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Fabrication of ultrasmooth complementary split ring resonators by an improved template stripping method on SU-8

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ABSTRACT

It has been demonstrated that metamaterials play a very important role in plasmonic devices. The behavior of surface plasmon polaritons (SPPs) is highly related with the roughness of metal/dielectric interface, for instance, a smooth metal surface can largely elongate the propagation length of SPP. In this work, an improved template stripping method is developed to fabricate ultrasmooth metamaterials of nanoscale complementary split ring resonators (CSRRs). By using an ultraviolet photoresist of SU-8 as the adhesive, nanoscale metal CSRR structures with ultrasmooth surface were peeled off from silicon template by employing a combined process of pre-baking, UV irradiation and post-baking. By optimization of the pre-baking temperature and the depth of the nanopatterns, ultrasmooth nanoscale CSRRs with a gap of 30 nm were obtained. Our results indicate that the approach of template stripping by implementation of an SU-8 adhesive layer is an effective method to produce elaborated hollow nanostructures with ultrasmooth surface and nanoscale gap, which may have potentials in sensing applications.

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1. Introduction

Studies on metamaterials consist of subwavelength structures have been one of the main concerns in physics and material science in the past decade. Split ring resonator (SRR) and its complementary structure (complementary split ring resonator, CSRR) were put forwards as meta-atoms to modulate electromagnetic waves [1,2], and they have been widely used in bandpass filters, planar waveguides and duplexers in microwave and Terahertz regions [3–6]. However, there is little work about the metamaterial of CSRR in visible region. The grain size of metal films (mostly deposited by evaporation) is usually comparable to the feature size of the nanoscale subwavelength CSRR structure in visible range. Thus, the roughness of metal films brings disadvantages to the fabrication of nanosized CSRR by traditional planar technology.

The electromagnetic properties of CSRRs mainly depend on the surface plasmon polaritons (SPPs), which occur very close to the metal–dielectric interface, and they can be affected by the surface morphology of the structures. Up to now, various technologies have been developed to fabricate ultrasmooth structures such as template stripping [7–10], nanografting [11], indentation lithography [12], focused-ion-beam direct-pattering of single-crystalline metal [13] and thermoplastic forming [14]. Among these technologies, template stripping, which is based on the mature nanofabrication of silicon template and the adhesions difference between epoxy/metal and metal/silicon, is the most convenient and widely used choice of fabricating full metallic ultrasmooth nanostructures in large area. Traditional stripping method makes sufficient contact between epoxy and metal pattern on template and has been the effective method for fabrication of various nanostructures [7,8]. However, when it comes to CSRR with complicated shape and nanosized gaps, unfortunately, the metal patterns are usually completely peeled off, which makes it hard to form hollow patterns by such method. Thus new fabrication methods and improved techniques are highly demanded.

In this paper, we developed an improved template stripping method by using an ultraviolet resist SU-8 as adhesive by employing a combined process of pre-baking, UV irradiation and post-baking. After careful optimization of the processing parameters, such as the pre-baking temperature and the pattern depth in silicon template, a metamaterial of clean CSRR patterns with ultrasmooth surface and a gap of 30 nm was fabricated successfully. This method enables fabrication of complicated hollow structures with ultrasmooth surface and nanoscale gap.

2. Fabrication processes

The schematic of the improved template stripping method was illustrated in Fig. 1. First, PMMA (495 k, 5%) electron resist was spin-coated on a silicon wafer. Then the sample was pre-baked on a hot-plate at 180 °C for 60 s and exposed by electron beam lithography system (Raith 150) at 20 kV. The exposed sample was developed in MIBK:IPA (1:3) developer at room temperature

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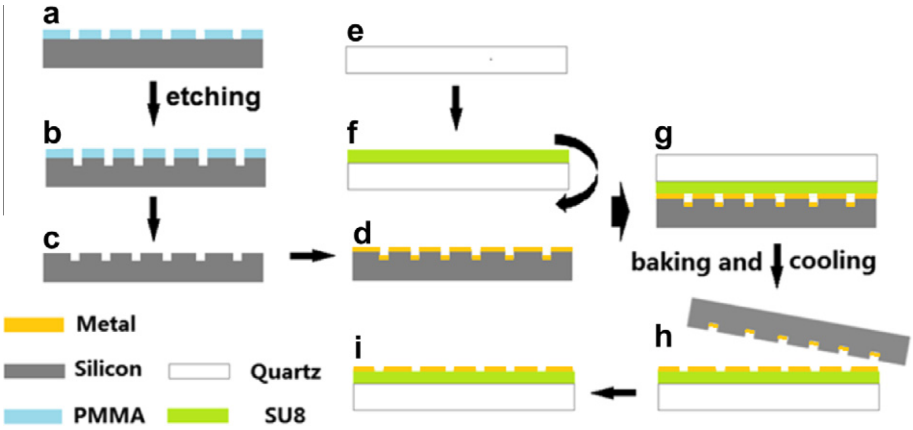

Fig. 1. The schematics of the improved template stripping process.

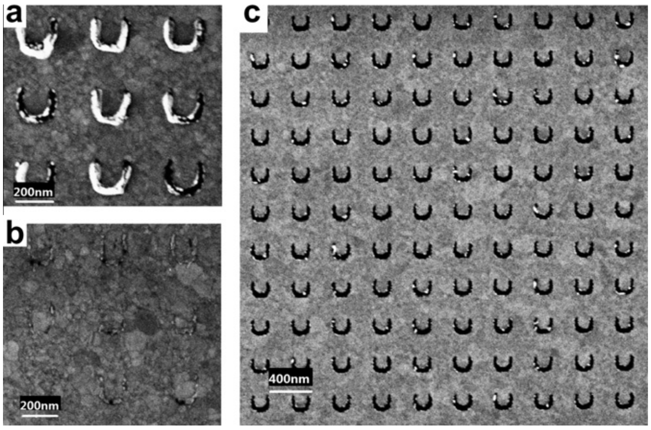

Fig. 2. The SEM images of stripped CSRR structures: (a) The stripped CSRR structures formed using pre-baking temperature of 175 °C, showing over pre-baking resulted excessive stripping; (b) The stripped CSRR structures fabricated with template pattern depth of 90 nm, showing too shallow depth resulted excessive stripping; (c) the stripped clean and regular CSRR structures achieved using prebaking temperature of 155 °C and template pattern depth of 160 nm.

for 40 s, rinsed in IPA for 30 s and blown dry by pure nitrogen gas. After that, the nano-patterns were formed in the electron resist layer, as shown in Fig. 1a. A following etching process was performed in an Inductively Coupled Plasma Reactive Ion Etching system (ICP-RIE, PlasmaLab System 100, Oxford Instruments) to transfer the resist patterns onto the silicon substrate (Fig. 1b). When the PMMA resist was removed by acetone, a template with nanoscale CSRR structures was prepared, as shown in Fig. 1c. Then a 30 nm Au film was deposited onto the silicon template (Fig. 1d) by electron beam evaporation (Peva-600E, Advanced System Technology).

On the other side, a quartz wafer (Fig. 1e) with a spin-coated SU-8 resist layer (with a thickness of about 5 μm) was used as a target substrate (Fig. 1f) to support the stripped metal structures. The SU-8 resist is a kind of polymer based on epoxy resin, which can be solidified by UV irradiation and displays high thermal/chemical stability and an unusual transparency, as well as good resistance to mechanical damages [15]. Then the target substrate was turned upside down and put onto the metal-covered template, as shown in Fig. 1g. The processes of pre-baking, UV irradiation and post-baking were taken in turn to solidify the SU-8 layer to make it tightly adhere to the Au film. By stripping the silicon template from the target substrate, the Au patterns were left on the SU-8 layer, as shown in Fig. 1h. The expected nanostructure was finally adhered to the target substrate (Fig. 1i). The residual gold in the silicon slits can be removed by I_2/KI solution, and the template can be reused. However, the stripping and wet-etching process will cause damage and contamination to the template, which will reduce the quality of stripped patterns. After carefully cleaned by alcohol, the template can be reused for at least five times with all completely formed nanoscale CSRR structures in large size.

3. Optimization

In our experiments, it was found that the fidelity of the stripped nanoscale CSRR structures largely depends on the efficient adhesion between the SU-8 resist and the metallic patterns, and the quality of silicon template. So a series of experiments were carried out for optimized processing conditions.

Firstly, the effect of the adhesion between the SU-8 layer and the Au patterns on the Si template was studied. In order to increase the adhesion, we took a pre-baking and UV irradiation process successively to solidify the SU-8 resist when it was in close contact with the Au patterns on template. It is known that bubbles can be easily formed in the interface when attaching a hard template onto glue coated substrate. Therefore, a pre-baking process before UV irradiation is necessary.

In order to optimize the pre-baking process, a series of pre-baking temperature ranging from 95 to 175 °C with a step of 20 °C were tested, and the baking time is fixed at 2 min. For the 95 °C pre-baking sample, a lot of bubbles formed in the interface and result in defects in the stripped metal film. With the increasing of pre-baking temperature, the viscosity of SU-8 decreased, and the bubbles in SU-8 layer can be easily squeezed out by the pressure that comes from the weight of quartz wafer. When the pre-baking temperature was increased up to 155 °C, the bubbles completely disappeared. However, further increasing the pre-baking temperature up to 175 °C, the viscosity of the SU-8 further decreased to a

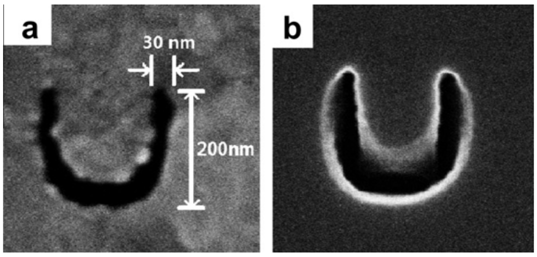

Fig. 3. Typical SEM images of the stripped CSRR structures with period of 400 nm (a) and the corresponding structures in the silicon template (b).

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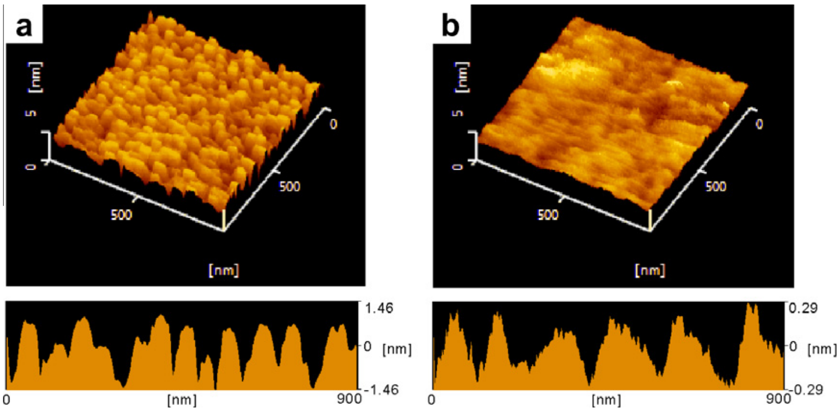

Fig. 4. Typical AFM image of the as-deposited rough metal film deposited by electron beam evaporation and the corresponding cross-section profile; (b) typical AFM image of the stripped smooth metal film and the corresponding cross-section profile.

point that it can flow easily into the template slits, leading to firm contacting of the SU-8 with the metal layer on the slit side wall and the bottom. Consequently, excessive stripping is unavoidable, as shown in Fig. 2a. Thus pre-baking at 155 °C for 2 min was fixed and used for further experiments.

After pre-baking, SU-8 layer was solidified by an UV exposure for 30 s with an overexposure dose of 300 mJ/cm^2. With such treatment, the template was easily stripped off. We also found that lots of small bumps appeared on the metal surface when the stripped samples were placed in air for some time. This instability of the samples mainly originates from the incomplete cross-linking of SU-8. SU-8 is a kind of chemically amplified resist, and the cross-linking reaction happens during the post-baking process, thus sufficient post-baking time is essential for the solidification and stability of SU-8. Based on systematic experiment, we found that post-baking at 95 °C for 30 min is sufficient to stabilize the SU-8.

Furthermore, the quality of silicon template is also very important to produce CSRR structures with good performances. In our work, the CSRR structures were formed on silicon template, consisting of deep slits with a gap of about 30 nm. After Au deposition, the top surface of silicon and the bottom of the slits were both covered by a layer of Au. Obviously, only the Au on the top surface of the template is expected to be peeled off to form the designed hollow CSRR structures. To achieve clean CSRR features on stripped sample, besides avoiding excessive high pre-baking temperature, an appropriate depth of template patterns is also important. In this study, nanopatterns on silicon templates were etched with different depth ranging from 90 to 240 nm. For template patterns with depth of 90 nm, the metal films on the slit bottoms would be peeled off, which lead to obvious defects (excessive stripped CSRR) in the final product as shown in Fig 2b (depth of 90 nm). For the spilt patterns with a gap of 30 nm in our experiments, the excessive stripping gradually disappeared with the increasing of the pattern depth, and a clear separated feature can be obtained when the etching depth reached 160 nm. It was also found that the nanopatterns formed on the template may suffer from deformation when the pattern depth is further increased, thus a critical depth of 160 nm was chosen in order to get elaborate structures. Using the aforementioned optimized process conditions, CSRR structures with good fidelity were finally fabricated, as shown in Fig. 2c.

4. Results

Fig. 3a show the SEM images of stripped CSRRs with a unit size of 200 nm, a gap of 30 nm, and a period of 400 nm, which has very good similarity in both shape and size with the silicon template shown in Fig. 3b. It is hard to be achieved by the approach of exposure/etching or traditional stripping method. Fig. 4a shows the atomic force microscope (AFM) images of the stripped and unstripped metal film deposited by electron beam evaporation and the corresponding cross-section profiles are shown in Fig. 4b. The root mean square (RMS) surface roughness was 0.4 and 1.3 nm in a 1 μm^2 area for the stripped gold surface (the side that in contact with the Si substrate surface after deposition) and the as-deposited metal surface, respectively. Obviously, using the improved template stripping method with an SU-8 adhesive layer, the surface roughness of the CSRR structures has been significantly improved.

It had been proved that the SPP propagation is highly correlated to the roughness of metal films, thus the surface roughness of CSRR structures is a very important factor that determines the performances of plasmonics. Although the template stripping method cannot reduce grain size and avoid SPP scattering by grain boundary, it can smooth metal surface to reduce the damping of SPP propagation. At the interface, the bumps of metal grain will scatter SPP [7] or generate unwanted random "hot-spots" [8], which both have negative effect on SPP propagation. The ultrasmooth metal surface can elongate SPP propagation length [7] and enhance refractive index sensitivity [9]. Most importantly, the nanosized gap can generate strong field enhancement, which could be useful in plasmonic sensing devices. In addition, we would like to point out that besides CSRR nanostructures, other hollow metal structures with complicated shapes can also be made by the improved template stripping method.

5. Conclusion

In this paper, we have developed an improved template stripping method to fabricate ultrasmooth nano-scale CSRR structures by using silicon as the template and SU-8 as the adhesive. After processes optimization, a metamaterial of CSRRs structure with ultrasmooth surface (RMS of 0.4 nm) was fabricated with a gap of 30 nm. This method enables fabrication of nanoscale complicated hollow structures with ultrasmooth surface on various kinds of substrates, and potentially could benefit the plasmonic devices in sensing applications.

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