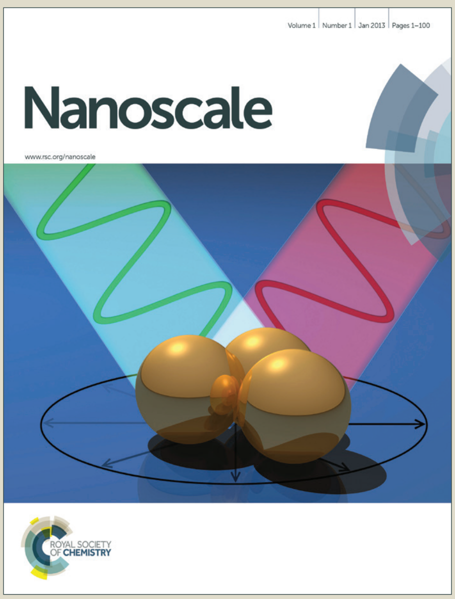
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Silica-gold bilayer-based transfer of focused ion beam-fabricated nanostructures

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Abstract: The demand for using nanostructures fabricated by focused ion beam (FIB) on delicate substrates or as building blocks for complex devices motivates the development of protocols that allow FIB-fabricated nanostructures to be transferred from the original substrate to the desired target. However, transfer of FIB-fabricated nanostructures is severely hindered by FIB-induced welding of structure and substrate. Here we present two (*ex* and *in situ*) transfer methods for FIB-fabricated nanostructures based on a silica-gold bilayer evaporated onto a bulk substrate. Utilizing the poor adhesion between silica and gold, the nanostructures can be mechanically separated from the bulk substrate. For the *ex situ* transfer, a spin-coated poly(methyl methacrylate) film is used to carry the nanostructures so that the bilayer can be etched away after being peeled off. For the *in situ* transfer, using a micro-manipulator inside the FIB machine, a cut-out piece of silica on which a nanostructure has been fabricated is peeled off from the bulk substrate and thus carries the nanostructure to a target substrate. We demonstrate the performance of both methods by transferring plasmonic nano-antennas fabricated from single-crystalline gold flakes by FIB milling to a silicon wafer and to a scanning probe tip.

Introduction

Focused ion beam (FIB) is a well-developed powerful tool for rapid nanostructure fabrication. However, substrate damage severely restricts its use on delicate substrates, such as semiconductors, magnetic materials, polymers, thin films, *etc*. For such substrates, either the lattice structures / constituting molecules are altered by the high-energy ions, or the surface of the substrate is deformed by the ion bombardment. A way to circumvent FIB damage is to perform structure patterning on a FIB-suited (i.e. robust and well-conductive) *source* substrate and transfer the resulting structures to a delicate *target* substrate.

As a matter of fact, the rapidly growing interest in constructing more complicated devices has already stimulated the development of various transfer techniques.[1–7] Particularly, Meitl *et al*.[1] reported an approach where polydimethylsiloxane (PDMS) stamps are used to transfer semiconductor structures and mica sheets *etc*., which are loaded to or released from the stamps by kinetic control of the adhesion between structures and PDMS. Jiao *et al*.[2] reported that a poly(methyl methacrylate) (PMMA) thin film is a good transfer mediator of nanostructures because it conforms and adheres well to nanostructures and can be easily removed. In this method the PMMA film is peeled off from the source substrate by soaking it in basic solution while the nanostructures remain embedded in it. Schneider *et al*.[3] demonstrated that cellulose acetate butyrate films can be separated from glass or SiO_2/Si substrates due to the water wedging effect and thus can be used to strip off graphene or metal structures from their substrates and transfer them to other substrates. Although these techniques proved to be successful for various applications, we found that they can hardly be applied to transfer FIB-fabricated nanostructures as detailed below.

Here, we present a transfer approach that is designed to work with FIB-fabricated nanostructures. To fit the requirements of both FIB fabrication (conductivity) and transfer (structure separation), the source substrate is a glass coverslip coated sequentially with a gold and SiO_2 layer (silica-gold bilayer). After FIB fabrication of nanostructures on top of this bilayer, a PMMA film is spin-coated onto the source substrate. The silica layer acts as a spacer layer between the nanostructures and the gold layer, while the gold layer, in addition to providing electric conductivity for FIB process, acts as a sacrificial layer for the lift-off of all the top layers from the coverslip. The nanostructures are fully released from the substrate by etching away the silica-gold bilayer. This approach has a 100% transfer efficiency and only requires very little amount of etchants. Furthermore, we also demonstrate that based on the silica-gold bilayer substrate, it is possible to perform *in situ* transfer of nanostructures inside a FIB machine. Taking advantage of various manipulation modules of the FIB machine, the *in situ* transfer may provide more fabrication capabilities with higher precision.

Results and discussion

Strong bonding between FIB-fabricated nanostructures and substrate

To demonstrate the difficulty of transferring FIB-fabricated nanostructures, we used FIB to fabricate an array of cross-shaped optical antennas[8,9] from a single-crystalline gold flake grown on a glass substrate,[10,11] and the conventional PMMA-mediated transfer method reported by Jiao *et al.*[2] was used to transfer the antennas to an indium tin oxide (ITO) coated glass coverslip. It turned out that only a few of the antennas were transferred successfully, while the others were lost either partly or completely (Figure 1). In fact, the lost parts were still staying on the source substrate (Figure S1 in Supplementary Information). These results suggest that FIB milling considerably enhances the bonding between the antenna and substrate such that the adhesion between PMMA and gold is no longer sufficient to detach the antenna from the substrate. Therefore, most previously reported transfer methods are not expected to work for FIB-fabricated nanostructures either. The low transfer yield shown in Figure 1 indicates that FIB-fabricated nanostructures need to be released from the substrate in a different manner.

Silica-gold bilayer-based transfer (*ex situ*)

Our new approach is depicted in Figure 2. A glass coverslip is coated with silica-gold bilayer and acts as the source substrate. Single-crystalline gold flakes are then deposited onto the source substrate and nanostructures are milled out of a gold flake by FIB (Figure 2a). Next, a PMMA film is spin-coated on the source substrate. Thus the nanostructures are embedded between the PMMA and silica layers. As the edges of the coverslip are usually covered by the PMMA film, a circular slit is scratched on the outer part of the substrate to make all layers accessible for further processing (Figure 2b). The sample is then inserted into ultrapure water slowly at an oblique angle. Because PMMA is hydrophobic and the adhesion between the gold layer and glass substrate is quite weak, the gold layer is separated from the coverslip by the surface tension force of water, which wedges in from the scratched slit (Figure 2c). The three layers (PMMA, silica, and gold) are peeled off in this way and float on the water surface (Figure 2d). A plastic sheet with a hole at center is used to pick up the floating membrane and the structures are positioned inside the hole. Once the plastic sheet (membrane holder) and the membrane have been dried in oven, they bond with each other firmly (Figure 2e). To locally remove the gold and silica layers, the structure is turned upside down and a small amount of gold etchant is dropped onto the gold layer at the structure position (Figure 2f), followed by silica layer etch (Figure 2g). Subsequently, the membrane is reversed again and placed on the target substrate. At this point it is possible to align the structures with the desired location on the substrate by moving the membrane holder. After the membrane within the hole adheres to the substrate, a circular cut is made along the edge of the hole, so that the holder can be removed (Figure 2h). As the last step, the PMMA layer is removed (Figure 2i).

To demonstrate the method we also used cross-antenna arrays because of their fine structural features, and the results are presented in Figure 3 (see also Figure S2 in Supplementary Information). All the antennas in the array were transferred to the SiO_2/Si substrate with all structural details maintained very well – only tiny differences between the structures before and after transfer are visible from the SEM images. This excellent result is ensured by the fact that the nanostructures are released by etching away the substrate instead of being detached from the substrate by the PMMA film. Once the nanostructures are separated from the substrate, the PMMA film is fully adequate to hold the structures and carry them to the target substrate without disturbing their original configurations.[2]

We would like to emphasize that the silica-gold bilayer plays an essential role in our approach. First, because the silica layer is thin enough, electrons and gallium ions are able to penetrate it and reach the gold layer. The substrate therefore exhibits sufficient electrical conductivity and thus avoids charging effects, which would harm both fabrication and imaging. Second, the gold layer separates from the glass coverslip very easily, leading to a simple and fast detachment process using only the surface tension of water. Once removed from the coverslip, the gold and silica layers can be etched away to completely release the nanostructures. Because it is only necessary to locally etch the gold and silica layers at the structure position and the thin layers are etched in the normal direction, very little amount (a few tens of microliters) of etchants are needed, and both etching steps are finished within one minute. The silica layer here mainly acts as a spacer layer between the structures and gold layer, yet the silica layer or both the gold and silica layers can also be kept and transferred to the target substrate if they are needed as a part of the desired device. In principle the silica-gold bilayer can also be replaced with other materials as long as they work for the transfer in similar ways, but silica and gold are still recommended since they are widely used in various technology fields and their processing techniques from high-quality coating to etching are well-developed and very popular.

There are a couple of reported approaches[12,13] where devices are fabricated on SiO_2/Si substrate and the SiO_2 layer is etched away to release the devices from substrate. They should also be valid for transferring FIB-fabricated nanostructures. However, for these approaches a large amount of SiO_2 etchant (HF) has to be used to immerse the whole sample, and the etch takes hours at high temperature due to the fact that the thin sacrificial layer has to be etched through from lateral direction with much lowered etch rate, which is known as a common problem in the epitaxial lift-off process.[14,15] Compared to these approaches, our approach is apparently user & environment-friendly. In addition, the source substrate in our approach is partially transparent, which allows optical inspection and characterization of the sample before the transfer.

On the right side of Figure 3, high-resolution SEM images of a single antenna before and after transfer to SiO_2/Si substrate are compared. In contrast to the smooth target substrate, the FIB-created roughness is evident on the source substrate, i.e., there are grooves on the silica layer and each antenna arm resides on a silica pedestal.

The roughness of the substrate may distort the scattering spectra of the nanostructures. It is well recognized, though, that such unwanted milling of substrate is hardly avoidable when fabricating precise structures for which the FIB patterns consist of many components. On one hand, over-milling is often needed to completely remove the material on top of the substrate, because the milling is usually inhomogeneous even for one single component of the pattern. On the other hand, sometimes the pattern design requires some components to more or less overlap, for example to eliminate the influence of stage drift or to compensate the inaccuracy of other parts, resulting in milling of the substrate at the pattern overlap area. Besides the milling of substrate, ion implantation may also cause modification or damage of the substrate, as mentioned above. Moreover, it is noticed that on the source substrate there are many small particles. These particles are actually redeposited gold originating from the last steps of milling. They are likely influencing the surface electrical resistance and contributing to optical scattering and absorption of the substrate. For instance, in the upper panel of Figure 1, no pronounced charging on the bare glass substrate is seen, which might hint at a lowered surface resistance around the nanostructure. On the target substrate, however, the redeposition particles are not seen anymore. This is because an extra processing step was implemented after the silica layer etch – a droplet of strongly diluted (1:100) gold etchant was put on the antenna area for 5 s, resulting in that the redeposition particles were etched away. According to our observation, the top/bottom surface (crystal plane {111})[10] of the gold flake has a quite high resistance against the etchant, while the lateral surfaces are etched much more rapidly (Figure S3 in Supplementary Information). Therefore, since the nanostructures are imbedded in PMMA and only the bottom surfaces are exposed to the etchant, such mild etch hardly harms the nanostructures (see Figure 3g), but is sufficient to remove the redeposition particles. As a conclusion, the well-defined and clean substrate in Figure 3e explicitly indicates the advantages resulting from the transfer of FIB-fabricated structures.

As a further demonstration, we transferred these cross-antennas again from the SiO_2/Si substrate to a glass coverslip using the same method as used for Figure 1. In contrast to the failed result in Figure 1, this time all the antennas were successfully transferred, as shown in Figure 3c. This result proves that the failure in Figure 1 is indeed caused by the FIB-induced strong bonding between the structure and substrate. In other words, the structures are likely welded to the substrate by FIB milling, such that the simple direct detaching methods are not valid anymore. It is also worth noting here that the antennas survived sample processing procedures such as oxygen plasma cleaning, SEM characterization and PMMA spin-coating, as shown above. This fact evidences that the bonding between the nanostructures and the target substrate is good enough for a wide range of applications.

In situ **transfer**

The idea of using a silica-gold bilayer substrate can be adapted to perform nanostructure transfer inside the vacuum chamber of the FIB machine, i.e. *in situ* transfer. For this purpose, the source substrate (Figure 4a) is slightly modified compared to the one used above. The glass coverslip is replaced by a silicon wafer, since a transparent substrate is not needed and silicon has a larger conductivity. In addition to the silica-gold bilayer, we introduce a thin chromium adhesion layer between the gold layer and silicon. The gold layer is thus firmly bonded to the silicon, while the silica layer can still be peeled off mechanically from the gold layer using micro-manipulators inside the vacuum chamber. Figure 4b-f illustrate a demonstration of this approach by transferring an optical antenna to a probe tip of scanning probe microscopy (SPM).[16–19] First, a small piece (6 × 8 μm) of the top layers (especially the gold flake and silica layer) was cut out using FIB milling. To mechanically strengthen the silica piece which was to be peeled off and moved later on, a rectangular window was milled out of the gold flake piece, followed by ion beam-induced deposition (IBID) of platinum on the exposed silica to form a solid frame. Unlike evaporated platinum on silica, the IBID platinum adheres on the silica layer very well (see Supplementary Information), making it possible to peel off the silica piece via the help of the platinum frame. After a nano-antenna was fabricated from the gold flake inside this frame, a micro-manipulator probe was approached to the platinum frame and connected to the frame by applying platinum deposition again (Figure 4b). The probe was then shifted slightly and slowly, and as a result the silica piece was detached from the gold layer (Figure 4c). Consequently, the small cut-out piece with the antenna sitting on the silica layer could be lifted and moved by the manipulator (Figure 4d). A SPM probe was now brought to the cut-out piece (Figure 4e). After being aligned to the apex of the SPM tip, the antenna and the adjacent silica piece was cut off by FIB and attached at the apex (Figure 4f). The final result of the transfer after the rest of the cut-out piece was moved away is shown in the inset of Figure 4f. Although the antenna was a bit misaligned due to the limited precision of the manipulator, there is no doubt that this issue can be fixed by flattening the apex to a platform of about 0.5 μm (e.g. using FIB milling). This experiment demonstrates the enormous potential of the *in situ* transfer approach for building sophisticated multi-element devices.

Experimental section

Instruments and materials

A FEI Helios NanoLab 600 Dual Beam system was used for the FIB fabrications, and a built-in Omniprobe manipulator was used to perform the *in situ* transfer. An e-beam resist (Allresist, AR-P 679.04) was used for spin-coating PMMA films. The typical thickness of the PMMA films is around 400 nm. For the silica-gold bilayer coated on glass coverslip, the SiO_2 and Au layer thicknesses are 60 and 50 nm, respectively. For the *in situ* transfer experiment, the SiO_2, Au and Cr layer thicknesses are 50, 30 and 5 nm, respectively. The thickness of the SiO_2 and Au layers are not very critical, but on one hand they should be sufficiently thick in order to be solid enough, on the other hand the SiO_2 layer should be properly thin to avoid charging. The tip of the SPM probe (Cantilever SNOM Sensor, WITec GmbH) used for the *in situ* transfer was a hollow pyramid of SiO_2 and coated with aluminum. The single-crystalline gold flakes were produced with the protocol reported in ref. 10. The gold etchant is a home-made solution of I_2 : KI : H_2O = 1 g : 4 g : 40 ml, and the SiO_2 etchant is a home-made mixture of 40% NH_4F : 48% HF = 50 : 1 (buffered oxide etch). A piece of overhead projector film was cut into the membrane holder with a square hole at center.

Transfer of nanostructures fabricated on glass coverslip to ITO-coated coverslip

To make the sample electrically conductive for FIB fabrication, the whole sample (including the gold flakes grown on the coverslip) was coated with a 10 nm thick gold film and the sample holder was in contact with the gold film. The same method and procedure as described in ref. 2 were used for the transfer. In particular, a PMMA film was spin-coated on the source substrate and then baked in oven at 170 °C for 2 hours. The sample was immersed horizontally in KOH aqueous solution (Sigma-Aldrich, 319376) at 50 °C. After 5 min, the sample was taken out and put into ultrapure water horizontally. As a result, the PMMA film was separated from the coverslip by the water surface tension, together with the gold film, gold flakes, and part of the nano-structures. The floating membrane was picked up by an ITO-coated coverslip and dried in oven at 100 °C.

Gold flake preparation on silica-gold bilayer substrate

Gold flakes grown on a glass coverslip were transferred to the substrate with the same process mentioned above. In the end, the PMMA film was dissolved in an e-beam resist remover (Allresist, AR 600-70) at 40 °C.

Transfer of nanostructures based on silica-gold bilayer

After the PMMA film was spin-coated on the silica-gold bilayer substrate, the sample was baked in oven at 150 °C for 30 min. The scratched slit on the layers was around 0.5 mm wide. For etching silica and gold layers and gold redeposition particles, the etchants were droped on the membrane (PMMA, silica, and gold layers) with a pipette, and after every etch, the membrane was rinsed with ultrapure water. When the membrane was placed on the SiO_2/Si substrate, a droplet of ultrapure water was used to form a thin spacer layer between the membrane and substrate. After the water dried up in ambient condition, the sample was heated in oven to 100 °C until it was completely dry. Thus the membrane within the hole of the membrane holder was fixed on the substrate. After the membrane holder was removed, the sample was put in oven again at 170 °C for 2 hours. The heating of the PMMA film at a temperature significantly higher than its glass transition temperature (around 105 °C) relaxes and flattens its lower surface that has conformed to the uneven source substrate, which ensures a good contact of the nanostructures to the target substrate. The PMMA film was finally dissolved with the vapor of the PMMA remover (Allresist, AR 600-70) at

~55 °C.[2] In the end, the sample was treated by O_2 plasma (HARRICK PLASMA, PDC-3XG) for 10 min to thoroughly remove the residual PMMA.

Conclusions

We have shown that a key issue of transferring FIB-fabricated nanostructures is how to effectively separate the nanostructures from the source substrate due to the strong bonding between the structures and substrate, which is probably caused by a FIB-induced welding effect. Based on a source substrate with pre-deposited silica-gold bilayer and by taking advantage of the weak adhesion between gold and silica, we have developed two (*ex* and *in situ*) transfer techniques which are applicable to FIB-fabricated nanostructures. Combined with FIB fabrication and versatile micro/nano-manipulation techniques, these approaches are promising for producing varieties of unique nanostructures for special or delicate substrates and constructing complex devices. Moreover, the strategy of using a silica-gold bilayer substrate can be applied to other nano-structuring techniques, for example electron-beam lithography, where an adhesion layer between structure and substrate is often used. The *in situ* transfer method is also not limited to FIB-fabricated nanostructures.

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Supplementary Information

Electronic Supplementary Information available: SEM images of the transfer experiments showing more details, time series of wet etch of gold flakes, explanation of the strong adhesion between the IBID-deposited Pt and the silica layer.

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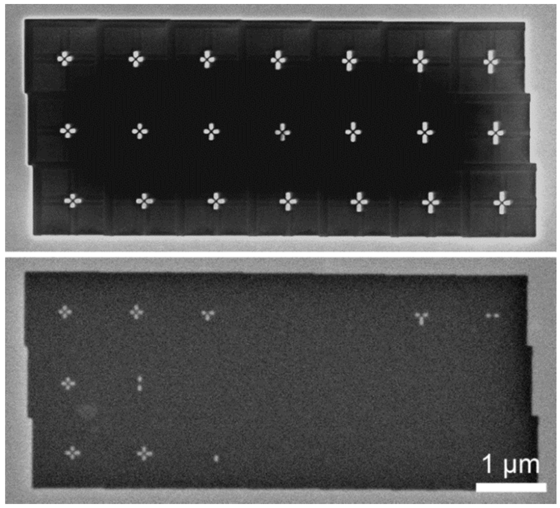

Figure 1. SEM images of a nano-antenna array after FIB milling (upper) and after being transferred to ITO-coated coverslip (lower, PMMA film still on top) using the method reported in ref. 2.

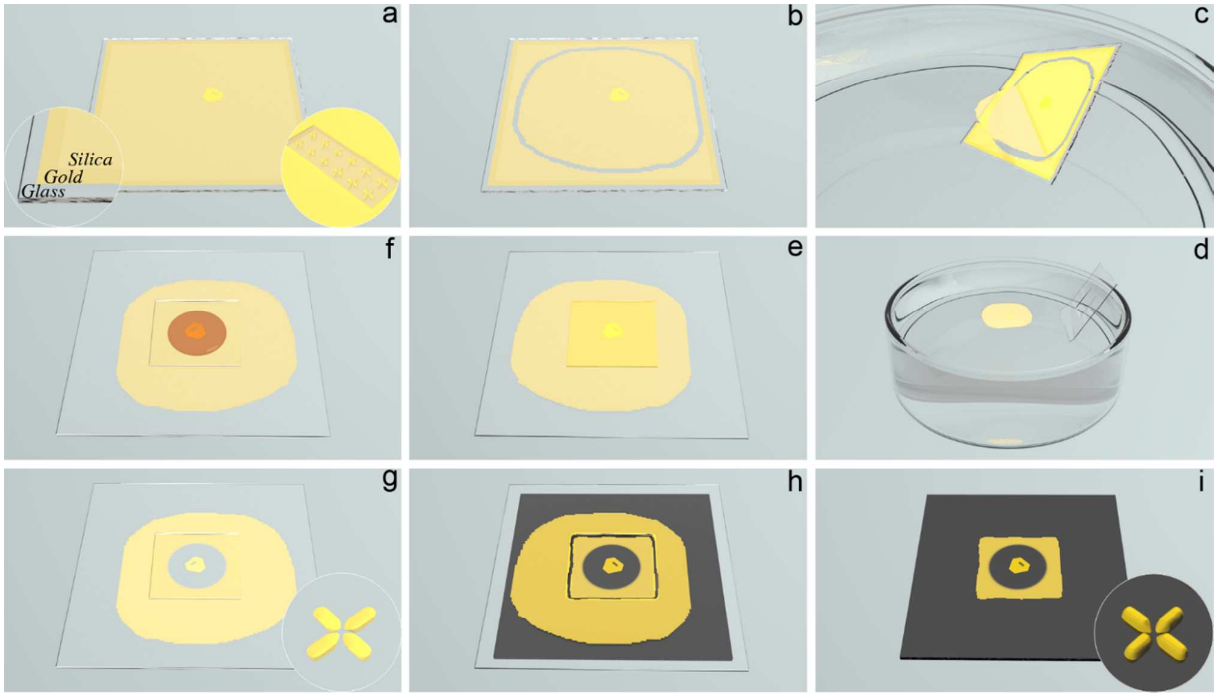

Figure 2. Schematic of the procedure of the silica-gold bilayer-based transfer approach. The objects are not in the same scale. a) A gold flake is deposited on a glass coverslip coated with Au and SiO_2 layers sequentially (left inset). An optical antenna array is then fabricated out of a gold flake using FIB milling (right inset). b) After the sample is spin-coated with a PMMA film, a circular slit is scratched through all the layers. c) The sample is dipped in water obliquely and slowly, such that the gold layer is separated from the coverslip. d) After the gold layer is separated completely, the PMMA-SiO_2-Au membrane floats on the water surface. A plastic sheet with a hole at center is used to pick up the membrane. e) The membrane is picked up with the gold flake positioned in the hole. The membrane is then fixed on the plastic sheet after being dried in oven. f) The membrane is placed with the gold layer up and a drop of gold etchant is put on the gold layer at the structure position. g) Both the gold and silica layers at the structure position are etched away. The inset shows the zoom of a reversed antenna (bottom up). h) The membrane is reversed again (PMMA film up) and placed on the target substrate, with the structures aligned to the desired position on the substrate. The membrane within the hole of the plastic sheet is cut out after it adheres on the substrate. i) The plastic sheet is removed and in the end the PMMA film is removed. The inset shows the zoom of an antenna on the new substrate.

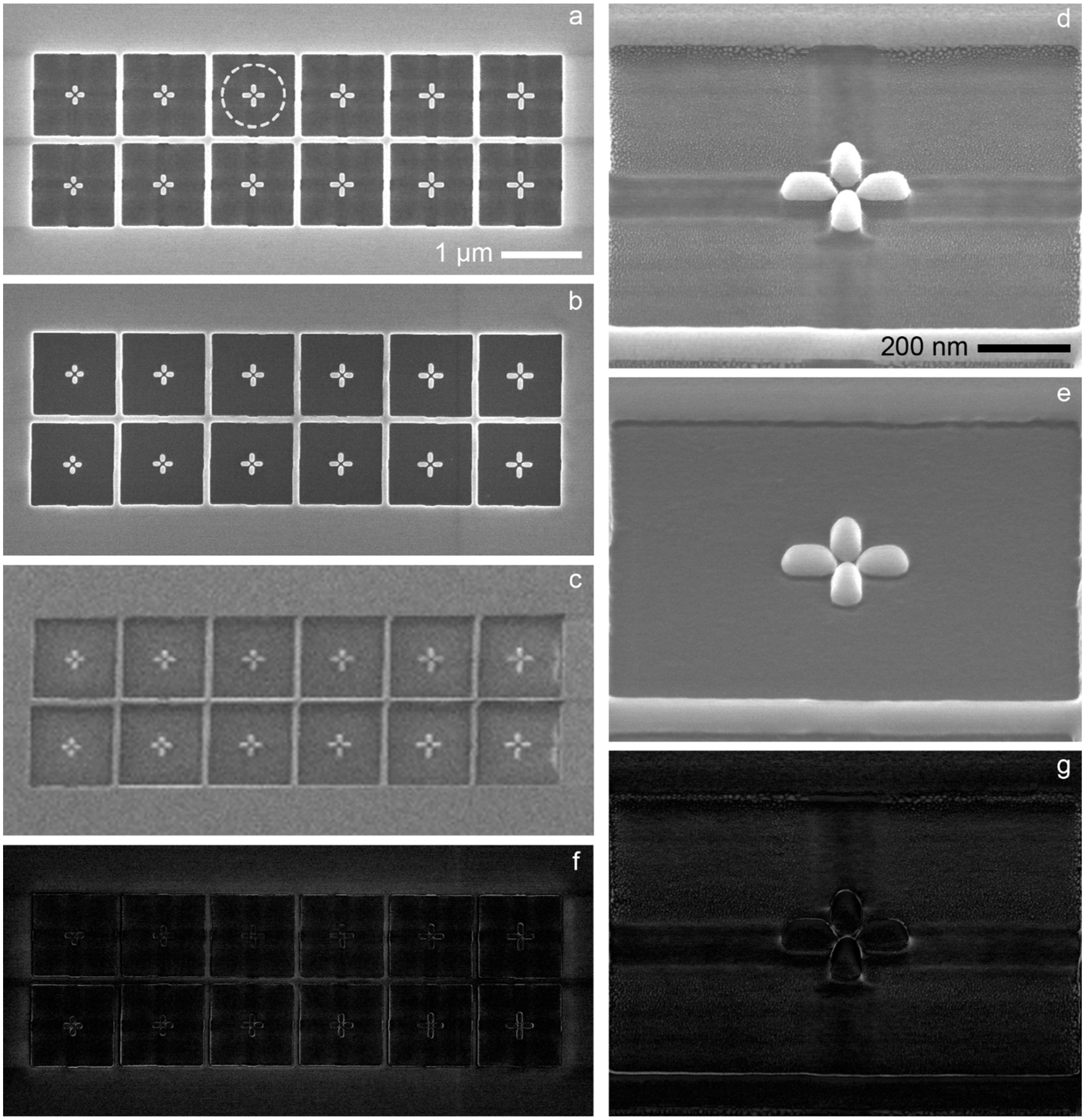

Figure 3. SEM images of a nano-antenna array after FIB milling (a), after being transferred to a SiO_2/Si substrate using the silica-gold bilayer approach (b, PMMA removed), and after being transferred again from the SiO_2/Si substrate to a glass coverslip using the same method as used for Figure 1 (c, PMMA removed, the bad image quality is due to the charging of substrate). d) and e) Zoom to the antenna in the dashed circle in (a) in 52° tilted view after FIB milling (d) and after transfer to the SiO_2/Si substrate (e, PMMA removed). f) Difference overlay of panels (a) and (b). The contrast of the two images were adjusted to be roughly the same and the absolute value of the difference (pixel by pixel) is shown. As a result the difference of the structures before and after transfer appears bright in a dark background. The two images are aligned with respect to the antenna in the dashed circle. The slightly larger mismatch for the structures far away from that antenna is most likely due to the drift of the electron beam or sample during the slow scan of the high resolution images. g) Same as (f) but for panels (d) and (e).

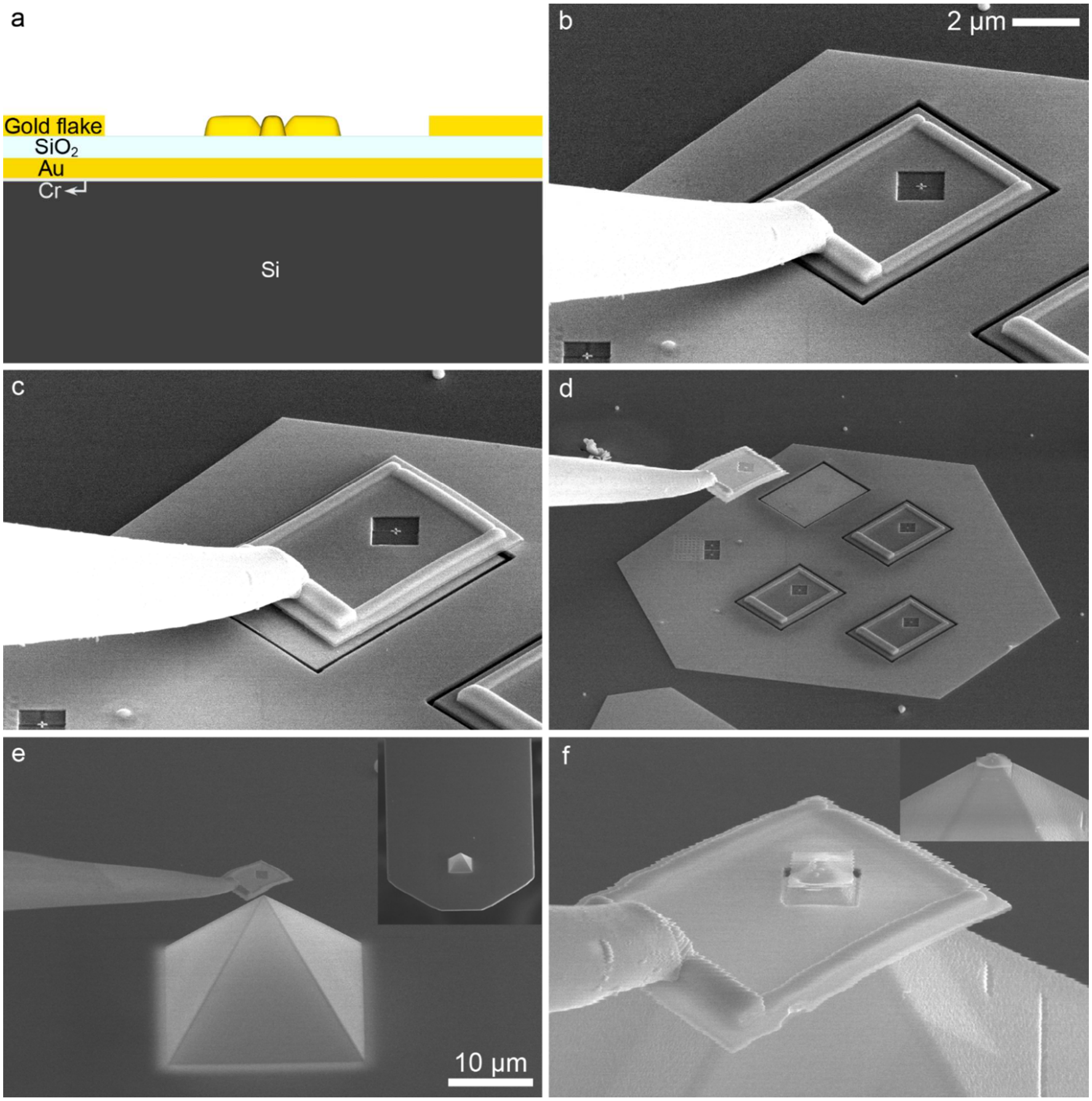

Figure 4. Silica-gold bilayer-based *in situ* transfer approach. a) Schematic of the substrate with a nano-antenna made from gold flake. b-f) SEM pictures of the procedure of transferring a nano-antenna to the tip of a SPM probe: the tip of a micro-manipulator probe contacts the thicker bar of the Pt frame and they are welded together by Pt deposition (b); the probe is shifted slightly such that the isolated piece is peeled off from the gold layer (c); the piece can then be lifted up (d) and the pyramid tip of a SPM probe is brought to the cut-out piece (e, the inset shows the cantilever of the probe); the antenna is aligned to the apex of the SPM tip by the manipulator, and the silica piece is cut off with FIB (f). The inset of (f) shows the final result where the residual part of the cut-out piece has been moved away. No extra gluing was performed here. All SEM images are in 52° tilted view.