I. INTRODUCTION

Graphene is a one atom thick hexagonal lattice of sp²-hybridized carbon atoms, which has attracted much attention recently due to its unique properties.1–3 By applying an electric field, the charge carriers in graphene can be tuned from holes to electrons, both having zero rest mass and high mobility. The combination of graphene electronic- and mechanical properties is interesting for both fundamental studies and electronic applications.4–9 Pristine graphene is charge neutral and exhibits excellent electronic properties. However, charged impurities, surface contaminants, and structural deformation contribute to local doping. This leads to an inhomogeneous charge density, the so-called electron-hole puddles.10–14 and a shift in the charge neutrality point, \( V_D \) (Dirac voltage). Impurities from microfabrication and sample handling are inherent in any graphene processing acting as external scattering centers and affecting the devices properties.15–21

Several techniques exist for cleaning graphene. Standard cleaning using solvents cannot remove all these residues.15,19–21 Most commonly, graphene is cleaned by high temperature annealing in inert, typically Ar/H₂, atmosphere.15,17,19,22,23 While this technique is able to remove most of resist residues through desorption, the coupling between the substrate and graphene may increase, leading to mechanical deformation of the graphene.21 Also, suspended graphene becomes rippled upon temperature cycling of only 100–200 K.24 Both these effects can cause degradation in device performance. Additionally, many substrates cannot sustain high temperature without oxygen atmosphere, which is incompatible with graphene, e.g., ferroelectric barium strontium titanium oxide (BSTO) thin films.25 Charged contaminants adhere very strongly to these substrates due to the polarized surface. This renders graphene on BSTO heavily contaminated and difficult to clean. Another common method is annealing by Joule heating.16,25 This technically simple technique can be done in situ in a cryostat. However, graphene is also in this case heated locally to high temperature leading to rippling or even breakage if too much current is applied. Just recently, mechanical cleaning of graphene was suggested.26–28

In this work, we use the mechanical cleaning method to obtain clean, atomically smooth graphene. We scan the tip of an atomic force microscope (AFM) in direct contact with graphene, removing contaminants in broom-like movements. Resist residues are efficiently brushed away, piling up outside the graphene flake. We show that this procedure produces atomically smooth graphene with improved electronic properties.

II. EXPERIMENT

We fabricate graphene using mechanical exfoliation on two different kinds of substrates:2; silicon with 290 nm thermally grown silicon dioxide and Nb-doped STO substrates with 50 nm BSTO thin films grown by pulsed laser deposition (PLD). On SiO₂, mono- and bilayer graphene are localized using optical microscopes and identified by its optical contrast.29 Control samples measured at low temperature and high magnetic fields confirm the number of layers through the quantum Hall effect.4 On BSTO, the contrast is significantly smaller and the number of layers is estimated from AFM measurements. Samples are patterned using electron beam lithography (EBL) and subsequent oxygen plasma etching. A second EBL step, followed by evaporation of typically 3 nm Ti and 60 nm Au and lift-off, defines electrodes. An overview of studied samples is shown in Table I.

We use AFM in tapping mode to observe the devices both before and after cleaning. In this mode, the AFM does not influence the graphene. Cleaning is done in contact mode using several different AFM probes and different forces. Electrical measurements are performed at room temperature before and after the cleaning. A voltage is applied to the conducting Si substrate acting as a back gate.

III. RESULTS

Typical height and phase (inset) images of graphene devices after fabrication are shown in Fig. 1(a). The height rms roughness, \( R_{\text{rms}} \), is 0.77 nm and 0.47 nm for a 0.5 × 0.5 \( \mu \text{m}^2 \) area of graphene and bare SiO₂ substrate, respectively. We then set the AFM to contact mode, pushing the TiN-coated Si tip in touch with the sample. While the tip is scanned back and forth over the sample, contaminants are mechanically...
TABLE I. Table of samples going through typical fabrication and cleaning procedures. $R_{\text{RMS}}$ is calculated over a $0.5 \times 0.5 \mu m^2$ area.

<table>
<thead>
<tr>
<th>Sample name</th>
<th>Substrate</th>
<th>Cleaning method</th>
<th>Graphene $R_{\text{RMS}}$ before—after cleaning (nm)</th>
<th>$V_D$ before—after cleaning (V)</th>
<th>Mobility before—after cleaning ($cm^2/Vs$)</th>
<th>Step height (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>S1</td>
<td>Wet SiO$_2$/Si</td>
<td>Hard</td>
<td>0.50 $\rightarrow$ 0.25</td>
<td>-</td>
<td>-</td>
<td>$\sim$1</td>
</tr>
<tr>
<td>S2</td>
<td>Wet SiO$_2$/Si</td>
<td>Hard</td>
<td>0.77 $\rightarrow$ 0.28</td>
<td>-</td>
<td>-</td>
<td>0.6–0.7</td>
</tr>
<tr>
<td>S3</td>
<td>Wet SiO$_2$/Si</td>
<td>Soft</td>
<td>0.30 $\rightarrow$ 0.12</td>
<td>$+12 \rightarrow -3$</td>
<td>4300 $\rightarrow$ 7700</td>
<td>0.5–0.6</td>
</tr>
<tr>
<td>S4</td>
<td>Wet SiO$_2$/Si</td>
<td>Soft</td>
<td>0.45 $\rightarrow$ 0.13</td>
<td>$+24 \rightarrow +2$</td>
<td>1200 $\rightarrow$ 1800</td>
<td>0.6</td>
</tr>
<tr>
<td>S5</td>
<td>Dry SiO$_2$/Si</td>
<td>Hard</td>
<td>0.65 $\rightarrow$ 0.21</td>
<td>$+20 \rightarrow -10$</td>
<td>1400 $\rightarrow$ 1800</td>
<td>0.6</td>
</tr>
<tr>
<td>S6</td>
<td>Dry SiO$_2$/Si</td>
<td>Hard</td>
<td>0.30 $\rightarrow$ 0.21</td>
<td>$+16 \rightarrow -1$</td>
<td>4200 $\rightarrow$ 5000</td>
<td>0.7</td>
</tr>
<tr>
<td>S7</td>
<td>BSTO / STO</td>
<td>Hard</td>
<td>1.82 $\rightarrow$ 1.17</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td>S8</td>
<td>BSTO / STO</td>
<td>Hard</td>
<td>1.60 $\rightarrow$ 1.17</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
</tr>
</tbody>
</table>

FIG. 1. (Color online) (a) Tapping mode AFM height measurement before cleaning. The inset shows the corresponding phase image. The surface is heavily contaminated after fabrication as seen in both height and phase images. The height rms roughness, $R_{\text{RMS}}$, is 0.77 nm and 0.47 nm for a $0.5 \times 0.5 \mu m^2$ area of graphene and bare SiO$_2$ substrate, respectively. (b) Tapping mode measurement of the same area as in (a) after contact mode cleaning in AFM. $R_{\text{RMS}}$ is now 0.28 nm and 0.29 nm for graphene and bare SiO$_2$ substrate, respectively. The arrows in (a) and (b) are pointing at the graphene. The scale-bars in (a) and (b) are all 1 $\mu m$. (c) Large-area tapping mode AFM height measurement of a graphene Hall-bar where the central part has been cleaned. The two ellipses point out beads with removed contaminants. The scale-bar is 2 $\mu m$ and the Z-scale is 25 nm. (d) Histogram of surface roughness from before (blue, dashed line) and after (red, solid line) cleaning, respectively.
pushed to the sides in a broom-like way. Typically three to five such scans are performed. The graphene is generally clean after only two scans and only minor improvement is seen after subsequent cleaning. In Fig. 1(b) the same area as in Fig. 1(a) is shown after four scans in contact mode. $R_{RMS}$ is now reduced to 0.28 nm and 0.29 nm for graphene and bare SiO$_2$ substrate, respectively.

An overview AFM image is shown in Fig. 1(c). The central area of the device is cleaned in contact mode. Around it, there are beads with pushed-away contaminants, as indicated by the two ellipses. The surface is significantly rougher outside the cleaned area. A histogram of the surface roughness before and after cleaning is shown in Fig. 1(d). The distribution becomes much more narrower after the cleaning.

Devices are fabricated on SiO$_2$ grown thermally using both wet- and dry oxidation. We measure $R_{RMS}$ for both the bare substrate and areas covered with graphene. While $R_{RMS}$ for pristine “dry” oxide is slightly less than that of “wet”, both show a wide range of $R_{RMS}$ $\sim$ 0.35–1.40 nm after fabrication. Corresponding values for graphene after fabrication are $R_{RMS}$

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**FIG. 2.** (Color online) (a) Resistance as a function of gate before (red, solid line) and after (blue, dashed line). The charge neutrality point moves toward zero after cleaning and the estimated mobility increases from $\sim$4300 cm$^2$/Vs to $\sim$7700 cm$^2$/Vs. The Z-scale is 8 nm. (b) and (c) AFM height images before and after cleaning of graphene on BSTO, respectively. The heavy contamination is removed and the atomic steps in the BSTO are clearly seen, including through graphene. Upper insets: AFM phase images. The phase response of both substrate and graphene is almost flat after cleaning, evident from the phase histograms (lower insets). All scale-bars are 500 nm.
~0.30–0.65 nm. After cleaning, we obtain \( R_{\text{RMS}} \approx 0.25–0.33 \) nm and \( R_{\text{RMS}} \approx 0.18–0.23 \) nm for wet and dry oxides, respectively. Corresponding values for graphene after cleaning are \( R_{\text{RMS}} \approx 0.12–0.25 \) nm for both types of oxide. The roughness of graphene depends more on the force applied in the AFM measurements of both bare SiO\(_2\) and graphene. A large contact force of 180 nN, the graphene is pushed down toward the substrate leading to similar roughness measurements with a small contact force are shown.

We find that the ferroelectric film, the contaminants are strongly adhered after cleaning. Resistance is measured as a function of gate voltage \( V_g \) applied to the Si substrate. We observe consistent changes in the charge neutrality point, \( V_{\text{NC}} = V_{D}, \) after cleaning. \( V_D \) is zero for ideal un-doped graphene. After fabrication, our devices show positive \( \Delta V_D \), indicating \( \Delta V_D \) increases after microfabrication to around 1.5–2.0 nm. After cleaning, it reduces to 0.6–0.7 nm, typical for clean graphene on SiO\(_2\).

Several samples are measured electrically before and after cleaning. Resistance is measured as a function of gate voltage \( V_g \) applied to the Si substrate. We observe consistent changes in the charge neutrality point, \( V_{\text{NC}} = V_{D}, \) after cleaning. \( V_D \) is zero for ideal un-doped graphene. After fabrication, our devices show positive \( \Delta V_D \), indicating \( \Delta V_D \) increases after microfabrication to around 1.5–2.0 nm. After cleaning, it reduces to 0.6–0.7 nm, typical for clean graphene on SiO\(_2\).

The measured step height corresponding to graphene increases after microfabrication to around 1.5–2.0 nm. After cleaning it reduces to 0.6–0.7 nm, typical for clean graphene on SiO\(_2\). Corresponding values for graphene after cleaning are \( R_{\text{RMS}} \approx 0.13 \) nm for graphene and \( R_{\text{RMS}} \approx 0.33 \) nm for bare SiO\(_2\), respectively.

IV. CONCLUSIONS

Mechanical cleaning of graphene using contact mode AFM is an easy way to obtain clean and atomically flat graphene after microfabrication. It improves the charge neutrality of graphene and, using moderate contact force, increases the mobility. This technique appears to be particularly indispensable in the case of graphene devices on ferroelectric BSTO. Despite its low throughput, we believe this strengthens the development of mechanical cleaning methods for graphene.

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