Support-Free Transfer of Ultrasmooth Graphene Films Facilitated by Self-Assembled Monolayers for Electronic Devices and Patterns

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ABSTRACT: We explored a support-free method for transferring large area graphene films grown by chemical vapor deposition to various fluoric self-assembled monolayer (F-SAM) modified substrates including SiO₂/Si wafers, polyethylene terephthalate films, and glass. This method yields clean, ultrasmooth, and high-quality graphene films for promising applications such as transparent, conductive, and flexible films due to the absence of residues and limited structural defects such as cracks. The F-SAM introduced in the transfer process can also lead to graphene transistors with enhanced field-effect mobility (up to 10,663 cm²/Vs) and resistance modulation (up to 12×) on a standard silicon dioxide dielectric. Clean graphene patterns can be realized by transfer of graphene onto only the F-SAM modified surfaces.

KEYWORDS: graphene, self-assembled monolayer, transfer, support-free, ultrasmooth, GFET, pattern

Transferring chemical vapor deposition (CVD)-grown graphene films from metal surfaces to target substrates while preserving their high quality is a major challenge, particularly for use in electronic devices.1−4 Thus far, transfer techniques have been reported involving the use of support materials such as polymers, namely, poly(methyl methacrylate) (PMMA), polydimethylsiloxane (PDMS), poly(bisphenol A carbonate) (PC), thermal release tape (TRT), and also through “electrochemical delamination”.13−22 The use of pressure sensitive tape has recently been described by Kim et al., who stated that this is an effective method for transfer of graphene from Cu foil and to various substrates.23 However, the polymer/tape-based methods suffer from support material residues left on the surface of the transferred graphene films, and additional cleaning process(es) that may lead to cracking or folding/wrinking of the graphene, resulting in a poorer performance of the graphene-based devices.24 Therefore, support-free transfer methods for CVD grown graphene films toward ultrasmooth graphene surfaces suitable for electronic devices are highly desirable.

Considering high performance graphene field-effect transistor (GFET) devices, it has been reported that the devices on SiO₂/Si substrates have a lower carrier mobility than suspended devices or devices on hexagonal boron nitride (h-BN) substrates.25,26 More recently, SAM (molecular organosilane self-assembled monolayer)-modified SiO₂/Si substrates have been reported to be efficient in modulating the electronic properties of graphene to make selectively doped FETs and boost field-effect mobility by attenuating surface polar phonon scattering.27−30 and perhaps by other factors such as improved surface flatness and fewer wrinkles and so on. However, support materials were all included in the fabrication of these graphene/SAMs devices, and the resulting surface contaminations may influence the as-measured transport performances.

Support-free transfer can be achieved by floating graphene films on the surface of solution through a slow and nearly static...
process or by enhancing the adhesion between the graphene and target substrates using physical approaches involving electrostatic and/or mechano-electrothermal forces.\(^{31-33}\) A transfer method to generate ultrasmooth and high quality graphene without contamination from the support materials, and that can modulate the electronic performance in useful ways, is needed.

Here we introduce a support-free method for transferring large area and ultrasmooth CVD-grown graphene films to trichloro-(1H,1H,2H,2H-perfluorooctyl)silane formed SAM (F-SAM) modified substrates including SiO\(_2\)/Si wafers, polyethylene terephthalate (PET) films, and glass. The target substrate was first pretreated to form a hydrophobic surface with the F-SAM (Figure 1). The graphene/Cu foil was then gently pressed on the modified substrate, followed by immersion in an aqueous copper etchant. Typically, graphene films tend to detach from the pristine SiO\(_2\)/Si wafer and become suspended in water due to the water molecules diffusing between the graphene and substrate (Figure 1b). In contrast, the hydrophobic groups terminating the SAM hinder the insertion of water molecules and thus maintain the high adhesion between graphene and the modified F-SAM-coated SiO\(_2\)/Si wafer during the process of Cu etching (Figure S1, Supporting Information, in which an atomic force microscopy/AFM image shows the transferred graphene without water at the interface between it and the F-SAM/Si wafer).\(^{35,36}\) This approach not only provides an efficient and clean route for transferring ultrasmooth CVD-grown graphene films onto different substrates, but also improves the electronic properties of the graphene devices due to the F-SAM on the substrate.

**RESULTS AND DISCUSSION**

In a typical experiment, a Si wafer with a 300 nm thick thermally grown oxide layer on the surface was used as the base substrate. For the untreated SiO\(_2\)/Si wafer, Figure 2a, the water contact angle was 71.1°, indicating a somewhat hydrophilic surface. The direct transfer of graphene onto this substrate was investigated, and no graphene was observed on the wafer after etching the Cu foil. A well-known technique was used to modify the surface of the SiO\(_2\)/Si wafer with SAMs, as shown in Figure S2. The SiO\(_2\)/Si substrates were first treated with O\(_3\) plasma, forming a highly hydrophilic surface with a water contact angle of 7.8° (Figure S3). During this pretreatment, silanol groups (\(-\text{SiOH}\)) were formed on the surface for the subsequent dechlorination reaction with silane coupling agents (see details in the Methods section). Briefly, vapor deposition in vacuum was used to form the SAMs rather than solution-based methods to avoid possible contamination of the transferred graphene by the solvents, here typically toluene, hexane, and ethanol. Three types of coupling agents, namely, trichloro(octadecyl)silane, trichloro(phenyl)silane, and trichloro(1H,1H,2H,2H-perfluorooctyl)silane, were used to form the SAMs, with the substrates thus referred to as CH\(_3\)–Si wafers, C\(_6\)H\(_5\)–Si wafers, and F–Si wafers, respectively.

X-ray photoemission spectroscopy (XPS), water contact angle goniometry, ellipsometry, and atomic force microscopy (AFM) measurements of thickness and roughness were carried out to evaluate each different SAM on the surface of the modified SiO\(_2\)/Si wafers. In the XPS survey spectra of the substrates ranging from 0 to 1000 eV (Figure S4), the C 1s peak with higher intensity compared with that of the pristine SiO\(_2\)/Si wafer was observed, indicating the successful formation of the SAMs. The F 1s peak was observed for the F–Si wafer, and no chlorine peak was observed for all three modified substrates, suggesting chemical bonding between these coupling agents and the silanol groups on SiO\(_2\). The water contact angles for the CH\(_3\)–Si wafers, C\(_6\)H\(_5\)–Si wafers, and F–Si wafers were 81.1°, 90.0°, and 105.2°, respectively. It is worth noting that the water contact angle of a wafer modified through the vapor deposition process is typically reported to be smaller than that obtained for SAMs deposited by the solution method, but it can provide a cleaner and smoother surface for the graphene transfer.\(^{36,37}\) The thickness of the SAMs were measured by ellipsometry to be 2.34 nm (CH\(_3\)–), 0.96 nm (C\(_6\)H\(_5\)–), and 1.37 nm (F–), respectively, which is consistent with the reported values for monolayer SAM coverage.\(^{28,37,38}\) The three modified substrates were used to attempt graphene transfer under similar conditions, as illustrated in Figure 1a. The F–Si wafer appears to maintain the most intact graphene film after transfer (Figure
2d), implying that the more hydrophobic surface affords a better transfer, or possibly other factors are also playing a role.

A 6.5 cm × 6.5 cm graphene film was transferred onto a 4-in. F–Si wafer (Figure S5, Figure 3a). The optical image in Figure 3c. The AFM data showed that the transferred graphene is a monolayer. Ten graphene regions as clean as we could respectively by PMMA and F-SAM methods were evaluated by AFM measurement. Around 1 nm, suggesting highly conformal contact between the graphene and the substrate. The thickness of the monolayer region is around 1 nm, suggesting highly conformal contact between the graphene and the substrate. The AFM data showed that the films were uniform and free of residue. The thickness of the monolayer region is around 1 nm, suggesting highly conformal contact between the graphene and the substrate.

The surface roughness of the graphene films transferred respectively by PMMA and F-SAM methods were evaluated by AFM measurement. Ten graphene regions as clean as we could find with the equal size of 5 × 5 μm² were selected and are shown in Figure S8 as a comparison. The average surface roughness of graphene transferred by the PMMA method is 1.20 nm (Table 1). The high surface roughness may be from a worse contact between graphene and the SiO₂ substrate and perhaps also the polymer residues. In contrast, the Rₚ is as low as 0.26 nm when the graphene was transferred by the F-SAM method, even lower than the roughness of the F-SAM itself. This observation suggests the F-SAM molecules under the graphene films function as a cushion to release the surface tension of graphene, yielding an ultrasmooth surface.

The optical images show that the CVD-grown graphene is mostly a monolayer but with randomly distributed bilayer patches, and the corresponding Raman spectra are shown in Figure 3d and Figure S9, respectively. For the monolayer graphene, the intensity of the 2D peak at 2683 cm⁻¹ with a full width at half-maximum (fwhm) of 28.7 cm⁻¹ is roughly twice that of the G peak at 1596.1 cm⁻¹, while the defect-related D-peak at 1345.3 cm⁻¹ has a relatively low intensity, indicating the high quality of the transferred graphene film and that it is a monolayer.

XPS measurements were performed to evaluate the surface cleanliness of the graphene transferred by the F-SAM method versus the PMMA method, Figure 3e. The C 1s peak of the graphene film transferred using the F-SAM method is located at 284.5 eV, corresponding to the binding energy of the sp² hybridized states, with a narrower bandwidth than the C 1s peak for the graphene transferred using the PMMA method. The broadening of the C 1s peak and a peak at 288.4 eV are attributed to the PMMA residue on the surface of the graphene. It is worth noting that the C 1s peak at 291.5 eV for the F-SAM transferred graphene corresponds to the binding energy of the F-SAM on the wafer (Figure S10), and the apparent decrease in the intensity is due to the complete coverage of the graphene film. The XPS spot size is about 900 μm.

The direct transfer of graphene onto transparent and/or flexible dielectric substrates can be achieved by a similar F-SAM transfer method. A similar F-SAM modification process as for the SiO₂/Si wafer can be applied to a glass substrate. The monolayer graphene deposited on glass has a sheet resistance of 500 Ω/□ and a transparency of ca. 97.0% at a wavelength of 550 nm, which is comparable to the theoretical transmittance of 97.7% for monolayer graphene at that wavelength. A PET film was treated with O₂ plasma for 20 min, and was then exposed to the vapor agent to generate a hydrophobic surface with a high contact angle (Figure S11), resulting in the successful transfer of graphene (details in Methods). The transfer of graphene onto these two types of substrates is illustrated in Figure 3f. The bendable graphene/PET film in Figure 3f exhibits a slightly higher transparency of 97.3% than graphene on glass at a wavelength of 550 nm. These results confirm the promise of the F-SAM method for transferring CVD-grown graphene onto glass and flexible substrates.

Three-terminal back-gated GFETs were fabricated to evaluate the electrical performance of the transferred graphene (Figure S12). A resist-free shadow mask process (Methods section) was used to form devices without any potential polymer residue contamination to the graphene surface which is usual for a fabrication process involving lithography. These shadow-mask devices have a typical channel length and width both at the 200 μm scale and electrical measurements are performed at room temperature and ambient pressure. From Figure 4a, the graphene transferred by the F-SAM method shows a positive voltage shift of ca. 5 V compared with that transferred using the PMMA method, indicating it is more

Table 1. Surface Roughness (Rₚ) Values of Graphene Transferred Respectively by PMMA and F-SAM Methods, Corresponding to the AFM Measurements in Figure S8

<table>
<thead>
<tr>
<th>samples</th>
<th>#1</th>
<th>#2</th>
<th>#3</th>
<th>#4</th>
<th>#5</th>
<th>#6</th>
<th>#7</th>
<th>#8</th>
<th>#9</th>
<th>#10</th>
<th>avg</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMMA-G</td>
<td>1.02</td>
<td>1.32</td>
<td>1.44</td>
<td>1.22</td>
<td>1.19</td>
<td>1.59</td>
<td>1.39</td>
<td>1.29</td>
<td>1.12</td>
<td>1.39</td>
<td>1.20 nm</td>
</tr>
<tr>
<td>SAM-G</td>
<td>0.27</td>
<td>0.29</td>
<td>0.25</td>
<td>0.27</td>
<td>0.32</td>
<td>0.25</td>
<td>0.20</td>
<td>0.26</td>
<td>0.28</td>
<td>0.26</td>
<td>0.26 nm</td>
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</table>
heavily p-doped, which is consistent with a reported graphene FET modified by the same F-SAM. The modulation of the charge carriers in graphene by the F-SAM molecules can be explained by a "built-in" electric field superimposed on the externally applied gate field. The generated electric field inside the F-SAM, was determined by $E_{\text{in}} = N(\mu_{\text{mol}}/e\mu_{\text{mol}})$, where $N$ and $\mu_{\text{mol}}$ are the areal density ($1-2 \times 10^{14}$ cm$^{-2}$) and length of the F-SAM molecules (1.1 nm), respectively, $\mu_{\text{mol}}$ is the dipole along the molecular axis ($\approx 2.202$ D was calculated using density functional theory), and $e$ is the dielectric constant inside the F-SAM. The negative electric field determined by the dipoles requires a positive shift of the voltage and thus agrees well with the p-doping of the graphene in the GFET device. The gate modulation ($R_{\text{max}}/R_{\text{min}}$ for each curve at typical plots in Figure 4b) of F-SAM-GFET transferred graphene is more than one order of magnitude (up to $12 \times$), whereas the gate modulation of PMMA-GFETs is $\approx 7$. This indicates that F-SAM transferred graphene has fewer surface charge trapping sites, leading to improved gate control in accordance with improved field-effect mobility as seen from the following detailed discussion.

To analyze the difference between the transferred graphene films (F-SAM versus PMMA methods) quantitatively, the carrier mobility ($\mu$) and the residual carrier concentration ($n_0$) of the samples can be extracted by using a well-accepted diffusive model. The carrier concentrations (electrons or holes) in the graphene channel $n_{\text{total}}$ can be approximated by

$$n_{\text{total}} = \sqrt{n_0^2 + n_G^2}$$  \hspace{1cm} (1)

where the residual carrier concentration ($n_0$) is generated by charged impurities located either in the dielectric or at the graphene/dielectric interface, and $n_G$ is the carrier concentration resulting from the difference between $V_G$ and $V_{\text{Drac}}$ as determined from the following equation:

$$V_G - V_{\text{Drac}} = \frac{e}{C_{\text{ox}}} n_G + \frac{n_0 \sqrt{2}\gamma n_G}{e}$$  \hspace{1cm} (2)

where $e$ is the electron charge, $C_{\text{ox}}$ is the gate capacitance $\approx 35$ nF·cm$^{-2}$ for 90 nm SiO$_2$ (11 nF·cm$^{-2}$ for 285 nm SiO$_2$), $h$ is the Planck constant $6.62 \times 10^{-34}$ J·s, and $V_F$ is the Fermi velocity $1.15 \times 10^6$ m·s$^{-1}$. The total resistance $R_{\text{total}}$ of the device can be approximated by

$$R_{\text{total}} = R_c + R_{\text{ch}} = R_c + \frac{1}{W \mu \sqrt{n_0^2 + n_G^2}}$$  \hspace{1cm} (3)

with $R_c$ is the contact resistance, $R_{\text{ch}}$ is the graphene channel resistance, and $\mu$ is the mobility. While $n_0$ is dependent on the applied gate voltage $V_{\text{g}}$, the relevant parameters, $\mu$ and $n_0$, can be extracted by fitting to the measured data, for instance, the typical plots in Figure 4b.

The statistics of the carrier mobility extracted from a dozen different experimental devices is exhibited (Table S1). The highest field-effect mobility of graphene transferred by the PMMA method was $4563$ cm$^2$ V$^{-1}$ s$^{-1}$ with $n_0 = 4.4 \times 10^{11}$ cm$^{-2}$, while the average values were $2996$ and $3022$ cm$^2$ V$^{-1}$ s$^{-1}$ for holes and electrons, respectively, suggesting the carrier mobility value is influenced by the polymer residue and/or cracks in the graphene generated during the PMMA-facilitated transfer process (Figure 4c). In contrast, the highest value obtained in F-SAM-GFETs is $\mu \approx 10,663$ cm$^2$ V$^{-1}$ s$^{-1}$ with $n_0 \approx 5.2 \times 10^{12}$ cm$^{-2}$, while the average values were $7293$ and $7173$ cm$^2$ V$^{-1}$ s$^{-1}$ for holes and electrons, respectively. The average field-effect mobility of F-SAM transferred graphene is $\approx 2.5$ times that of the graphene transferred by the conventional PMMA-facilitated process, indicating a graphene of higher quality.

The sheet resistances of graphene films on SiO$_2$/Si and PET substrates transferred by F-SAM and PMMA methods were measured using a four-point probe method to exclude the contact resistance. Figure 4d compares the statistics of the sheet resistance. The average values were respectively $\approx 800$ and $\approx 700$ Ω·sq$^{-1}$ for graphene on SiO$_2$/Si and PET substrates transferred by F-SAM method, while the values were $\approx 1500$ and $\approx 1800$ Ω·sq$^{-1}$ for the PMMA transfer method. The sheet resistances of graphene transferred by the F-SAM method are only half the values from PMMA transferred graphene, and this improvement can be attributed to the doping effect, lower roughness$^{48-50}$ and higher quality of the F-SAM-transferred graphene.

Inspired by the selective transfer of graphene onto F-SAM modified surfaces, we realized clean patterning of a graphene membrane on a SiO$_2$/Si substrate without using the conventional photoresist or beam resist coating and removal processes. First, patterned Cu dots/ribbons were thermally evaporated onto a SiO$_2$/Si substrate through a shadow mask (Figure 5a), then the F-SAM layer was deposited on this substrate in vacuum. By etching away the Cu patterns, a reverse patterned SAM was obtained. Afterward, a graphene film was transferred onto the modified substrate and imitated the configuration of the SAM pattern to form a graphene pattern after removing the Cu foil. As shown in Figure 5b, graphene was not transferred to the square regions where no SAM existed except for some graphene fragments, thus forming a continuous graphene membrane with regular square holes. By using the shadow mask with arranged slits, graphene microribbons were obtained as exhibited in Figure 5c. It should be noted that this patterning method combines the transfer and pattern into a one-step

![Figure 4. Electrical characterization of fabricated graphene field effect transistors (GFET): (a) gate-dependent conductance, $V_G = 20$ mV. (b) Normalized $R_{\text{tot}}$ as a function of $V_G - V_{\text{Drac}}$ of GFETs where graphene were transferred by the PMMA and F-SAM methods. (c) Statistical value of extracted field-effect mobility from a dozen devices. (d) Sheet resistance of graphene transferred onto SiO$_2$/Si and PET by F-SAM and PMMA methods, respectively.](image-url)
SiO2/Si wafer, glass and PET require transparent, conductive, and flexible surfaces with a hydrophobic SAM. This method yields clean, ultra-smooth substrate surfaces that are free from structural defects, and the absence of polymer residues. The fluoride chemistry of graphene, due to ultrasmooth surfaces, limited the contact between graphene/Cu and the substrate to improve the electrical measurements when using a 4156 analyzer.

Graphene Patterns. Using a graphene mesh with square holes as an example, 100 nm patterned Cu dots (square, 150 μm × 150 μm) were first thermally evaporated onto a SiO2/Si substrate through a shadow mask. The F-SAM was then deposited on this substrate in vacuum for 6 h. By etching away the Cu patterns, a reverse patterned SAM was obtained. Afterward, a graphene film was transferred onto the modified substrate and imitated the configuration of the SAM pattern to form the graphene pattern after removing Cu foil. Finally, the obtained graphene pattern on the wafer was immersed in deionized water to remove the residues. Further sonication in water can be used to clean the surface without ruining the pattern.

Characterization. The water contact angle was measured by a drop shape analyzer, DSA100. Raman spectroscopy was performed with a WaTeC micro-Raman instrument using 532 nm laser at ambient temperature. A ZEISS optical microscope (AxioCam MRc5) was used to characterize the morphology of the transferred graphene. The thickness of the SAMs were determined using an Alpha XPS system (Thermo Fisher Scientific, Inc.) with a 90 μm beam size. The electrical properties were measured with a 4200-SCS semiconductor characterization system (Keithley).

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsnano.5b06842.

AFM image of single crystalline graphene, schematic of chemical modification, water contact angle of SiO2/Si wafer after O2 plasma, XPS surveys, image of Cu/graphene on treated wafer, optical image of graphene transferred by PMMA method, the number of cracks on graphene films transferred by different methods, comparison of AFM images, Raman spectrum of bilayer graphene, XPS of F−Si wafer, water contact angles of glass and PET, optical image of GFET, and the statistics of carrier concentration and mobility (PDF).

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Notes

The authors declare no competing financial interest.

Fabrication of Graphene FETs. Au/Ti (60/2 nm) was deposited in an e-beam evaporator for the metal contact of the three-terminal graphene device in a vacuum of 2.0 × 10−6 Torr. A resist-free shadow mask process was used to define source and drain contacts, and the device isolation was achieved by probe-tip patterning. The devices had a channel length of 200 μm with a channel width of 200 μm. The process, which did not use lithography, was used to avoid the e-beam or photo resist contamination of the graphene surface, and the relatively large active region of the shaped graphene channel was for better macroscopic observation. The electrical measurements were performed at room temperature and ambient pressure on a Cascade probe station with an Agilent 4156 analyzer.

CONCLUSION

In summary, an effective method has been developed for the support-free transfer of CVD-grown graphene films using SiO2/Si wafer, glass or PET film substrates modified in a simple way with a hydrophobic SAM. This method yields clean, ultra-smooth and high-quality graphene films for applications that require transparent, conductive, and flexible films, and for fundamental research on, for example, the mechanics and chemistry of graphene, due to ultrasmooth surfaces, limited structural defects, and the absence of polymer residues. The F-SAM introduced in the transfer process also improved the carrier mobility of the graphene films. Patterned graphene was obtained by transfer to patterned F-SAM modified surfaces. This simple and cost-effective process appears to be scalable.

METHODS

General SAM Treatment of the Substrates. Three coupling agents, CH3(CH2)3SiCl (trichloro(octadecyl)silane), CH3SiCl3 (trichloro(phenyl)silane), and CF3(CF2)2(CH2)3SiCl3 (trichloro(1H,1H,2H,2H-perfluorooctyl)silane), were used to form SAMs individually under as close to identical conditions as possible on SiO2/Si wafer, glass and PET film target substrates. Typically, these substrates were first exposed to O2 plasma for 10 min to modify the surface with silanol groups (–SiOH) for further chemical reaction with the molecules, while the PET films were treated twice. The agent solution (50 μL) was added to a vial and put into a vacuum desiccator for 6 h at room temperature. The obtained substrates were rinsed with ethanol and dried with nitrogen before use.

Graphene Transfer. Graphene films were grown on Cu foils as reported by Li et al. Then, the graphene on one side was etched by exposing it to an O2 plasma for 10 min. After a rolling treatment to flatten the foil, the Cu foil/graphene was gently placed on the SAM modified substrates and then immersed in an aqueous etchant (0.1 M (NH4)2S2O8). A roller was further used to squeeze out excess air in between graphene/Cu and the substrate to improve the contact. After the Cu foil was etched away, the substrates were thoroughly rinsed with deionized water and dried with nitrogen.

The typical PMMA-based transfer method comprises the following steps: (1) spin-coating a layer of PMMA on one side of a graphene/Cu foil followed by the removal of the other side of the graphene by an O2 plasma; (2) floating the film on aqueous (NH4)2S2O8 to etch away the Cu foil; and (3) subsequently transferring the film to the target substrate, and removing the PMMA layer by acetone.
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