Supplementary Information

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

Xiaofei Wu,¹ Peter Geisler, Enno Krauss, Ren é Kullock, and Bert Hecht*

Nano-Optics and Biophotonics Group, Experimentelle Physik 5, Universität

Würzburg, Am Hubland, 97074 Würzburg, Germany

*E-mail: hecht@physik.uni-wuerzburg.de

¹Present Address: Experimentalphysik III, Universität Bayreuth, Universitätsstraße 30,

95447 Bayreuth, Germany

• Additional results of the PMMA-mediated transfer method reported by Jiao *et al.*



Figure S1. Transfer of FIB-fabricated nanostructures using the method reported in ref. S1. a) SEM overview of the gold flake and the nano-antenna arrays after FIB milling (left) and after being transferred to ITO-coated coverslip (right, PMMA film still on top). The lower array is the one shown in Figure 1. b) SEM image of the area on the source substrate where the antenna arrays were located (dashed rectangle in (a)) after the transfer process.

It is seen that although the gold flakes, small gold particle and the 10 nm gold coating were all transferred successfully, most of the nano-antennas are still staying on the source substrate. This is strong evidence for the enhanced bonding of nanostructures to the substrate due to FIB milling. The bad quality of the SEM image in Figure S1b is due to charging of the substrate now being a bare glass coverslip.

• Additional results of the silica-gold bilayer based *ex situ* transfer method



Figure S2. SEM overview of the gold flake and the antenna arrays after FIB milling (left) and after being transferred to a SiO_2/Si substrate (right, PMMA removed) using the silica-gold bilayer approach. The upper array is the one shown in Figure 3.

The nice transfer result shown in Figure S2 demonstrates the remarkable performance of the silica-gold bilayer-based transfer approach.

• Gold flake etching experiment



Figure S3. Etching of gold flakes as a function of time. Etchant I_2 : KI : $H_2O = 1$ g : 4 g : 40 ml. The flakes are around 50 nm thick. Scale bar: 50 μ m.

Gold flakes were grown on the glass coverslip, and there was no gap between the flakes and the coverslip. It is seen from Figure S3 that the etch is much faster in lateral direction than in normal direction, and the top surface (crystal plane of $\{111\}$) has a significantly high resistance against the etch. The etch rate in normal direction is estimated to be < 2 nm/s. Therefore, for the gold redeposition particle etch in Figure 3, the 5 s etch with 100 times diluted etchant had almost no influence to the nanostructures, since only the bottom (also $\{111\}$ plane) of the structures were exposed to the etchant. It is worth to point out that the gold flake etch behaves just like the inverse of gold flake growth. In the latter case, the lateral size of a flake increases much faster than the thickness.

• Strong adhesion between the IBID-deposited Pt and the silica layer

According to our observation, the adhesion between the IBID platinum and silica layer is actually very strong, e.g. a Pt bar deposited on a small area is sufficient for peeling off a silica piece with a much larger area (more than 10 times larger) from the gold layer. This guarantees the Pt frame can peel off the silica piece from the gold layer, although the contact area of the Pt frame and silica is much smaller than that of the silica and gold.

We propose that this is attributed to three possible reasons. First, the mechanisms of the Pt deposition on silica and silica deposition on gold are different. The former is an ion beam-assisted chemical vapour deposition, while the latter is just a physical evaporation deposition. For the former case, a chemical gas precursor is injected to the substrate and absorbed on its surface. The precursor on the surface is then decomposed by the ions and more importantly by secondary particles (electrons, atoms and ions) emitted from the FIB scanned substrate and thus some of the decomposition products are deposited on the substrate.^{S2} In this sense the substrate material plays an important role in the chemical decomposition reaction during IBID. This could result in an extraordinary adhesion between the deposition and the substrate, as it is known that the reason why Cr and Ti adhere on SiO_2 very well is that they react with SiO_2 . Additionally, the deposited material does not consist of pure Pt, instead it is an amorphous compound including pronounced proportions of C and Ga in addition to Pt (precursor Methylcyclopentadienyl(trimethyl)platinum(IV), C9H16Pt).^{S3} Moreover, the property of the silica where the Pt is deposited is also modified due to FIB milling to open the gold flake window, i.e. Ga ions and maybe also some Au atoms are implanted into the silica. All these aspects should be able to explain the much stronger adhesion than that between the silica and gold layers.

References:

- [S1] L. Jiao, B. Fan, X. Xian, Z. Wu, J. Zhang, and Z. Liu, J. Am. Chem. Soc. 130, 12612 (2008).
- [S2] P. Chen, H. W. M. Salemink, and P. F. A. Alkemade, J. Vac. Sci. Technol. B 27, 2718 (2009); P. Chen, "Three-dimensional nanostructures fabricated by ionbeam-induced deposition", Ph. D. Thesis, Delft University of Technology, 2010.
- [S3] T. Tao, J.S. Ro, J. Melngailis, Z. Xue, and H. D. Kaesz, J. Vac. Sci. Technol. B 8, 1826 (1990).