High resolution photolithography using arrays of polystyrene and SiO₂ micro- and nano-sized spherical lenses

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Abstract. Photolithography mask made of close-packed array of micro- and nano-sized spherical lenses allows to obtain the ordered structures and provides highest “optical resolution/cost” ratio between all existing photolithography and laser direct writing methods. In this letter, we present results of modeling the propagation of a plane wave falling on the array of quartz (SiO₂) microspherical lenses and focusing in the image reverse photoresist layer. We present here experimental results on fabrication of ordered arrays of submicron wells and columns and substrate preparation for growth of monocrystalline nanowires on metal surface using photolithography with mask of SiO₂ microspheres. Such ordered nano-sized arrays of wells and columns can be used in fabrication of further growth of monocrystalline nanowires, quantum dots and production of plasmon structures.

1. Introduction

Ordered arrays of micro- and nanostructures are objects of growing interest because of their unique properties and wide range of applications in nanoelectronics and plasmonics. Production of plasmonic and dielectric metasurfaces involves fabrication of large area periodic arrays of metallic and dielectric submicron structures. The latter fact impose extra requirements on photolithographic systems resolution leading to rise of equipment cost, lithography process cost and as a result to end-product price increase.

Industrial scanner-steppers are capable of forming the structures with 14 nm resolution. However, those systems are very expensive and are not suitable for scientific research. Research institutes and universities are commonly equipped with projection or laser lithography systems, which typical resolution is under the diffraction limit and optical scheme with corresponding resolution limit in 0.5-1 μm range.

Electron-beam lithography allows nanoscale resolution structure fabrication. However, it is very area-limited as a direct writing method. Projection lithography method through array of micro- and nanosized spherical lenses is based on focusing of incident plane wave under the lens. When the spheres are closed-packed an ordered array of micro- or nanostructures can be produced.

The purpose of this work was to develop fabrication method of ordered arrays of nanoscale columns or holes by means of photolithography through arrays of microspheres. These structures can
be used to create an antireflection coating or plasmon structures as well as a mask for selective area growth of monocrystalline NWs or quantum dots [1].

2. Experiment

In this work we developed technology of fabricating a mask for synthesis of NWs or quantum dots via deposition of close-packed polystyrene (PS) and SiO2 microspherical lenses one monolayer thick layer with further photolithography.

We carried out photolithography on glass substrate covered with layer of aluminum and layer of diluted AZ1505 optical photoresist over it. SiO2 microspheres 1.5 μm in diameter were deposited on the surface of the photoresist. Due to polarity of SiO2 microspheres and hydrophobicity of the photoresist, before the deposition of microspheres on wafer the sample was processed in the TMAH based developer. Treatment of the structure with the developer makes its surface hydrophilic due to reduction of hydrophobic groups and deposited microspheres uniformly distribute along the surface of the photoresist.

Deposition of microlenses was carried out via spin-coating method [2]. In order to obtain uniform distribution of the spheres we optimized the deposition parameters and found optimal balance between deposition time and rotation speed. In our experimental series we varied both parameters: the speed of the first centrifugation step was 300 rpm and dwell time time was 40 s while the last step was 2000 rpm for 10 s. From SEM images shown in Figure 1 (a, b) it is seen that during the deposition of microspheres with the spin-coating method homogeneous large area arrays of self-organized SiO2 microspheres with a diameter of 1.5 μm were obtained.

After that the photoresist covered with ordered array of microspheres was subjected to projection photolithography with 405 nm wavelength. The photoresist was then developed and the aluminum layer was removed through the opened windows in the photoresist with etching in acid solution. As a result, openings in aluminum with diameter of about 550 nm were formed on the substrate as shown in Figure 1 (c). This ordered array of holes can be used as a mask for synthesis of NWs or quantum dots.

In the next part of our experimental work we fabricated nanoscale holes array in SiO2 layer deposited on Si wafer partially covered with different metal layers. Formation of such a mask is desirable for synthesis of NWs or quantum dots. Initial approach to solving this problem was photolithography through the mask of SiO2 microspheres with the following etching of SiO2 through the prepared resist mask. In the course of our work it was found that size of the holes depends on several factors: size of the microspheres, type of photoresist, photoresist exposure duration. It is well known, that SiO2 etching provides broadened hole edges, so in order to obtain holes in SiO2 without etching involvement we used the reverse photoresist approach. The optimum photoresist thickness was determined via simulation described as follows in Section 3.

To obtain mask with desired morphology we deposited three metals (Cr, Ti, Mo) on different regions of Si (111) wafer via thermal evaporation in a vacuum. The reverse photoresist and SiO2 microspheres were then deposited with spin-coating on the wafer and exposed to radiation. As a result of the photoresist development ordered array of photoresist columns on the silicon surface and various metals was obtained. As one can see from Figure 1 (d) images obtained with SEM diameter of the columns is in the 220-300 nm range. Further, silicon oxide was deposited on the nanostructured substrate and after lift-off the resist was removed with SiO2 and ordered array of holes was obtained as shown in the AFM image shown in Figure 1 (e). This structure is suitable for growth of NWs or quantum dots.
Figure 1 (a, b, c, d, e). SEM images of ordered SiO₂ microsphere lens array on metal-covered glass substrate (a), top view of the SEM image of SiO₂ microsphere lens array (b), optical image of nano-structure array on metal-covered glass substrate after sphere-enhanced photolithography and metal etching (c) an ordered array of columns of the reverse photoresist on silicon and the diameter of the columns is in the range 220-300 nm (d), and AFM image of an ordered array of holes obtained by lift-off the removed resist with SiO₂ (e).

3. The modeling
In the course of the work 3D modeling of the propagating a plane incident electromagnetic wave through SiO₂ microspherical lenses into a photoresist layer with different photoresist thicknesses from 2 μm to 0.2 μm was carried out to determine the optimal radiation absorption by an optical reverse photoresist. For modeling, the length of the incident plane wave was chosen equal 405 nm in that the projection photolithograph, with which were carry out an experiments described above, has an i-line mercury lamp with a wavelength of 405 nm. On the figures 1 (a-f) presented different resist thicknesses (2 μm, 0.45 μm, 0.2 μm, 0.3 μm, 0.4 μm, 0.5 μm) for comparison. Comparing the simulation results shown in figure 1 (a-f) we conclude that for the wavelength equal to 405 nm and for diameter SiO₂ microspheres equal 1.5 μm the optimum photoresist thickness is 0.45 μm. These obtained data of modeling were used in the experiment to obtain ordered columns by photolithography using microspheres as shown in figure 1 (g). The profile of the absorbed light in the photoresist layer in figure 2 (b) has two expressed maxima which coincides with the column profile obtained experimentally as shown in figure 2 (g) This result makes it possible to use a reverse photoresist to obtain an array of ordered columns with nanometer sizes.
4. Conclusion

Results of the numerical simulation to determine the optimum thickness of the optical image reverse photoresist (0.45 μm) which allows to make a high quality photolithography when the photoresist is exposed through 1.5 μm SiO₂ microlens array. Here we present ordered layers of microspherical SiO₂ and PS lenses deposition technology on substrates with photoresist. Using the developed technology ordered openings array with a diameter about 550 nm as well as columns with a diameter of the order of 220-300 nm are obtained. The substrate preparation technology for growth of NWs using photolithography with SiO₂ microspheres mask on silicon and different metals was tested. With use of this method nanostructured substrates have been fabricated that can be used for further realization of plasmon structures as well as growth of monocrystalline NWs and quantum dots.

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