Lift-off Free Nanofabrication of Suspended Plasmonic Nanohole Arrays To Overcome Mass Transport Limitations in Bio-sensors

Ahmet A. Yanik[1,2], Min Huang[1,2], Alp Artar[1,2], Tsung-Yao Chang[3], and Hatice Altug[1,2,*]

[1]Department of Electrical and Computer Engineering, [2] Photonics Center, Boston University, Boston, MA 02215
[3]Department of Electrical and Computer Engineering, M.I.T, Cambridge, MA 02139
altug@bu.edu

Introduction

Surface plasmonic devices based on nano-apertures have taken much interest recently due to their potential for biosensing, photovoltaics and information technology applications [1]. However, fabrication of surface plasmonic devices often requires focused ion beam (FIB) or negative resist electron beam lithography (EBL). FIB is an operationally slow technique which requires expensive infrastructure. Method intrinsically causes damaging of the surface and tapering of the structures. EBL tools are easier to operate and more commonly available. On the other hand, fabrication of plasmonic devices using EBL involves multiple resist layers and metal lift-off processes which limits the aspect ratio (width/depth) and the resolution of the features. In this article, we introduce a lift-off free fabrication technique based on single layer positive resist EBL and reactive ion etching (RIE). The simplicity of this fabrication technique allows us to fabricate nanostructures with extremely high yield/reproducibility and minimal surface roughness.

Results and Discussion

Application of the technique is demonstrated in fabrication of suspended plasmonic nanohole hybrid biosensors merging nanoplasmonics and nanofluidics. In conventional biosensing approaches, the analytes simply stream pass over the sensing surface, and the analyte transport towards the sensor is only through the diffusion processes. Accordingly, sensitivity of the surface biosensors are often controlled by the analyte delivery rate to the sensing surface instead of their intrinsic detection capabilities. At low concentrations, this limitation, commonly known as mass transport limitation, causes impractically long detection times [2]. Our hybrid detection platform enables targeted analyte delivery to the sensing surface to overcome the mass transportation limitations [3]. Here, the subwavelength size nanoholes act as nanofluidic channels connecting the microfluidic chambers on both sides of the sensors as well as enabling resonant light transmission due to the extraordinary light transmission (EOT) effect.

Fabrication technique based on positive resist electron beam lithography (EBL) and reactive ion etching (RIE) is summarized in Fig.1a. Initially nanohole pattern is defined in the positive e-beam resist, and then the nanohole pattern is transferred from e-beam resist to SiN membrane with RIE. After plasma cleaning of the surface a directional metal deposition processes is used to deposit Ti (5 nm) and Au (125 nm) metal layers defining the suspended plasmonic sensors with nanohole openings. Deposition process is extremely reliable; large areas of metallic nanohole arrays are repeatedly obtained without clogging the openings with extremely high yield/reproducibility and minimal surface roughness [3].

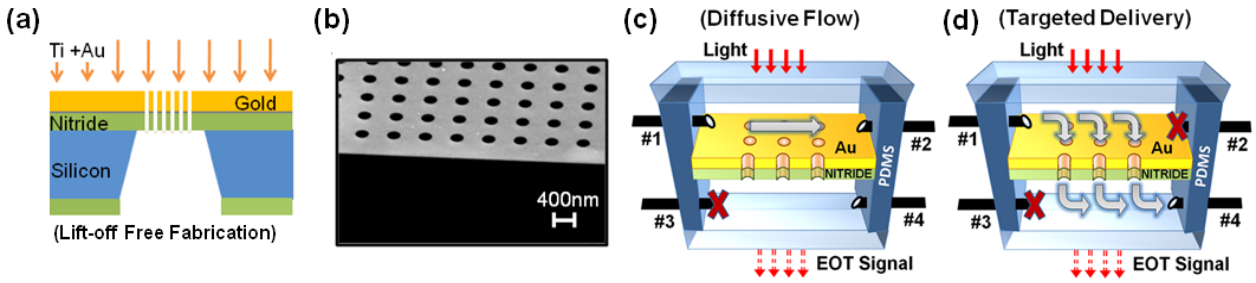

Fig. 1 *(a) Suspended plasmonic nanohole arrays are fabricated using a single layer lithography process and metal deposition. (b) Scanning electron images of the nanoholes are shown. multilayered microfluidic scheme allows 3-D control of the convective flow enabling (c) diffusive and (d) targeted delivery of the analytes to sensing surface.*

Biosensors are mounted in a multilayered microfluidic channel system based on poly(dimethylsiloxane). As illustrated in Figure 1(c-d), this multi-inlet/outlet fluidic platform allows us to actively control the fluidic flow in three dimensions through the plasmonic nanohole openings. Convective flow over different surfaces of the plasmonic sensor is realized by running the solutions in between input→output lines on the same side, such as 1→2/3→4 (Fig. 1(c)). In targetted delivery scheme, the convective flow is steered perpendicularly towards the plasmonic sensing surface by allowing the flow only through one inlet/outlet on either side of the plasmonic sensor (Fig. 1(d)).

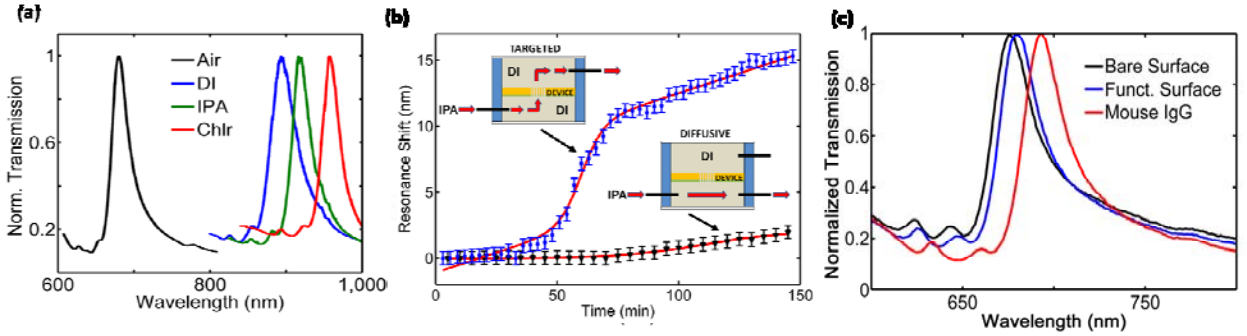

Fig. 2 (a) Bulk refractive index sensitivity of the plasmonic nanohole arrays are obtained in different solutions. (b) Efficiencies of the passive and targeted delivery of the analytes are compared. (c) Detection of biomolecules is demonstrated using mouse IgG antibodies.

Quality of the fabricated plasmonic nanohole arrays is reflected as spectrally narrow resonances and large refractive index sensitivities of $\Delta\lambda/\Delta n = 630nm/RIU$ (Fig. 2(a). Time dependent spectral measurements are performed to compare the efficiency of the analyte delivery for the diffusive and the targeted schemes. To quantify the analyte transport efficiency of the both delivery schemes, a lower viscosity analyte solution (IPA) with higher refractive index is introduced from the bottom inlet. The plasmonic sensor responses only to the refractive index change due to the perpendicularly diffused or actively delivered IPA solution depending on the implemented flow scheme. This way, a good quantitative measure of the transport limit is created in the perfect collection case. As shown in Fig. 2b in real time measurements, the directed delivery results in a much more efficient analyte delivery to the biosensor area. Experimentally observed resonance shifts are fitted to a sigmoid function of form $A_b + (A_t - A_b)/\left(1 + e^{-k(t-t_0)}\right)$ which is superposed to a background with $C_t(t-t_0) + C_0$ due to increasing refractive index of the bulk medium in the top channels as IPA concentration increases. The mass transport rate constants are obtained as $k_{diff}=0.0158\ min^{-1}$ and $k_{targ}=0.2193\ min^{-1}$ for the diffusive and targeted transport schemes, respectively. This corresponds to more than 14-fold improvement in rate constants which is crucial for enhancing the performances in immunoassay based applications. Biosensors are also tested in detection of mouse-IgG antibodies as shown in Fig. 2(c). Initially, functionalization of the gold surface causes a red shifting of 4nm of the plasmonic resonances with respect to bare surface. Capturing of the mouse-IgG antibodies at a concentration of $n_{mouse\text{-}IgG}=0.5mg/ml$ resulted in additional red shifting of $\Delta\lambda = 14\ nm$.

Conclusions

We have introduced a lift-off free nanofabrication scheme which can find wide range of applications in nanoplasmonics field by eliminating the need for FIB lithography and lift-off processes in EBL. This technique is demonstrated in development of a hybrid biosensing system merging nanoplasmonics and nanofluidics in a single platform enabling targeted delivery of analytes to the sensor surface. We also introduced a multilayered micro/nanofluidics scheme allowing three dimensional control of the analyte flow and direct steering of the convective stream to sensor surface to overcome mass transport limitations.

References

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