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Metal-film-assisted ultra-clean transfer of single-walled carbon nanotubes

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ABSTRACT

Transfer printing of nanomaterials onto target substrates has been widely used in the fabrication of nanodevices, but it remains a challenge to fully avoid contamination introduced in the transfer process. Here we report a metal-film-assisted method to realize an ultra-clean transfer of single-walled carbon nanotubes (SWCNTs) mediated by poly(methyl methacrylate) (PMMA). The amount of PMMA residue can be greatly reduced due to its strong physical adhesion to the metal film, leading to ultra-clean surfaces of both the SWCNTs and the substrates. This metal-film-assisted transfer method is efficient, nondestructive, and scalable. It is also suitable for the transfer of graphene and other nanostructures. Furthermore, the relatively low temperature employed allows this technique to be compatible with nanomaterial-based flexible electronics.

1 Introduction

Incorporating nanomaterials into nanoelectronic or flexible electronic devices has been one of the central goals for the application of nanotechnology. Currently the growth of nanomaterials is not compatible with the semiconductor process, because of the requirement of the high growth temperature and the specific substrate. For example, horizontally aligned single-walled carbon nanotubes (SWCNTs) and large-area graphene of high quality can be synthesized on quartz substrates [1–3] and copper foils [4], respectively, at a temperature of around 1,000 ℃. Thus transferring synthesized nanomaterials to silicon wafers or flexible substrates plays a crucial role in the fabrication of nanoelectronic and flexible electronic devices [5–8].

On the other hand, nanomaterials are very sensitive to surface contamination due to the huge surface to volume ratio. The performance of nanodevices will be severely deteriorated by the surface or interface

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contaminations. Thus an ultra-clean transfer method, which can guarantee clean surface of transferred nanomaterials, has been a key challenge in this field.

The current transfer methods generally utilize a sacrificial layer as the transfer media. Typically this sacrificial layer is an organic material, such as poly(methyl methacrylate) (PMMA) [9–13], which is adhesive to nanomaterials, such as SWCNTs and graphene. The nanomaterials are first coated with an organic film. Then the organic film together with the nanomaterials is separated from the growth substrate and attached to the target substrate. Finally, this organic film is removed by dissolving in acetone, leaving the nanomaterials on the target substrate. However, the organic residue cannot be fully removed from the nanomaterials and the target substrate, resulting in a poor contact between the nanomaterials and the post-fabricated metal electrodes. It has been proposed that annealing in argon (Ar) atmosphere can further remove the residue, since most of the organic materials can decompose at high temperature [9]. However, this treatment cannot be applied to plastic substrates because the substrates themselves cannot withstand high temperature. Therefore we cannot obtain clean SWCNTs or graphene on flexible substrates by the conventional polymer-mediated transfer. Metal films such as Au can also be employed as the sacrificial layer to peel off nanomaterials from the growth substrate [14–16]. However, the metal residue will affect the performance of nanodevices as well. So far, an effective method to transfer SWCNTs and other nanomaterials with no residue remaining is still lacking.

Here we show that this problem can be overcome by slightly modifying the conventional PMMA-mediated transfer printing technique. By depositing an additional metal film (e.g. Ti, Au, Cr, Al, or Ni) onto the PMMA sacrificial layer, SWCNTs can be transferred to the target substrate with little residue left, which is verified by atomic force microscopy (AFM), Raman spectroscopy and transmission electron microscopy (TEM) characterization. We demonstrate that it is the strong adhesion between the metal film and the PMMA film that enables the PMMA to be cleanly peeled off in acetone, leaving ultra-clean SWCNTs on the target substrate. With this efficient and nondestructive method, we could even fabricate ultra-clean complex structures via multiple transfers. Moreover, this metal-film-assisted technique is compatible with flexible substrates owing to its operation at relatively low temperature, and will play an important role in future flexible electronics and optoelectronics.

2 Experimental

The metal-film-assisted transfer technique is schematically shown in Fig. 1. Horizontally aligned SWCNTs were synthesized on a stable temperature-cut (ST-cut) quartz substrate via chemical vapor deposition (CVD) [1–3]. PMMA solution (Mw = 495 k, 4 wt.%) was spin-coated onto the quartz substrate and then baked at 180 ˚C for 2 min to form a 190 nm-thick dense film [9]. This PMMA/SWCNT film was separated from the quartz substrate in NaOH aqueous solution (1 mol·L^{-1}, 100 ˚C), and then attached onto a SiO_2/Si (thickness of the SiO_2: 300 nm) substrate, as illustrated in Fig. 1(a). The process above is the same as the conventional transfer technique. Afterwards, an additional metal film (20 nm Ti) was deposited onto the PMMA surface. The metal-coated PMMA could be peeled off from the substrate after dipping into acetone for 5 min (Fig. 1(b)), leaving clean SWCNTs on the substrate (Fig. 1(c)). A reference specimen was also made by the conventional PMMA-mediated transfer method (Fig. 1(d)). The substrate was also dipped into acetone, and it usually takes a longer period (about 30 min) to dissolve the PMMA. By comparing the AFM images of the metal-film-assisted transferred SWCNTs (Fig. 1(c)) and the conventional transferred SWCNTs (Fig. 1(e)), we can clearly see that by both methods SWCNTs are accurately transferred to the target substrate. However, the SWCNTs transferred by the metal-film-assisted method are neat and clean, while the specimen transferred by the conventional method shows a lot of PMMA residue on the sample surface. This difference was also confirmed by Raman spectroscopy, as shown in Figs. 1(f)–1(h). The SWCNTs transferred by the metal-film-assisted technique have a lower intensity of D mode, which mainly originates from defects and impurities, compared with the conventional transferred

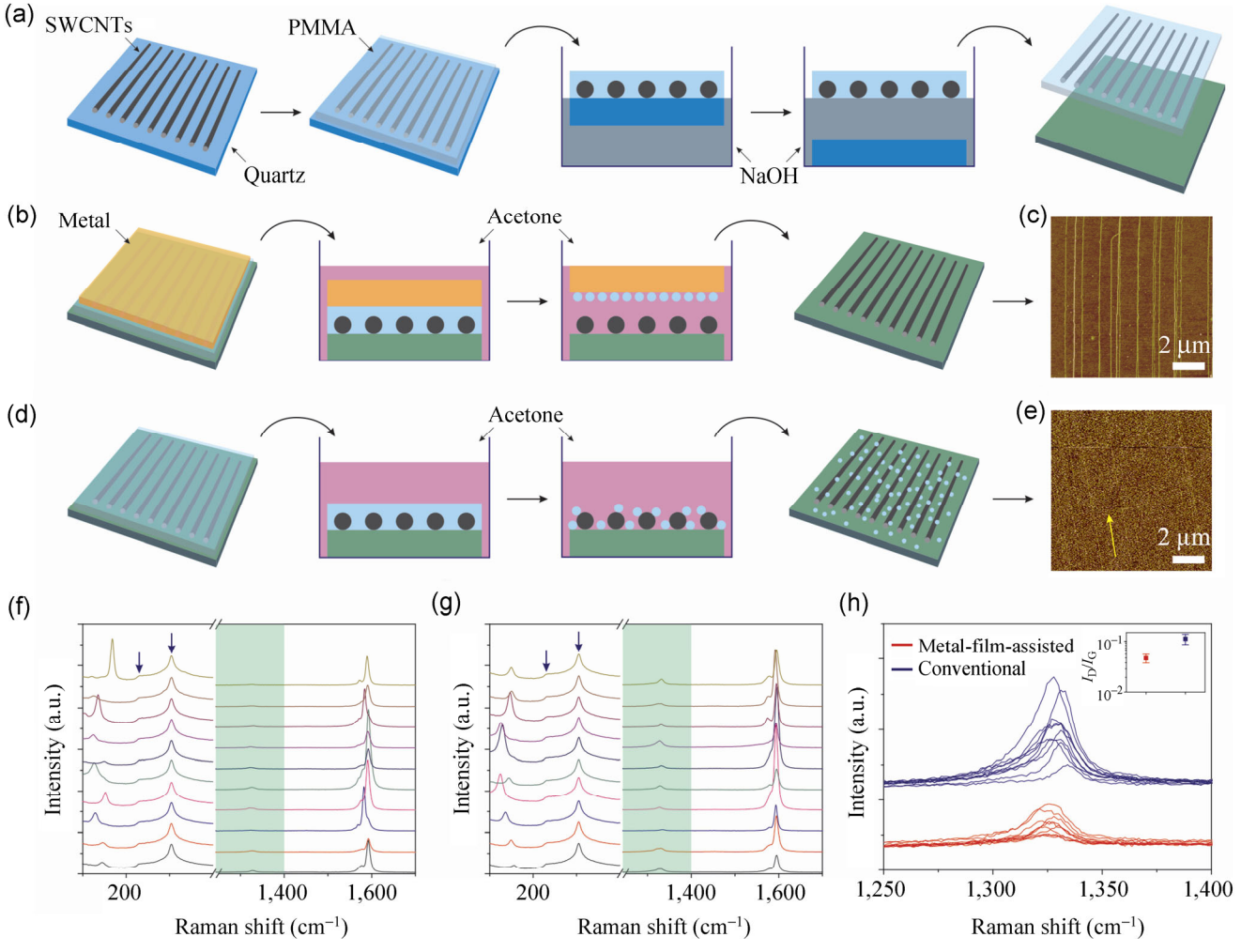

Figure 1 Flow charts of the metal-film-assisted transfer technique and the conventional method. (a) Schematic illustration of transferring PMMA/SWCNT film from quartz onto the target substrate. Aligned SWCNTs are grown on a quartz substrate by CVD, then spin-coated with a PMMA film as the sacrificial layer. Then the PMMA/SWCNT film is separated from the quartz substrate in NaOH aqueous solution (1 mol·L^{-1}, 100 ˚C), and attached onto a SiO_2/Si (thickness of the SiO_2: 300 nm) substrate. (b) The process of removing PMMA film by the metal-film-assisted transfer technique. After depositing a metal film on the PMMA surface, the metal-coated PMMA can be peeled off from the substrate in acetone within 5 min, leading to clean surfaces of SWCNTs and substrate. (c) The AFM image of the SWCNTs of (b). (d) The process of removing PMMA film by the conventional method. After dissolving in acetone for 30 min, an amount of PMMA residue still covers the substrate and the SWCNTs. (e) The AFM image of the SWCNTs of (d). The direction of the aligned SWCNTs is marked by the yellow arrow. Raman spectra of SWCNTs transferred by (f) metal-film-assisted transfer technique and (g) the conventional method. The spectra were obtained at 10 random spots with a 633 nm excitation laser. The Raman spectra of Si substrate are marked by the blue arrows. The D modes are highlighted by green area for a direct comparison between the two methods. (h) The high-magnification of the D modes in (f) and (g). The inset shows the average intensities of D mode normalized by G mode.

SWCNTs. From both the AFM and Raman results, it is evident that the metal-film-assisted method can effectively reduce the amount of PMMA residue.

The aforementioned metal-film-assisted technique was further utilized to transfer SWCNTs from quartz to the flexible substrate. The PMMA/SWCNT film peeled off from quartz was attached onto a polyethylene terephthalate (PET, thickness: 70 μm) substrate and a thin film of Ti (optimized thickness: 20 nm) was then deposited onto this specimen by electron-beam evaporation. The PET substrate was then immersed in acetone to dissolve the PMMA. Figure 2(a) presents the sample surface at different dissolving times from 30 to 400 s. With the increase of dissolving time, the

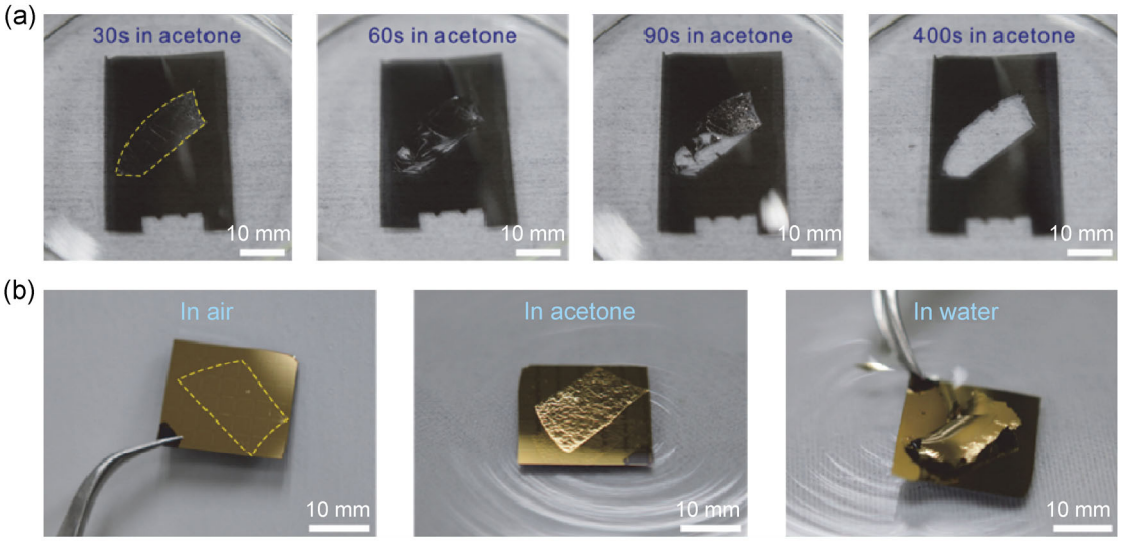

Figure 2 Photographs of the process of peeling off metal-coated PMMA film. (a) Photographs of Ti (20 nm)-coated PMMA/SWCNT film on a PET (thickness: 70 μm) substrate immersed in acetone for different times. The Ti/PMMA film is peeled off gradually in 400 s. (b) Photographs of Au (50 nm)-coated PMMA/SWCNT film on a SiO_2/Si (thickness of the SiO_2: 300 nm) substrate. The Au/PMMA film cannot be peeled off from substrate in acetone, but a multitude of wrinkles appear on the surface of Au/PMMA film. By inserting the substrate into water immediately after removal from acetone, the Au/PMMA film can be peeled off spontaneously. The Au/PMMA film is marked by yellow dashed line.

Ti/PMMA film was gradually peeled off. Subsequent scanning electron microscopy (SEM) characterization showed that the SWCNTs were perfectly left on the PET substrate, verifying the suitability of the metal-film-assisted transfer method for flexible substrates. The application of the low temperature metal-film-assisted transfer on flexible substrates is of great importance, since these flexible substrates will deform target or melt during high temperature annealing, which is the general method to further remove the PMMA residue. Therefore, the metal-film-assisted transfer is superior to the conventional method particularly for flexible target substrates.

Further experiments showed that a variety of metal films (Au, Cr, Al, and Ni) with appropriate thickness can replace the Ti film, and that all of them can help to reduce the PMMA residue on the transferred SWCNTs as effectively as the Ti film. It is interesting that we could even peel off the metal-coated PMMA film entirely if the deposited metal film is thick enough. As shown in Fig. 2(b), 50 nm Au was deposited onto a PMMA/SWCNT film on a SiO_2/Si (thickness of the SiO_2: 300 nm) substrate. In this case, the metal film could not be peeled off from the substrate in acetone spontaneously. However, a multitude of wrinkles appeared on the surface of the Au/PMMA film. The formation of these wrinkles can be attributed to the partial and uneven separation between Au/PMMA and the Si substrate induced by acetone dissolution and penetration. By soaking the substrate into water immediately after removal from acetone, the whole wrinkled film can be directly peeled off from the substrate, and the whole Au film becomes suspended on the water surface. The separation in water is caused by the surface tension of water. Note that a proper dissolution time in acetone is essential to make sure the SWCNTs are perfectly left on the target substrate without damaging them during the peeling-off process. In this case, we can also obtain clean transferred SWCNTs. Furthermore, this thick-film method can be applied to transfer nanostructures grown on substrates with relatively rough surfaces, such as graphene on copper foils, because on these relatively rough substrates, a thicker metal layer is required to form a continuous film.

3 Results and discussion

TEM was used to further show the advantages of the metal-film-assisted transfer technique. To make a comparison, SWCNTs were transferred from quartz onto home-made SiN_x TEM grids by the conventional transfer method and the metal-film-assisted transfer technique, as illustrated in Figs. 3(a) and 3(d) respectively. The TEM images show that the whole SWCNT was covered by PMMA residue after the acetone dissolving process by the conventional method (Figs. 3(b) and 3(c)). However, if we deposited a thin film of Ti (20 nm) onto the PMMA/SWCNT film before the grid was immersed in acetone, the TEM images present clean surfaces of the SWCNTs, as the amount of the PMMA residue is greatly reduced and only small pieces of PMMA residue are attached on the SWCNTs (Figs. 3(e) and 3(f)). The key role of the additional metal film was thus verified by the TEM characterization.

To further improve the cleanliness of transferred SWCNTs, it is rather effective to integrate the metal-film-assisted transfer technique together with high temperature annealing. Both of the aforementioned TEM samples were simultaneously annealed at 400 °C under the protection of Ar atmosphere for 2 h. This post-treatment can further decompose PMMA residue remaining after dissolution in acetone, as shown in Fig. 4. In the case of the conventional method (Figs. 4(a)–4(c)), some bulk residue is still present after the annealing process, which arises from the large amount of PMMA residue as shown in Fig. 3(b). In contrast, on the annealed sample made by the metal-film-assisted transfer method (Figs. 4(d)–4(f)), we cannot find any apparent residue on the entire suspended SWCNTs at low magnification (Fig. 4(d)), and the high magnification TEM images show the ultra-clean surfaces of SWCNTs (Figs. 4(e) and 4(f)). Since the metal-film-assisted transfer method has removed most of the PMMA (Figs. 3(e) and 3(f)), the tiny amount of PMMA residue remaining is easily decomposed completely at high temperature. Therefore, we can obtain ultra-clean SWCNTs by integrating the metal-film-assisted transfer technique and a post-annealing process.

On the basis of the phenomena observed during the dissolution procedure and the improved surface cleanliness verified by AFM and TEM characterization, we can propose a mechanism for the metal-film-assisted transfer. In the metal-film-assisted transfer, the top surface of the PMMA film is coated by a metal thin film deposited by electron-beam evaporation, while the bottom surface is attached to the target substrate after peeling from quartz. We speculate that the contact between the substrate and the PMMA is weaker than that between the metal film and the

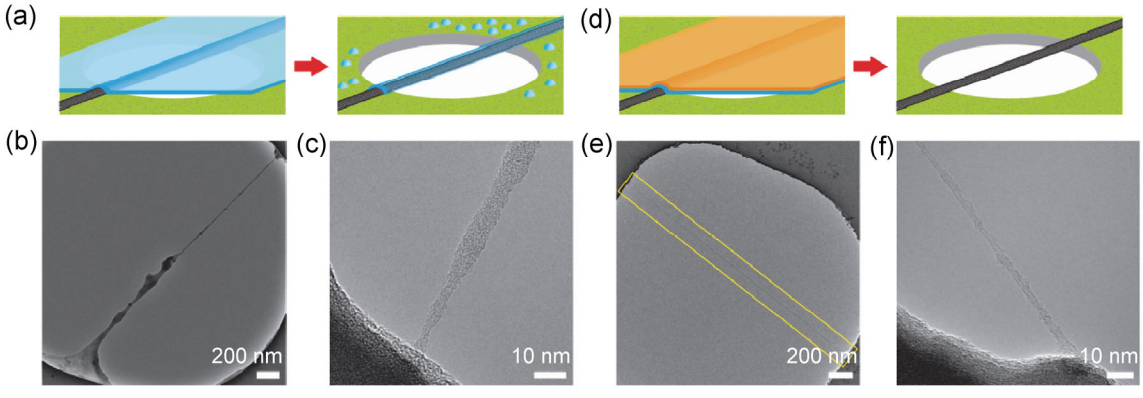

Figure 3 A comparison by TEM between the conventional method and the metal-film-assisted transfer technique. (a) A schematic illustration of transferring suspended SWCNTs by the conventional method. (b) Low-magnification and (c) high-magnification TEM images of SWCNTs transferred by the conventional method. (d) A schematic illustration of transferring suspended SWCNTs by the metal-film-assisted transfer technique. (e) Low-magnification and (f) high-magnification TEM images of SWCNTs transferred with the metal-film-assisted transfer technique. The SWCNTs are marked by the yellow rectangle in (e).

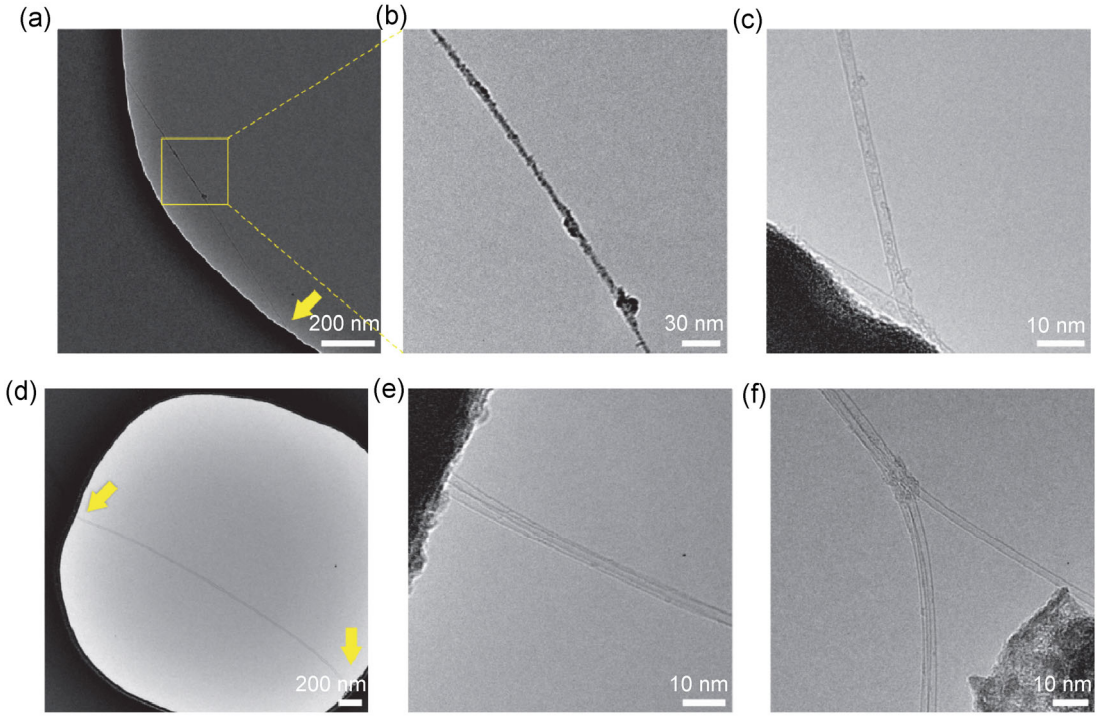

Figure 4 A comparison of TEM images of SWCNTs transferred by the conventional method and by the metal-film-assisted technique. The SWCNTs transferred by both methods are further annealed at 400 °C under the protection of Ar atmosphere for 2 h to further remove the PMMA residue. (a) A low-magnification TEM image of suspended SWCNTs transferred by conventional method, showing some bulk residue after the annealing process. (b) A medium-magnification TEM image of the yellow square area in (a). (c) A high-magnification TEM image of the area marked by the yellow arrow in (a). (d) A low-magnification TEM image of SWCNTs transferred by metal-film-assisted transfer technique, showing no apparent residue on the entire suspended SWCNTs. (e) and (f) High-magnification TEM images of areas marked by yellow arrows in (d).

PMMA. Therefore, when immersed in acetone, the dissolution of PMMA is initiated from the bottom surface of the PMMA. When the PMMA near the bottom surface is dissolved, the residual PMMA film can be peeled away from the substrate with the help of the metal film, leaving clean SWCNTs on the substrate. However, in the case of the conventional transfer method, the PMMA is dissolved from both the top surface and the bottom surface. The dissolution rate at the top surface is higher than that at the bottom surface since the top surface is fully exposed to acetone. Therefore, the whole PMMA film is dissolved into small pieces, and there is inevitably some residue left on the surfaces of the SWCNTs and the target substrate. In other words, the function of the additional metal film is actually to prevent the dissolution of the PMMA film from the top surface and to increase the mechanical strength of the metal-coated PMMA film; thus the PMMA film is removed in large blocks before the PMMA is fully broken into small pieces. Therefore, the transferred SWCNTs on the target substrate are clean with little residue. This mechanism is strongly confirmed by the different times needed to dissolve the PMMA. The metal-film-assisted method requires only about 5 min to peel off the PMMA, since only the PMMA near the bottom surface is dissolved. By contrast, in the conventional method, the dissolution of PMMA takes much longer (about 30 min), since the PMMA film must dissolve gradually from top to bottom to eventually uncover the SWCNTs. Because the mechanism above relies little on the type of polymer, we believe that this method would also work for other organic media used in transfer technology.

The clean and efficient technique we have developed makes it possible to fabricate more complex nanostructures via multiple transfers. In fact, multiple transfers have been widely used in the fabrication of nanodevices [17–22]. However, the PMMA residue

makes it difficult to realize a good contact between nanostructures from different layers, which hampers further applications of the transfer printing technique. Here we demonstrate that the metal-film-assisted transfer technique significantly outweighs the conventional method in multiple transfers. Figures 5(a)–5(c) show the AFM and SEM images of cross-stacked SWCNT networks on a Si substrate fabricated by transferring horizontally aligned SWCNTs twice. In the high-magnification SEM image (Fig. 5(c)), we can clearly see that some SWCNTs appear brighter where they make a crossing with other SWCNTs, which is a typical phenomenon in SEM when a semiconducting SWCNT contacts a metal, as is illustrated in our previous work [23, 24]. The contrast difference along a semiconducting SWCNT in the SEM image (Fig. 5(c)) reveals that a Schottky barrier is formed at the contact point where it contacts with a metallic SWCNT [25]. This is a strong evidence of a good contact between SWCNTs from different layers. In addition, SWCNT/graphene structures on a Si substrate were also fabricated by transferring SWCNTs from quartz and graphene from copper foils, successively. Note that a thicker metal film (100 nm in thickness) should be used to transfer graphene from copper foils, since the rough surface of copper foil leads to a rough PMMA film and further hinder the formation of a continuous metal film. Figures 5(d)–5(f) show an optical micrograph and SEM images of SWCNT/graphene stacked structures on a Si substrate. Similarly, some of the SWCNTs appear brighter near the contact point with graphene, which again proves the high-quality contact between nanostructures. Such high-quality contact can be used to verify the cleanliness of the metal-film-assisted transfer technique and suggests that this transfer technique will pave the way for fabricating high-quality nanodevices by multiple transfers.

4 Conclusions

We have developed a metal-film-assisted technique to realize ultra-clean transfer of SWCNTs from quartz

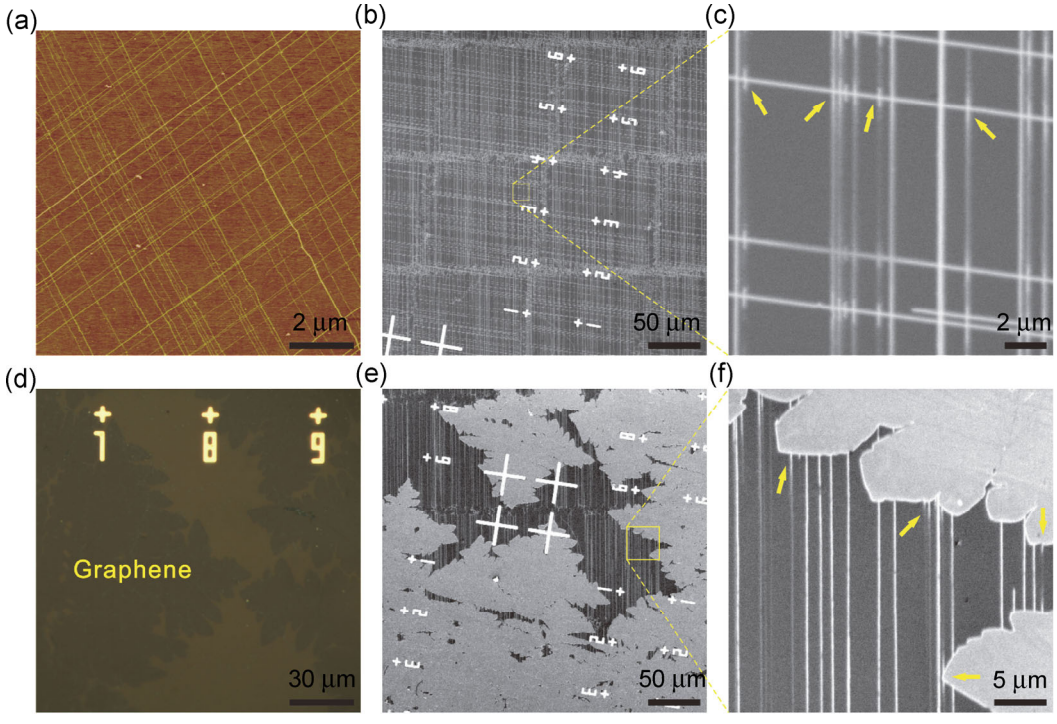

Figure 5 Fabrication of complex nanostructures via multiple transfers. (a) AFM and (b) and (c) SEM images of cross-stacked SWCNT networks on a SiO_2/Si (thickness of the SiO_2: 100 nm) substrate. Some of the SWCNTs appear brighter near the crossing with other SWCNTs (marked by the yellow arrows). (d) Optical microscopy and (e) and (f) SEM images of the SWCNT/graphene stacked structures on a SiO_2/Si (thickness of the SiO_2: 100 nm) substrate. Some of the SWCNTs appear brighter near the contact point with grapheme (marked by the yellow arrows).

to silicon substrates or even to flexible substrates. The surfaces of SWCNTs transferred by the metal-film-assisted method are much cleaner than those obtained by the conventional PMMA-mediated transfer, as shown by AFM and TEM results. By integrating the metal-film-assisted transfer with post-annealing, the small amount of remaining PMMA residue can be removed. This efficient and clean technique can be used to fabricate complex nanostructures by multiple transfers, such as cross-stacked SWCNT networks, and SWCNT/graphene stack structures. Since the clean surface ensure the good contact between nanostructures from different layers and between nanostructures and deposited metal electrodes, this metal-film-assisted transfer method should have widespread applications in nanomaterial-based electronics, especially in flexible electronics.

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