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High index dielectric metasurfaces and colloidal solutions: from fabrication to application

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Abstract. High index dielectric nanoparticles and meta-materials have been proposed for many different applications, including light harvesting, sensing and metalenses. However, widespread utilization in practice also requires large-scale fabrication methods able to produce homogeneous structures with engineered optical properties in a cost effective manner. Here, it is presented a facile fabrication method for silicon nanoparticles which is scalable to 4-inch wafers and can produce a wide range of nanoparticle shapes on demand. We also show that the fabricated nanoparticles can be detached from their support using a simple substrate removal technique and then transferred to colloidal suspension. We will finally discuss some uses of the fabricated systems. For the metasurfaces, we will demonstrate complete absorption due to far field interference effects. For the nanoparticles colloids we will show the possibility of realizing an intrinsically chiral structure composed of a low-loss dielectric resonator and we will study optical trapping phenomena for different particle sizes and shapes.

1. Introduction

Electromagnetic (EM) metasurfaces are structured material layers with subwavelength thickness that appears smooth to an impinging EM field because the feature spacing is small compared to the incident wavelength. In particular, the use of high-index dielectric (HID) nanoparticles as metasurface building blocks, or meta-atoms, allows for extremely flexible control of the optical properties by engineering their morphology. Similar to their metallic counterparts, HID nanoparticles support multiple resonances[1] and are able to efficiently convert propagating fields into sub-wavelength volumes. HID nanoparticles also have additional advantages, such as thermal stability, reduced Ohmic losses and CMOS fabrication compatibility. These features render HID metasurfaces highly interesting for applications like wavefront manipulation, light managment, directional antennas, light harvesting, ultra-fast optical switching and molecular sensing, to name a few.[2]

The diverse pool of potential applications highlights the need for flexible and cost-effective fabrication methods that would enable realization of large-scale nanostructured HID materials with well-defined physical and optical properties. Many of the available techniques have their limitation: lithographic methods cannot cover large areas and the features are generally planar. On the other hand, wet chemistry synthesis of HID nanoparticles is far from being as mature as for metal nanoparticles, several methods are capable of producing high quality spherical silicon particles with a range of sizes have been recently reported.

In this document, we first demonstrate a method for large-scale fabrication of different silicon nanostructures using a modified version of the hole-mask colloidal lithography (HCL) technique. Afterwards, we present a method for removing such Si particles from a supporting substrate and used it to realize colloidal HID solutions. We show that this methodology can be used to detach and dissolve nanoparticles of diverse shapes ranging from disks to chiral particles. In the final part of the document, we will discuss some applications of the different fabricated systems.

2. Metasurface fabrication.

Figure 1a shows the basic principle for the fabrication of HID metasurfaces. A sacrificial layer, for example a resist or a SiO2 film, is formed by spin coating or deposition, on top of a deposited Si film. Polystyrene beads are then dispersed on the substrate. After a thin metal film evaporation and tape stripping, a hole mask is formed. The sacrificial layer is then etched through the holes in the mask, followed by evaporation of hard mask (Ni or Cr) nanoparticles of a chosen shape. These hard mask particles can be transferred to the poly-Si film underneath by anisotropic etching.

The method described can be used to produce homogeneous metasurfaces over 4-inch wafers, as shown in Figure 1b. Most importantly, it is also highly versatile and capable of fabrication of nanoparticles of different shapes simply by changing the conditions for deposition of the Ni mask as shown in Figure 1c and d.

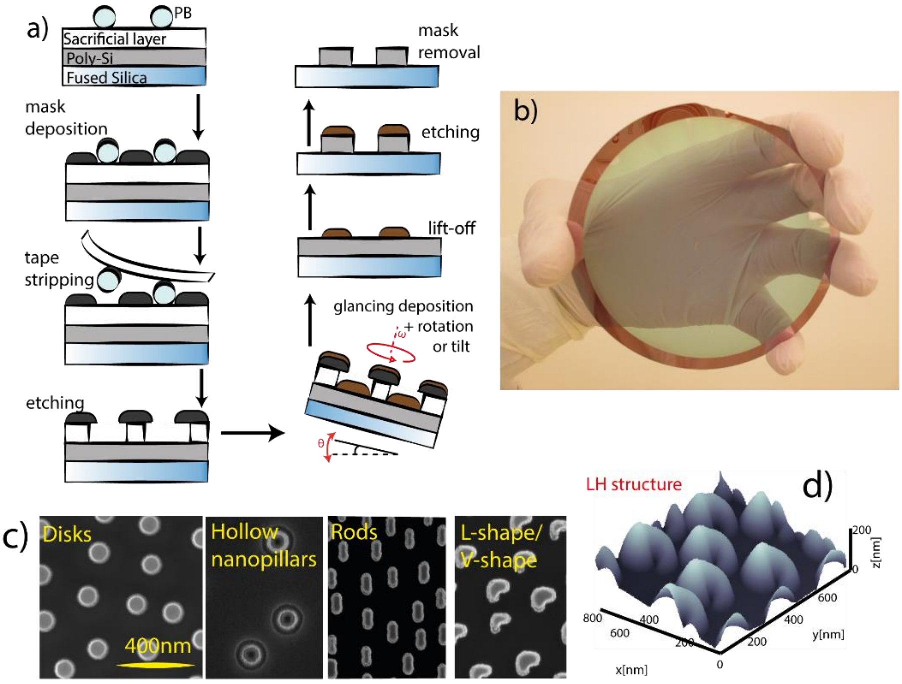

Figure 1. a) Schematic of HID metasurfaces fabrication. b) image of a 4-inch substrate covered with Si nanodisks. c) SEM images of different HID metasurfaces. d) Atomic force microscope image of a metasurface composed of 3D chiral Si nanocrescents.

3. Colloidal solutions

The fabricated nanoparticles can be removed from the substrate to realize colloidal. The as-fabricated metasurface is first dipped into a HF solution (5%) to selectively etch the substrate. Subsequently, diluted HF (0.1%) is drop-casted on the metasurface until the particles disperse into the solution. The HF solution is then left to evaporate. Finally, the particles are dispersed by sonication in an aqueous solution of cetrimonium bromide (CTAB, typically 1-5mM). The colloidal solution is stable for several months. Note that this method can in principle be adapted to substrates prepared by any large-scale fabrication method. The optical extinction measurements of the metasurface (b) and colloidal solution (d) composed of nanodisks, reveal the presence of tunable multiple resonances, typical of HID nanostructures.

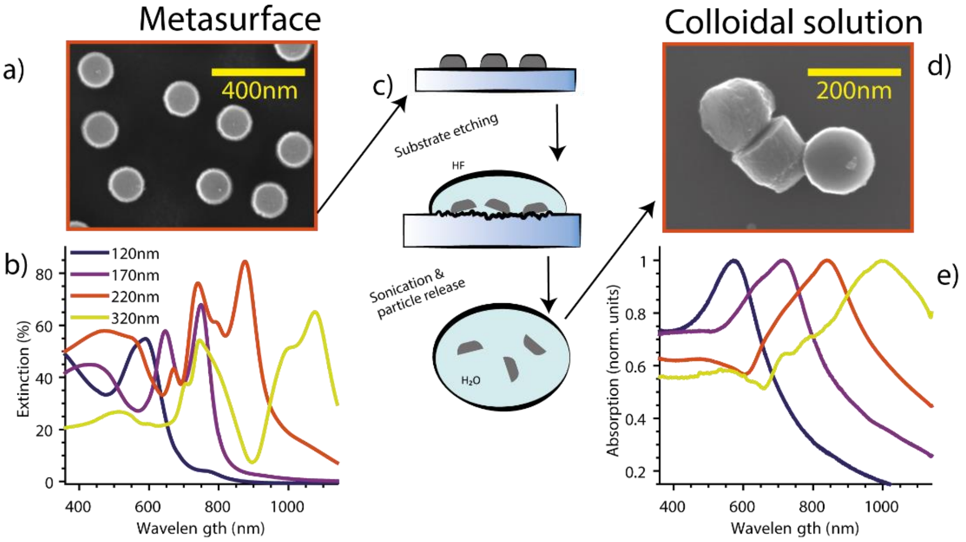

Figure2. From HID metasurface to colloidal solution. Using HF, the substrate supporting the Si nanoparticles can be etched. After HF dilution and evaporation, the particles can be released in solution by sonication, resulting in a colloidal suspension of HID nanoparticles. SEM image of a fabricated metasurface composed of Si nanodisks (a) and after redispersion onto a Si chip (d). The optical extinction spectra for nanodisks of different diameters on a substrate and in solution are shown in Figure b and e respectively.

4. Applications of the different systems

Figure 3 shows an overview of some of the applications that can be studied with the aforementioned fabrication methods. In figure 3a it is shown that a silicon metasurface excited in a total internal reflection configuration can absorb at least 97% of incident near-infrared light due to interferences between coherent electric and magnetic dipole scattering from the silicon nanopillars that build up the metasurface and the reflected wave from the supporting glass substrate. This “near perfect” absorption phenomenon loads more than 50 times more light energy into the semiconductor than what would be the case for a uniform silicon sheet of equal surface density, and it is irrespective of incident polarization.

Figure3b shows an optical trapping experiment using a solution composed of chiral colloidal HID nanoparticles. We found that the dispersed Si nanocrescents could be readily manipulated at the single particle level in solution using optical tweezers.[31] We demonstrate how a trapped nanoparticle can be moved across a surface using a power of the order of 50 mW at the sample. We also estimated the trapping stability based only on the probability density function of the average displacement.

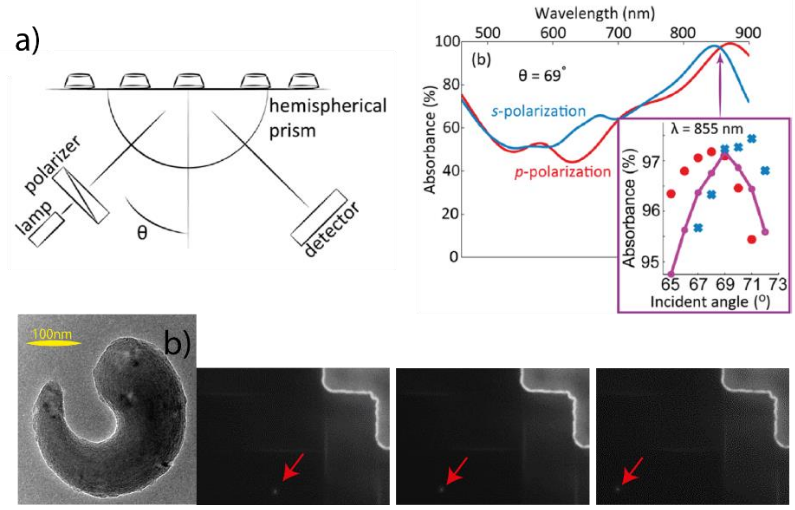

Figure3. Applications of HID nanostructures. a) Total internal absorption. A metasurface composed of nanodisks is illuminated in total internal reflection (left). By playing with the interference between the different modes, absorption can be boosted and it can reach up to 97% (right), irrespective to the polarization of the illuminated beam. b) Optical trapping of chiral nanocrescents. Using a 1064nm laser, chiral crescents (see left of a micrograph image) dispersed in solution can be trapped in 3D and easily manipulated. Selected images of a trapping experiment are shown on the right. The top right feature is a reference structure and the particle is indicated with an arrow in each movie frame.

5. Conclusions

We have demonstrated a method for efficient fabrication of large-scale metasurfaces composed of HID nanoresonators. Nanostructures of different shapes and sizes can be realized by modifying the process parameters. If desired, the nanostructures can be dispersed in a colloidal solution simply by selectively etching the substrate and then removing the particles by sonication. We present some applications for the two systems, namely perfect total internal absorption and the possibility of 3D optical trapping of such chiral nanostructures. Finally, the availability of tunable colloidal HID nanoparticles of non-spherical shapes opens wide avenues for further research applications. Similar to the case of colloidal plasmonic nanoparticles, we expect that many opportunities can be realized in the fields of bionanotechnology and nanomedicine.

These results are a collection of this previously published work:

- *Odebo Länk, et al. Nano Letter, 2017*
- *Verre et al., Advanced Materials, 2017*
- *Verre et al., Advanced Optical Materials, 2018*

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