Unconventional Photolithography with Self-Assembled Plasmonic Nanostructures

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***Abstract*—To overcome the diffraction limit of conventional optical photolithography the electromagnetic design of a novel plasmonic bow-tie nanostructure is presented. The proposed structure can be fabricated by self-assembling dielectric spheres on a layer of photoresist, followed by metal deposition and removal of the spheres. The metallic bow-tie nanostructure can focus light to expose the photoresist. The intensity distribution in the photoresist is calculated by solving Maxwell's equations, to predict the clearance profile after development. The simulations provide the size of the nanospheres, the optimum thickness of the metallic bow-tie antennas and the photoresist, the optimum wavelength of the illumination and the development conditions, in order to obtain the desired pattern of holes arranged in honeycomb lattice.**

***Index Terms*—Photolithography, Metamaterials, Plasmonic nanoparticles.**

I. Introduction

The resolution of conventional optical lithography is limited by the wavelength of the exposing electromagnetic radiation [1], [2]. To overcome the diffraction limit and to allow the fabrication of specific nanostructures without using a mask created by electron beam lithography several techniques of unconventional optical lithography have been researched. Among these techniques the lithography with self assembled microlens arrays has been successfully demonstrated [2], with the typical period of the fabricated patterns in the range of a few hundreds of nanometers. At a fixed wavelength of the exposing light, the period can be reduced by decreasing the size of the dielectric spheres. However as the size of the dielectric spheres becomes subwavelength the focusing effect is gradually lost. The deep subwavelength nanoparticle array behaves as a homogenous layer with effective electromagnetic material properties.

Plasmonic resonances of metallic nanoparticles can allow future reduction in the size of the fabricated nanopatterns [3], [4]. The plasmonic resonances depend strongly on the size and shape of the nanoparticles and allow the fabrication of patterns with feature sizes smaller than 100 nm. Proper design with a full-wave electromagnetic solver allows the adjustment of the dimensions of the resulting pattern in function of the size of nanoparticles.

With an advance in computational electromagnetics a large number of commercial and freely available solvers are available for Maxwell's equations. Nevertheless the numerical simulation of metallic nanostructures presents specific challenges, which cannot be easily handled by any of the existing numerical solvers. At radio and microwave frequencies the metals can be modeled as perfect electric conductors. However as the working frequency is increased, and the sizes of the metallic structures is decreased, the wavelength of the exposing wave can became comparable or smaller than the penetration depth. At high frequencies metals may become transparent and dispersive material model has to be considered to account for the vibration of bounded electrons as well as the motion of the free electrons in the lattice of metallic atoms. When the size of the metallic components is smaller than the mean free path of the conduction electrons (~50 nm), the bulk electric permittivity of metals has to be modified to take into account the scattering of the conduction electrons form the particle boundary, which leads to an additional loss mechanism. The metallic structures can have resonances with very high quality factor; therefore, the use of time domain solvers (e.g. finite difference time domain method) can lead to extremely long runtimes. In addition, metallic structures, can concentrate the electromagnetic energy in very small volumes, much smaller than the smallest geometrical dimensions of the setup, leading to very large mesh sizes that make the solution by frequency domain solvers (e.g. finite elements) challenging.

In this paper the electromagnetic design of a novel metallic bow-tie nanostructure for unconventional photolithography is presented and the details of the electromagnetic calculations are discussed. The electromagnetic intensity distribution in the photoresist is calculated and utilized to predict the clearance profile after development, in order to determine the setup of the unconventional lithography and to estimate the process conditions.

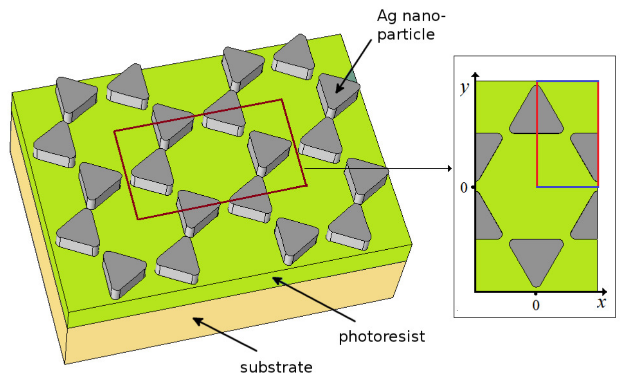
II. Computational Lithography of the Metallic Bow-Tie Array

Colloidal chemistry can manufacture a large variety of dielectric nanospheres. Such nanoparticles can be self assembled with the Langmuir-Blodgett technique on a photoresist covered glass substrate to form a single layer of nanoparticulate film. Metal deposition through the monolayer film as a mask results in a periodic structure of bow-tie shaped metallic nanoparticles formed in the spacing between the dielectric spheres. The spheres are removed by dissolution resulting in a very large area of bow-tie array on top of the photoresist layer. The metallic bow-tie structure is modeled as an array of rounded-corner triangular slabs as it is shown in Fig. 1.a. The side of the triangles is 50 nm, the thickness is

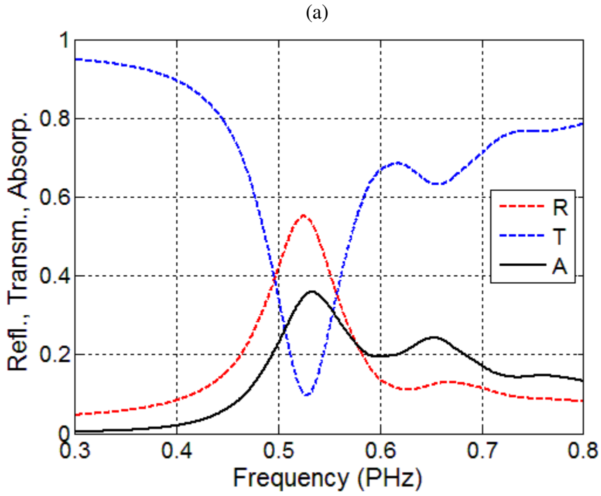
20 nm and the radius of the rounding is 4 nm. The thickness of the photoresist is 20 nm.

The transmission and reflection data of the bow-tie structure are calculated with the time domain and with the frequency domain solvers of CST Microwave Studio. The simulations are performed for the rectangular supercell shown in the inset of Fig. 1.a with periodic boundary conditions. The exposing light is a polarized plane wave propagating in the z direction with the magnetic component lying in the gap of two triangular elements. The periodicity of the structure is modeled with PMC and PEC boundary condition in the x and y directions respectively and the computational domain is reduced to one quarter with symmetric conditions. In the z direction the computational domain is truncated with waveguide ports that launch the exposing electromagnetic wave, absorb the outgoing waves and automatically calculate the transmission-reflection data. The material of the bow-tie array is silver, the photoresist is PMMA and the substrate is glass. The dispersive electric permittivity of the materials is from the SOPRA database [4].

The absorption spectrum, $A = 1 - T - R$, shown in Fig. 1.b has its highest peak at 0.532 PHz indicating plasmonic resonance at this frequency. Note that the time domain and frequency domain solvers produce similar results.



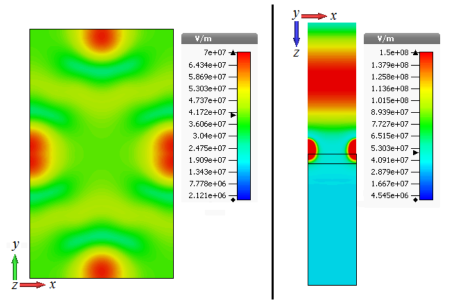
(a)



(b)

Fig. 1 The setup of the unconventional plasmonic lithography with silver bow-tie array on top of the photoresist supported by the glass substrate and the top view of the supercell used in the simulations are presented in (a). The calculated transmission, reflection and absorption data are shown in (b)

The distribution of the electric field intensity averaged over all phases at the resonant frequency 0.532 PHz is presented in Fig. 2 for two cutting planes of the computational volume. The large intensity spots correspond to the gaps which are aligned with the polarization of the source. The field distributions indicate that the photoresist is properly exposed and after dissolution the required holes pattern can be obtained. In the full paper detailed dissolution models [5], [6] will be applied to predict the final clearance profile of the resist after development.



(a) (b)

Fig. 2. The averaged electric field at 0.532 PHz in different cutting planes of the unconventional lithography setup: (a) in the xy plane at the bottom of the photoresist layer and (b) in the xz plane at $y = 0$ which passes through the gaps of two adjacent metallic nanoparticles

III. Conclusions

The proposed metallic bow-tie nanoarray, which can be fabricated by self-assembling dielectric nanospheres followed by metallic deposition, can support plasmonic resonance which leads to enhanced electromagnetic fields localized in the gaps of the nanostructure. Therefore the resist layer can be exposed and after dissolution holes arranged in honeycomb lattice with dimensions as small as a few tens of nanometers can be obtained. The simulations provide the size of the nanospheres, the optimum thickness of the metallic bow-tie antennas and of the photoresist, the optimum wavelength of the illumination and the development conditions, in order to obtain the desired pattern of holes.

Acknowledgement

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