  $R_c$  extracted from measurements made in air coincide well with  $R_c$  extracted from measurements made in vacuum.

Fig. 5(a) shows the measured resistance (combined probing pad, contact, and graphene channel) versus contact separation of typical TLM structures as a function of contact interface conditioning. The measured resistance is the aggregate value calculated from the linear I-V characteristics for large voltage sweeps (0 V to 0.5 V, 0.01 V steps). Two notable observations are (1) the total resistance is greatly reduced by the UVO treatment and (2) the change in resistance with L (contact separation or channel length) for test structures



FIG. 5. Electrical characteristics of graphene devices. (a) Total resistance vs. contact separation for different TLM test structures without and with UVO treatment. (b) Width normalized contact resistance and graphene sheet resistance with and without UVO treatment.

with and without UVO treatment is similar. These observations provide a first indication that the contact resistance is strongly affected by the UVO treatment but the channel resistance is not. The contact resistance and the channel resistance were extracted from a linear fit to the data for  $L > 5 \,\mu m$ and the width normalized contact resistance and graphene sheet resistance (R<sub>s</sub>) are plotted in Fig. 5(b). The contact resistance was reduced more than 2 orders magnitude for a 25 min UVO treatment. As alluded to by the AFM and Raman studies discussed above, even a 10 min UVO treatment was found to remove enough residue to improve contact formation between the graphene surface and metal, as substantiated by the nearly  $100 \times$  reduction in the width normalized contact resistance.

By using a UVO treatment, we obtained  $R_c$  as low as 184  $\Omega \mu m$  (not corrected for the pad resistance) for 25 min UVO treatment, which is a very low normalized contact resistance to CVD-grown single layer graphene. We note that our linear extrapolations of  $R_c$  were limited to data collected for devices with  $L > 5 \mu m$ , but we have included data points for devices with  $L = 5 \mu m$  for completeness. Data for short L was excluded from the extrapolation because we consistently observed pronounced deviations from a linear fit to the data at shorter L. This observation may be due to the polycrystalline nature of the CVD-grown single layer graphene itself and shorter channels comprised of a single domain (Fig. 2). Another possible reason is that the graphene channel region near the contact is perturbed by the contact metal, which is more pronounced for short channel devices.<sup>5,12,25</sup>

 $L_T$  is obtained from intercept of the fitting line in the TLM extraction with the x-axis. The intercept occurs at  $2L_T = -0.82 \,\mu$ m, yielding an experimentally determined value for  $L_T = 0.41 \,\mu$ m. We calculate an expected value for  $L_T$  from  $R_C$ , W, and  $R_S$  by using the relationship defined in Refs. 18 and 19 and with the simplification for  $L_T \ll L_C$ . The calculated value of  $L_T = 0.44 \,\mu$ m is in very close agreement with the value extrapolated from the linear fit.

Importantly, during the UVO treatment of the contact region, the graphene channel of the device remained masked by the photoresist. From the nearly unchanged values for sheet resistance, we conclude that the channel properties of the devices are not greatly affected by our contact treatment method. For completeness, we have characterized the room temperature field-effect properties of the TLM test structures

by applying a voltage to the heavily doped substrate with regard to the source contact while measuring the drain current. The carrier mobility can be extracted by fitting the I-V<sub>g</sub> curve using Eq. (1) of Ref. 26. To obtain a more accurate mobility, the vacuum measured results at  $V_{DS} = 0.1 V$  and  $V_{GS}$  swept from -60 V to 60 V were fitted (measurement conditions where the neutral point is restored to within a few volts from  $V_{GS} = 0$ ). The averaged electron and hole mobilities for all channel lengths are  $1773 \pm 574 \text{ cm}^2/\text{Vs}$ ,  $3264 \pm 32 \text{ cm}^2/\text{Vs}$ ,  $2178 \pm 178 \text{ cm}^2/\text{Vs}$ , and  $1725 \pm 383$ cm<sup>2</sup>/Vs, for no UVO treatment, 10, 16, and 25 min UVO treatments, respectively. The average mobility is found to be largely independent of the UVO treatment and we ascribe any variations in the mobility to the polycrystalline nature of the CVD-grown single layer graphene itself and structural imperfection introduced during the mechanical transfer of the graphene.

We have determined through AFM, Raman, and preliminary XPS studies that a major contributor to high contact resistance and poor device reproducibility of CVD-grown single-layer graphene devices is the resist residue left on the graphene surface after photolithographic processing. UVO is demonstrated to be a convenient and useful process for removing interfacial contamination from graphene and reducing contact resistance to record low values ( $<200 \,\Omega \,\mu m$ ) for photolithographically defined, metal contacts deposited onto CVD-grown monolayer graphene. Moreover, the channel properties of graphene devices are not significantly degraded at the expense of improved contact properties. These results contribute to increasing the likelihood that technologically relevant, CVD grown, single-layer graphene will find use in commercial electronic device applications.

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- <sup>27</sup>See supplementary material at http://dx.doi.org/10.1063/1.4804643 for UVO treatment details, surface roughness, and Raman D-to-G peak intensity ratio changes during the UVO treatment process.