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Fabrication of Large Area Periodic Nanostructures Using Nanosphere Photolithography

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Abstract Large area periodic nanostructures exhibit unique optical and electronic properties and have found many applications, such as photonic band-gap materials, high dense data storage, and photonic devices. We have developed a maskless photolithography method—Nanosphere Photolithography (NSP)—to produce a large area of uniform nanopatterns in the photoresist utilizing the silica micro-spheres to focus UV light. Here, we will extend the idea to fabricate metallic nanostructures using the NSP method. We produced large areas of periodic uniform nanohole array perforated in different metallic films, such as gold and aluminum. The diameters of these nanoholes are much smaller than the wavelength of UV light used and they are very uniformly distributed. The method introduced here inherently has both the advantages of photolithography and self-assembled methods. Besides, it also generates very uniform repetitive nanopatterns because the focused beam waist is almost unchanged with different sphere sizes.

Keywords Microspheres · Photolithography · Nanostructures

Introduction

Large area periodic nanostructures exhibit unique optical and electronic properties and have been applied into many areas, such as photonic band-gap materials [1], high dense data storage [2], and photonic devices [3]. To fabricate these periodic nanostructures, standard photolithography methods cannot easily reach the resolution required. High-resolution methods such as e-beam lithography and focal ion beam milling are too slow to reach a large area because of their inherent serial property. Nano-imprint methods are fast to be applied, but it needs to use the mold, which requires the same resolutions as the patterns. So, it also benefits from the development of fast, economic, and high throughput fabrication methods with a high resolution.

We have developed a novel photolithography technique, the Nanosphere Photolithography (NSP) technique [4, 5], which utilizes a self-assembled ordered monolayer of hexagonally close packed (HCP) micro-spheres as nano-jets [6] to generate sub-wavelength regular patterns over a large area on standard photoresist. Here, we will fabricate *periodic metallic nanoholes perforated* in gold and aluminum films *using NSP technique*. We used NSP to produce a large area of hexagonally packed nanopillars of negative photoresist with a strong undercut. Using these nanopillars, we produced large area uniform nanoholes perforated in different metal layers with controlled thickness by lift-off process. The diameter of the nanoholes is about 180 nm and the period of the hexagonal array is 1 μm, which is depending on the size of the microspheres used. The exposure wavelength we used for NSP is a broadband source centered about 400 nm, which is far greater than the pattern size.

Simulation Results

Figure 1 shows the 3D-FDTD simulations of light's intensity profile for silica microsphere with the diameter of 1 μm for a conventional UV lithography *i*-line ($\lambda = 365$ nm).

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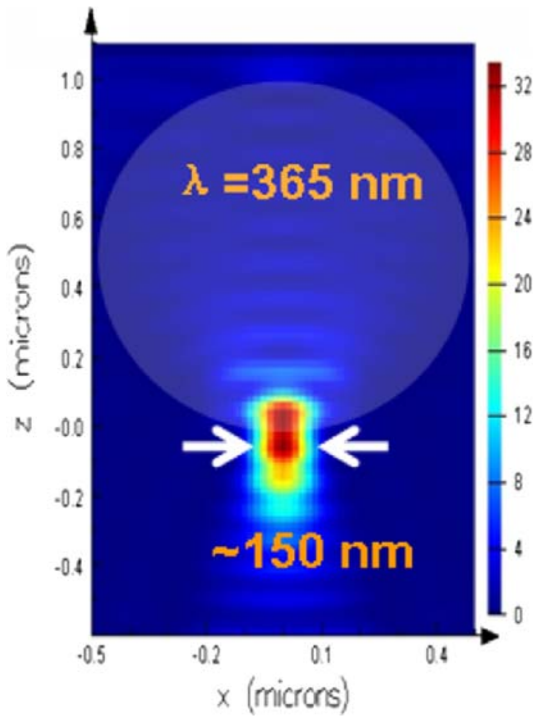

Fig. 1 One example of the focus process by the silica micro-sphere of 1 μm

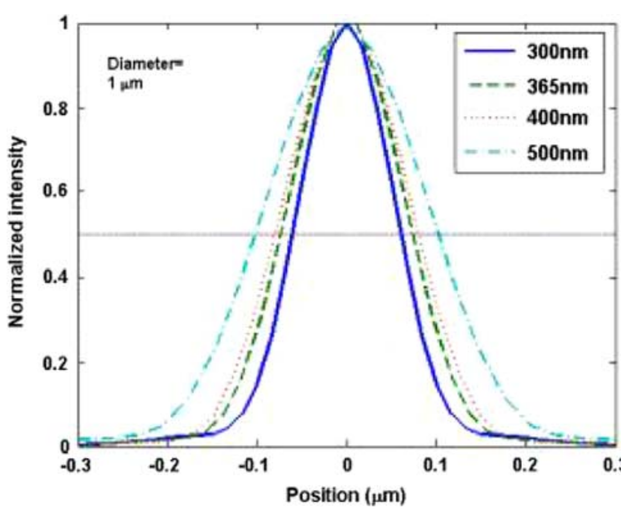

Fig. 2 The normalized intensity of the focused light versus the position with different wavelengths of UV light

The focused light's intensity is about 30 times as strong as the input light's and the full width at half-maximum (FWHM) of the focused light is about 150 nm, even smaller than half of the wavelength. Figure 2 is the normalized light intensity cross section after being focused by silica micro-sphere of 1 μm diameter with different wavelengths of UV light from 300 to 500 nm. It shows that the FWHM values of the focused light are getting smaller when the wavelength is scaling down. FWHM of the light intensity is a good measure of the photoresist exposure [5], so it is possible that the features we produced will scale down using the UV light of smaller wavelengths.

Experiment

The major processing steps are shown in Fig. 3. A standard commercial positive or negative photoresist such as *Shipley 1805* was spun on the substrate. A large area of HCP monolayer of silica microspheres was formed by the

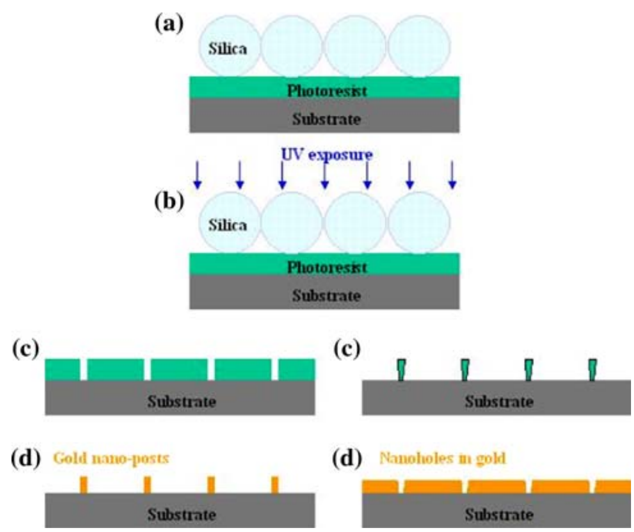

Fig. 3 The schematic process of NSP for fabrication nanostructures; from (**b**) to (**c**), it switches to two different processes with positive or negative photoresist used

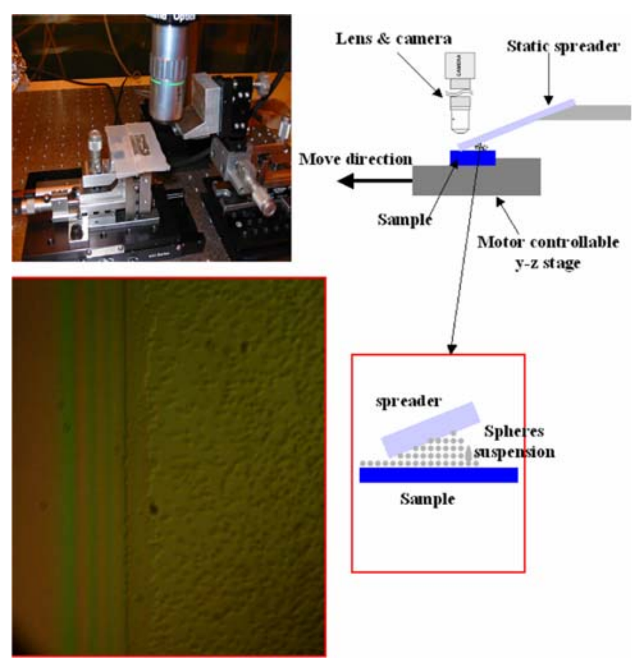

Fig. 4 The setup used for forming a large area of HCP micro-/nano-spheres and one snap shot of the spheres forming HCP monolayer

convective self-assembly setup shown in Fig. 4. *The silica microspheres were bought form Bangs Laboratory, Inc.* (http://www.bangslabs.com/index_static.php). The setup was placed on an optical table in the UV protection clean room. The high-resolution electrical motor made the movement of the stage with the sample. The method utilizes the convective force of water to push the spheres into a close monolayer along the meniscus as the stage moves. In *the* area of the meniscus, the thin water layer is easily evaporated and the microspheres stay on the surface. The samples were exposed by conventional photolithography instrument (Quintel Q-4000) under low exposure energy with a broad wavelength centered at 400 nm. Before development, the spheres were removed by ultrasonication in D.I. water. The photoresist was developed by AZ-300 MIF developer. The metal layers were deposited by electron-beam evaporator (Edwards Auto-306).

Results

A large area of HCP monolayer of silica microspheres was formed as shown in Fig. 5a. The Scanning Electron

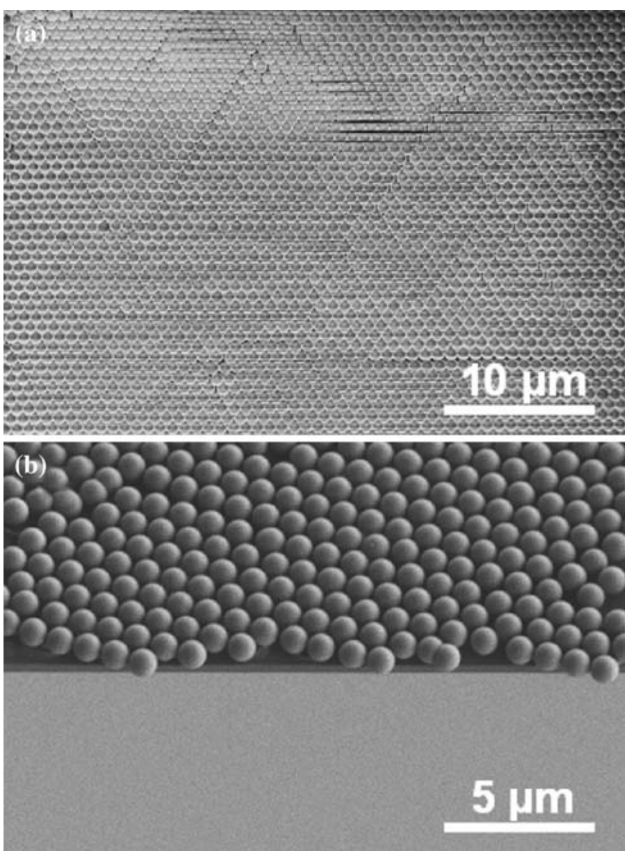

Fig. 5 (**a**) A typical SEM image of a large area of HCP monolayer of silica microspheres forming on top of photoresist; (**b**) the titled view of the spheres on photoresist

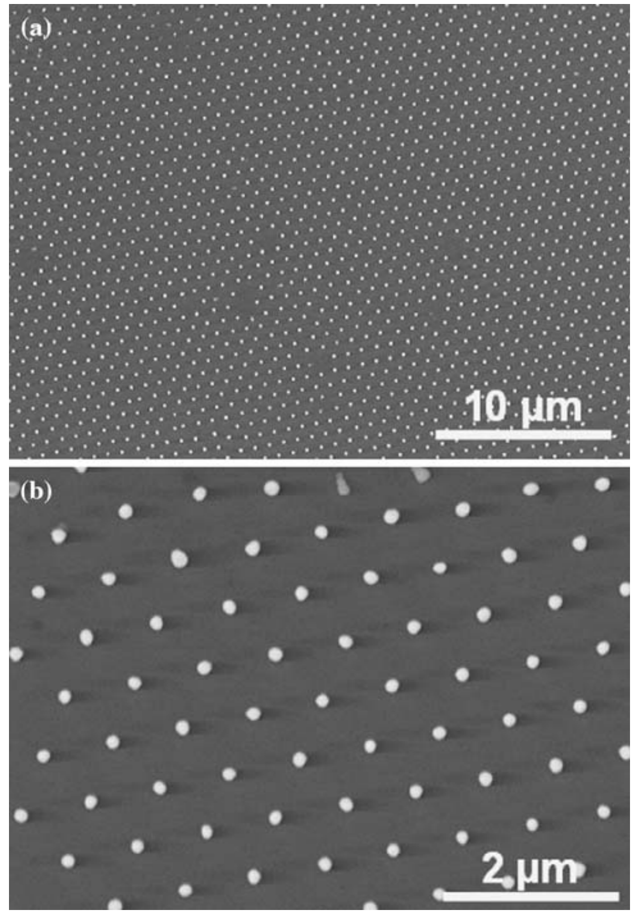

Fig. 6 (**a**) A typical SEM image of a large area of uniform nanopillars of photoresist after development; (**b**) the enlarged view of the nanopillars

Microscope (SEM) image shows the top view of a typical monolayer of silica spheres with the diameter of about 1 μm on top of photoresist. The area of the microspheres can be as large as millimeters *by millimeters*. Figure 5b shows the tilted enlarged view of the microspheres on photoresist. As shown in the figure, there seems an adhesion force between the spheres that makes the spheres close to each other. Figure 6a shows a large area of hexagonally packed nanopillars of photoresist after development. In the array there are some defects including the pillars falling off and an empty region without pillar, but they have a very small ratio. Figure 6b is the enlarged view of the nanopillars. The diameter of the nanopillars is about 180 nm and the periodicity of the hexagonal array is 1 μm, identical to the diameter of the microspheres. The height of the photoresist pillars is about 500 nm, which is depending on the photoresist thickness. Figure 7a and b shows the SEM images of the nanoholes perforated in gold and aluminum films separately, which were formed by lift-off on the photoresist nanopillars. The diameter of the nanoholes is about 180 nm and they are very uniform. Besides using

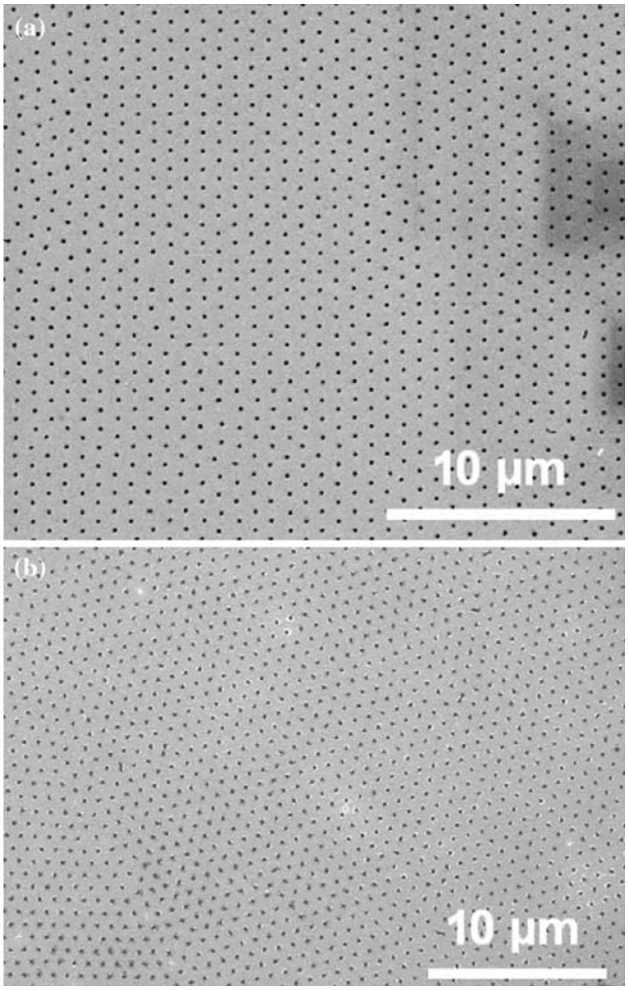

Fig. 7 SEM images of hexagonal distributed uniform nanoholes (diameter of about 180 nm) perforated in the gold (**a**) and aluminum (**b**) films fabricated by our lithography technique combined with lift-off process

1-μm microsphere, different sizes of microspheres can be applied in NSP, so the periods of the nanohole arrays produced can also be changed.

Conclusions

We have presented a large area of uniform metallic nanoholes of about 180 nm produced by Nanosphere Photolithography technique with a broadband wavelength centered at 400 nm. Our simulation results show that even smaller nanoholes with tunable periods can be generated with a shorter wavelength. The technique *demonstrated* here supplies an alternative routine for manufacturing large areas of periodic nanostructures.

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