ARRAYS OF METALLIC NANOPILLARS IN HOLES FOR PLASMONIC DEVICES

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ABSTRACT

We have developed a well-ordered metallic array of nanopillars in nanoholes fabricated simply by magnetron sputtering into a plastic nanohole array over a large area. The pillar height and pillar-hole gap were controllable to nanometer scales through the sputtering time and initial nanohole design, constructed by a nanoimprint technique with high reproducibility. Our new fabrication method, without any resist, etching, or lift-off process, is simpler than previous methods [1]. By using this substrate, we observed a new plasmon resonance dip, related to the nanogaps in reflection spectra from the nanopillar-hole array. This unique fabrication method will be utilized to study highly sensitive plasmonic devices and surface enhanced Raman scattering (SERS) active substrates.

KEYWORDS: Nanopillar, SERS, Nanohole, plasmonic device

INTRODUCTION

Periodic metallic nanostructures have attracted much attention for their interesting optical properties and their potential use in biosensors. Various fabrication method have been investigated to realize simple yet accurate nanostructures over large area such as nanosphere lithography, colloidal lithography and nanoimprint technique. Among them, nanoimprint technique is particularly recognized as a potentially low-cost solution for fabricating nanoscale structures over large area. In previous microTAS proceedings, we discussed the surface plasmon resonance properties of gold nanohole substrates and their application to bioanalytical measurements [2, 3]. Alhough accurate gold nanostructures for actual sensing devices were realized reproducibly, we need to farther improve their sensitivity for low-concentration biomolecular measurements by making accurate nanogap structures over a large area with a simple and reproducible technique. Recently, Oh et al. produced nanometric gaps fabricated by an effective process of atomic layer deposition and selective etching [1]. It is a very important step that they realized metallic nanogaps which was difficult to construct by using electron beam (EB) lithography or focused ion beam (FIB) technique. However this process is required multiple steps and to precisely control the deposition time and selective etching condition.

Here we report the development of gold nanopillar-hole pair structures solely by combining a magnetron sputtering method and UV nanoimprint technique. The height of the nanopillars and the gap between pillar and hole can be controlled by tuning the hole depth and sputtering time. Reflection spectra from the substrates show that the characteristic of plasmon resonance dip can be attributed to the generation of a hotspot between the pillar-hole pairs.

EXPERIMENTAL

Figure 1 shows a schematic illustration of process that the metallic nanopillar-hole pair structures were fabricated. The polymer nanohole structures were replicated from glassy carbon (GC) mold after dispensing oligomer and exposing it to UV light. After rinsing replicated substrates with ethanol and water carefully, gold was deposited by RF sputtering with rotating the tray at 24 r.p.m. The substrates were set parallel to the gold target with a distance of 85 mm. The thickness of gold was controlled with increasing sputtering time. The sputtering was operated at 0.5 Pa with 10 s.c.c.m Argon atmosphere and the power was set to 100 W.

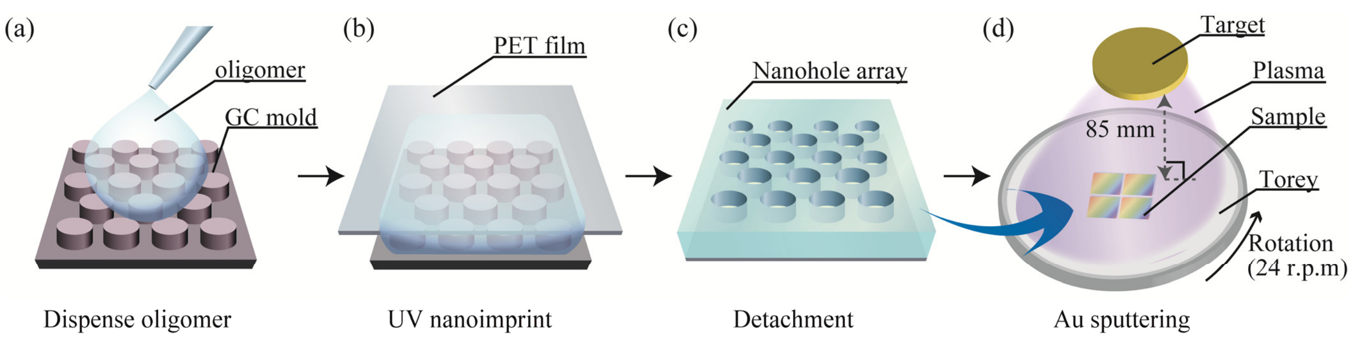

Figure 1: Schematic illustration of nanopillar-hole pair structures.

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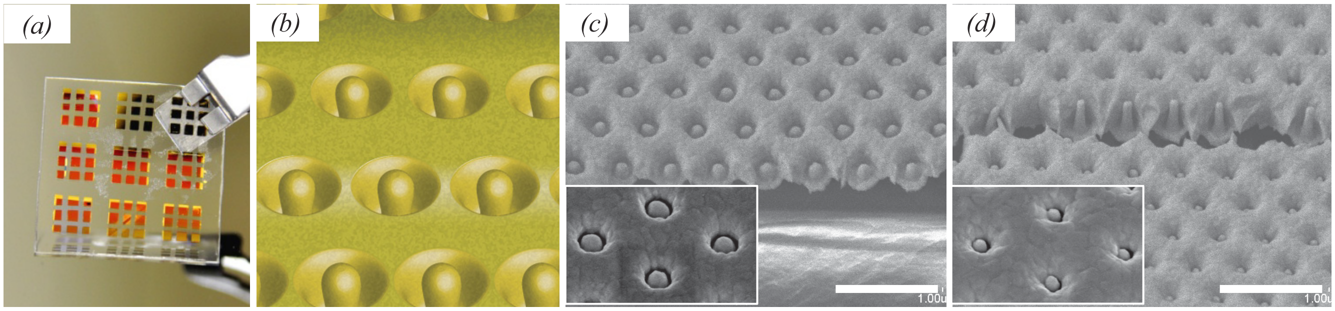

Figure 2: (a) Photograph of gold nanopillar / nanohole array chip. (b) Schematic illustration of gold nanopillar-hole array obtained solely by using sputtering on a nanohole array substrate fabricated with the UV nanoimprint method. (c), (d) Cross sectional SEM images of the substrates when 200 nm and 400 nm gold layers were sputtered on the nanohole array with a depth of 100 nm, a diameter of 300 nm and a periodicity of 500 nm. (Inset) Magnified images of each substrate at a granted angle. Allscale bars are 1 µm.

RESULTS AND DISCUSSION

Fig. 2(a) shows a photo of a plasmonic array substrate. Nanostructures were manufactured over a large area, as illustrated in Fig. 1(b). Almost no defect structures can be obtained with mm^2 scale. Figure 1(c) and (d) show SEM images of pillars with heights of 150 and 350 nm. Each gold nanopillar stands alone surrounded by a nanohole due to selective gold deposition at the bottom center of the nanohole and top of the substrate. Interestingly these structures cannot be fabricated when the hole depth was thinner or deeper than 100 nm such as 50 nm and 150 nm. This indicates that these nanopillar / nanohole array structures are obtained when there is a particular hole diameter / depth ratio. Also we already observed that we can manufacture these structures not only by sputtering gold but also silver and copper, allowing wider applications. Especially Ag nanopillar/nanohole structures are more suitable for SERS active substrates for its high sensitivity. Fig. 3 shows cross sectional SEM images and corresponding schematic illustrations for gold nanopllars in nanoholes when the gold was puttered with a thickness of 0, 100, 200, 300 and 400 nm, respectively. Figure 4(a-c) shows a dependency on gold film thickness of the pillar height, hole and pillar diameters and the gap. Pillar height increased linearly with the sputtered gold thickness. The pillar-hole gap decreased to below 20 nm. These properties were observed by SEM images in Fig. 3. When the gold was sputtered about 400 nm, the aspect ratio of pillar height / diameter becomes around four with standing alone. These nanostructures are difficult to fabricate with EB lithography. Reflection spectra were obtained from our substrate with the pillar height of 150 nm. We also collected reflection spectra from a gold nanohole array made by using vapour deposition [3]. The nanopillar-hole substrates showed two distinctive dips at wavelengths of 600 nm (dip-1:blue arrow) and 750 nm (dip-2:red arrow). Dip-1 was also observed in reflection spectra from the gold nanohole array, which was attributed to a propagating surface plasmon resonance [3]. Thus dip-2 shown in Fig. 5(a) might occur due to plasmon coupling between the gold pillar and the hole. This would explain the higher sensitivity of dip-2. We will apply these substrates as SERS active substrates for measuring bimolecular interactions.

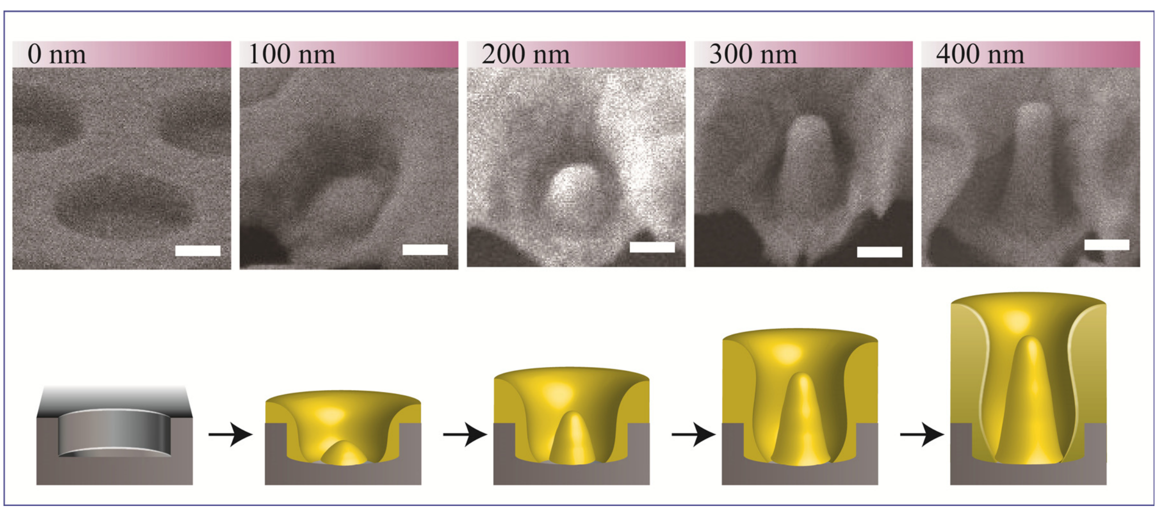

Figure 3: Cross sectional SEM images and corresponding schematic illustrations for gold nanopillars in nanohole when the gold was sputtered with a thickness of 0 nm, 100 nm, 200 nm, 300 nm and 400 nm on the nanohole array substrate. All scale bars are 100 nm.

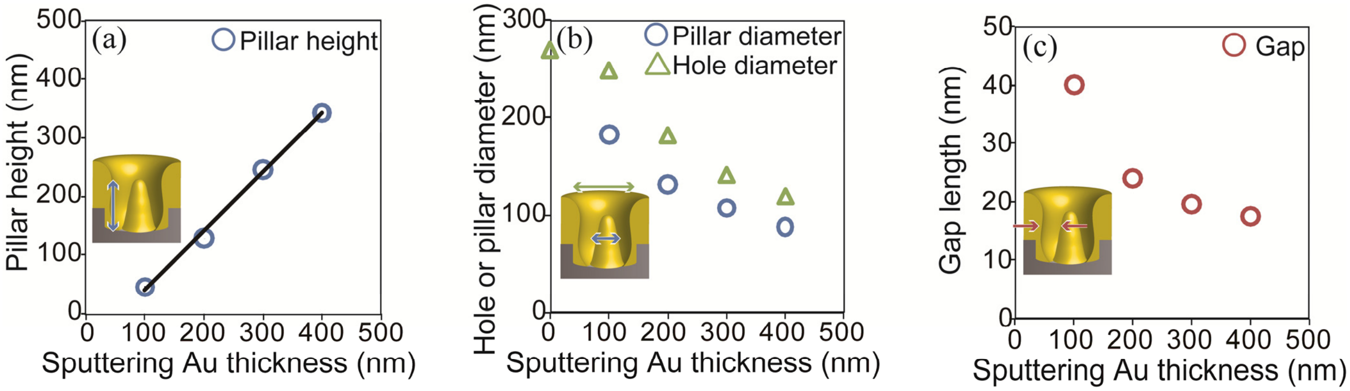

Figure 4: Dependence of (a) nanopillar height, (b) pillar and hole diameter and (c) gap distance as a function of the sputtering Au thickness.

CONCLUSTION

In this study, we fabricated gold nanopillar array embedded in nanohole for plasmonic studies by combining a magnetron sputtering method and UV nanoimprint technique. The height of the nanopillars and the gap between pillar and hole can be controlled by tuning the hole depth and sputtering time. Reflection spectra from the substrates show that the characteristic of plasmon resonance dip can be attributed to the generation of a hotspot between the pillar and hole pairs. These structures will be applied for SERS measurement.

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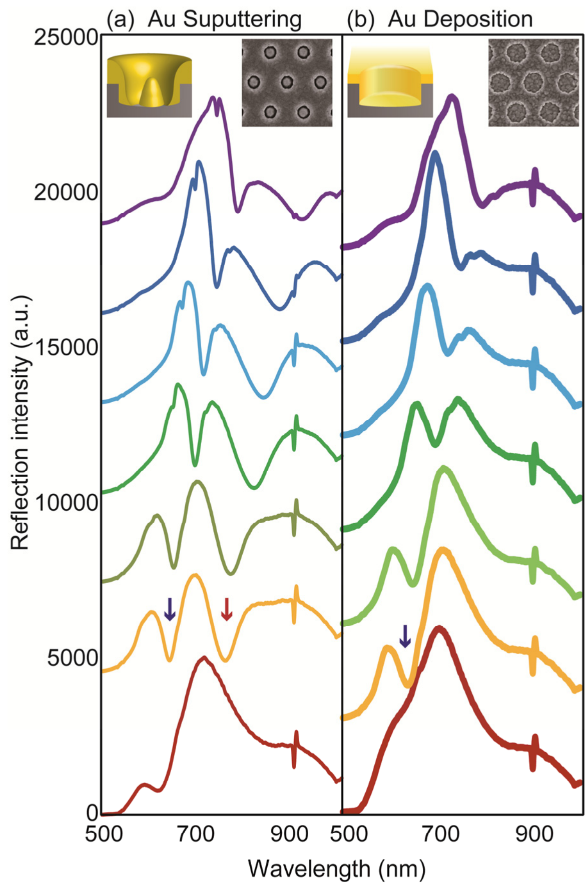

Figure 5: Reflection spectra from (a) gold nanopillar / nanohole substrates and (b) gold nanohole substrates fabricated by electron beam deposition method when various refractive index liquids were cast on the substrates (-- air (1.0), -- water (1.333), -- ethanol (1.36), -- glycerol (1.47), -- oil (1.516), oil (1.584, -- oil(1.7).