Three-dimensional Coaxial Plasmonic Metamaterial at Visible/UV Frequencies

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Abstract

We demonstrate a three dimensional metamaterial in the blue/UV spectral region composed of coupled coaxial plasmonic waveguides. Cathodoluminescence spectroscopy shows evidence for localized modes in the cavities and coupling between the coaxes. Results of interferometric transmission measurements at $\lambda = 476$ nm are presented and show a positive phase shift of 180° compared to a reference beam through air.

1. Introduction

Metamaterials are artificial materials composed of sub-wavelength building blocks. They provide an effective means to control the refractive index for light. By designing such metamaterials, the index can be made negative, giving rise to interesting properties as negative refraction and backwards phase propagation. Also special cases as $n = 0$, for which light propagates without a phase advance, or e.g. $n = 10$ for which light has a very small effective wavelength can be achieved. Most negative index metamaterials use sub-wavelength resonators as building blocks (e.g. split ring resonators). However, such designs suffer from high Ohmic losses and fabrication limitations when the resonances are pushed from the IR towards the visible part of the spectrum. Furthermore, the index highly depends on angle of incidence and polarization of the incoming light. These limitations can be circumvented by using metal-insulator-metal (MIM) waveguides [1] where the coupling of plasmonic waveguides can result in a material with an isotropic negative index in the visible/near UV [2].

Recently, a three-dimensional metamaterial composed of an array of coupled plasmonic coaxial waveguides was reported theoretically [3]. This material can be designed to have a highly tunable negative refractive index in the UV/blue spectral region with a relatively high figure of merit. The spherical symmetry of the coaxes results into a material with a nearly polarization independent refractive index which is nearly constant over a large angular range. Here, we show the experimental realization of such a coaxial metamaterial in the blue/UV spectral region. We characterize this material with cathodoluminescence (CL) spectroscopy and determine the optical response with interferometric transmission measurements.

2. Fabrication of coaxial waveguides

The fabrication of arrays of coaxes on a silicon substrate is a many-step process starting with electron beam lithography (EBL) using the negative high-resolution resist hydrogen silsesquioxane (HSQ). Development was performed in 25% tetramethylammonium hydroxide (TMAH) solution at room temperature followed by drying in a critical point dryer to prevent collapsing of the structures. Rings up to 300 nm in height, having a 90 nm outer and 50 nm inner diameter, can be fabricated using a 100 keV electron beam. Thinner resist layer can result in even smaller feature sizes. Scanning electron microscope (SEM) images of the resulting rings are shown in Figure 1a.

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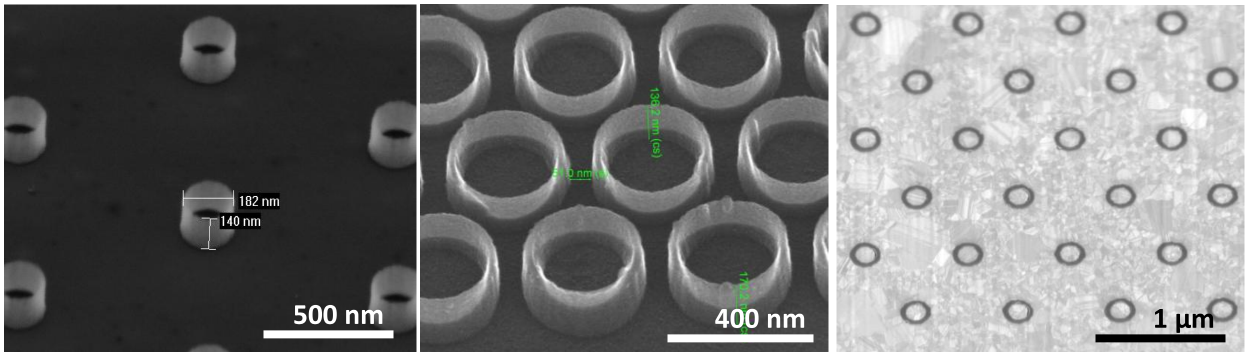

Figure 1 SEM images taken after different steps in the fabrication process. (a) Exposed and developed HSQ on Si substrate (b) coaxes after etching the structures into Si and (c) final metamaterial where the rings are infilled with Ag and polished with FIB shaving. The Si rings appear as dark circles in the Ag surrounding.

Next, the structures are transferred into the Si substrate with reactive ion etching using a gas flow of 35 sccm CHF_3 and 5 sccm SF_6, 125 W RF power, a pressure of 10 mT and 400 V DC bias. Figure 1b shows SEM images of the structures made in Si after etching. The resulting hollow Si pillars are infilled with Ag by thermal evaporation after removal of excess resist in HF (10 minutes dip in 1% HF solution). The excess silver is removed using focused ion beam (FIB) shaving using 30 keV Ga^+-ions, with the sample normal tilted 90° with respect to the ion beam, resulting in a polished surface as shown in Figure 1c.

Although the above procedure works very well on a bulk silicon substrate, some adhesion difficulties were encountered when fabricating the structures onto a silicon membrane, the latter being desirable for optical transmission measurements. Therefore we explored a procedure where a layer of hard-baked Novolak (HPR-504) is added as an etch mask between the silicon substrate and the HSQ. This additional layer has the advantage that the Novolak has a very good adhesion to the silicon as has the HSQ to the Novolak. Furthermore, due to the very high etch selectivity between HSQ and Novolak (1:47) we can use a thinner HSQ layer, resulting in a higher resolution or deeper structures.

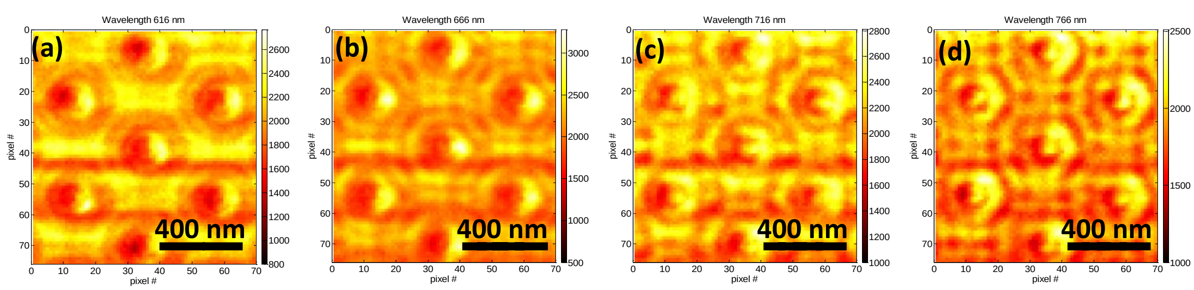

Figure 2 Cathodoluminescence measurements on a hexagonal array of 200 nm diameter coaxes with 800 nm center to center spacing. The color scale represents the intensity of the collected light. Emission is collected at (a) 616 nm (b) 666 nm (c) 716 nm and (d) 766 nm.

3. Cathodoluminescence spectroscopy

Cathodoluminescence spectroscopy allows mapping of the local density of states (LDOS) of the structures with a high spatial resolution of 10 nm. We use a 30 keV electron beam in a conventional SEM to excite the sample. The generated light is collected by an Al parabolic mirror over a large solid angle and sent to a spectrometer. Figure 2 shows CL images on a hexagonal array of 200 nm diameter coaxes with an 800 nm center to center spacing for different wavelengths. Sharp spatial features are observed corresponding to the individual coaxes. Furthermore, the images show interference patterns of surface plasmons scattered off the coaxes, demonstrating efficient coupling between coaxes. This coupling is essential for the material to act as an isotropic metamaterial.

4. Interferometry

The optical response of the fabricated material is investigated by measuring the phase shift of light transmitted through the sample. Preliminary results of interferometric measurements on a hexagonal array of coaxial plasmonic waveguides (215 nm inner and 360 nm outer diameter) fabricated on a 1300 nm thick silicon membrane are shown in Figure 3. The phase advance of light transmitted through the metamaterial is measured relative to a reference hole through the sample. The histogram in Figure 3 shows a large number of datapoints from multiple measurements on the same sample. The width is determined by the inaccuracy in the position of the moving mirror in the interferometer. A phase shift of 176° is obtained at a wavelength of 476 nm. The corresponding refractive index of 2.2 is in agreement with our calculations. For the determination of the index we assumed a 1200 nm thick Si substrate having an index of 3.5 at λ = 476 nm and a metamaterial thickness of 150 nm.

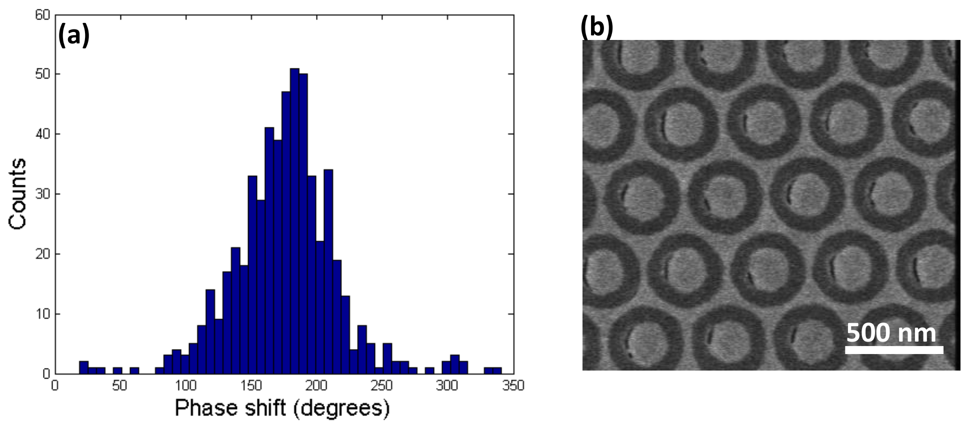

Figure 3 (a) Interferometric measurements on a hexagonal array of 260 nm diameter coaxes with 400 nm center to center distance. A phase shift of 176° is observed, corresponding to a refractive index of 2.2 (b) SEM image of the measured structures.

4. Conclusions

We fabricated a three-dimensional metamaterial composed of coupled coaxial plasmonic waveguides. The metamaterial is made of Si rings embedded in Ag by combining electron beam lithography, reactive ion etching and focused ion beam shaving. Cathodoluminescence spectroscopy show sharp spatial features corresponding to individual coaxes and we demonstrated coupling of the coaxes. Interferometric transmission measurements revealed a phase shift of 176° at λ =476 nm, corresponding to a refractive index of 2.2.

5. References

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