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Large scale fabrication of silicon Mie nanoresonators: an alternative to gold ?

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Abstract

High index dielectric nanoparticles have been proposed for many different applications. The multiples optical properties of such called Mie resonators allowed us to question there use as an alternative to the metal, especially gold. Actually, the dipolar and quadupolar resonances, coupled with a strong near field can open new perspective in plasmonic uses. However, widespread utilization in practice also requires large-scale production methods for crystalline silicon nanoparticles, with engineered optical properties in a low-cost manner. Here, we demonstrate a facile, low-cost, and large-scale fabrication method of crystalline spherical silicon colloidal Mie resonators in water, using a blender as explained figure 1.



Figure 1: Fabrication of Silicon Mie resonator using a blender. The various diameters give a full range color spectrum scattering.

1. Introduction

Metallic nanoparticles are usually used for its scattering properties (optical antennas), near-field enhancement (SERS, MEF, coupling) and absorption (heat generation for medical issues, local chemistry). These three elementary bricks open a huge number of optical applications. But metallic nanoparticles have two strong limitations: the nar-

row optical range (need to change metal) and the corrosion for other metals than gold. We can so consider the high index dielectric nanoparticles as resonator for scattering and near-field enhancement applications. As metallic nanoparticles, the Mie resonators have electric dipolar (ED) and quadupolar modes, but the positive dielectric permittivity allow them to generate magnetic dipolar (MD) and quadripolar modes as visible on the FDTD simulations figures 2. The position of these modes is only dependant on the size of the nanoparticles, and we are able to cover the full range visible spectrum by tuning the diameter. Mie resonators can so be consider as a good alternative to gold for plasmonic applications.

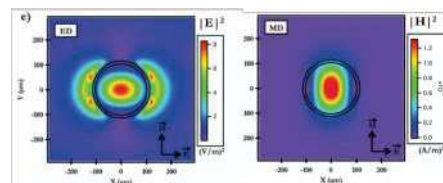


Figure 2: FDTD simulation on silicon nanoparticle. On left, the electric dipolar mode, on right, the magnetic dipolar mode.

2. Fabrication and applications

2.1. On substrates

Silicon Mie resonators on substrate can be fabricated using a femtosecond laser printing [1], but we propose a downsizing of the microelectronic well known techniques. These techniques are based on an etching (wet or dry) of a silicon wafer through a mask. Here we first demonstrate a wet etching of silicon weakly crystalline nano-layer by TMAH solution [2]. With this technique we can produce a wide size range nanoparticles, from 100nm to micron with very thin gap (down to 10 nm). But the clearance angle of the nanoparticles do not offer a very interesting optical proper-

ties. A dry etching is secondary developed using SF_6/O_2 cycles alternated with O_2 cycles [3]. This technique allow us to create nanocylinders with a very low clearance angle, then with a diameter range 70nm-210nm. We are able to cover the full visible range, and use them for nano-painting as visible figure 3.

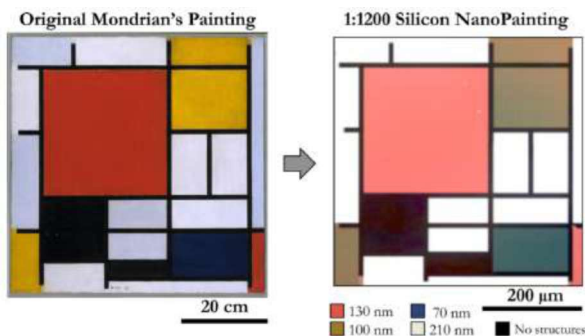


Figure 3: On the left, the original Mondrian painting *Composition in red, yellow, blue and black*, 1921 (59.5 x 59.5 cm^2). On the right, the nanopainting reduction (1:1200, 500 x 500 μm^2) made with silicon nanoparticles on glass

2.2. Colloidal Mie resonators

The chemical synthesis of silicon Mie resonator is a huge challenge, because the native oxidation of the silicon into silica. One approach have been developed based on a annealing at high temperature of a rich silicon layer, followed by centrifugations in HF [4]. Even through this method produce colloidal Si nanoparticles, we propose a more efficient and safe alternative method. Our method is based on the grinding of silicon pellets in water using a simple blender (see figure 1) [5]. The obtained nanoparticles are polydisperse with an almost spherical shape and the diameters controlled in the range 80-200nm by a centrifugation process. Then the size distribution of silicon nanoparticles enables broad extinction from UV to near-infrared, confirmed by Mie theory when considering the size distribution in the calculations. Thanks to photolithographic and drop-cast deposition techniques to locate the position on a substrate of the colloidal nanoparticles, we experimentally demonstrate that the individual silicon nanoresonators show strong electric and magnetic Mie resonances in the visible range.

3. Color application

Both techniques produce Silicon nanoparticles with a size range around 80-200nm. Both present a full visible spectrum scattering properties.

Figure 4 show the Gamut chromacity diagrams of the scattering of our silicon nanoparticles obtained with both techniques. We can observed than the nanoparticles made on substrates, even through they cover all the visible spectrum, lakes sparkles compared to our colloidal synthesis.

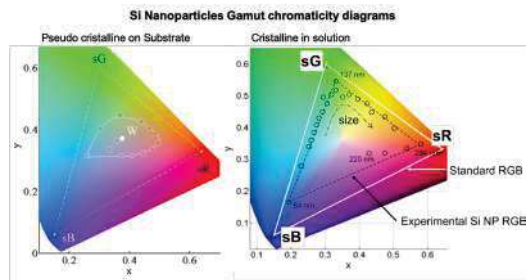


Figure 4: Gamut chromatic diagrams of Silicon Mie resonators. Left on substrate, right in solution.

4. Silicon equal to gold ?

Finally, might silicon Mie resonators replace gold in optical applications ?

We perform FCS (Fluorescence correlation spectroscopy) on both silicon and gold nanodimers [6]. We demonstrate than, for fluorescence enhancement, Silicon dimer with 20 nm gap size is equal to gold, and even better for higher gape size.

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