



Coaxial Plasmonic Metamaterials for Visible Light

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## Summary

Optical metamaterials are materials built from sub-wavelength building blocks, and can be designed to have effective optical properties that are not found in natural materials. A much-studied class of metamaterials uses small noble-metal resonant structures as building blocks, which have a magneto-electric response. These metal structures, however, suffer from high Ohmic losses near the metal plasma frequency and only work for a limited bandwidth. Furthermore, operation in the visible or UV spectral range requires scaling down of the structures to dimensions which are impossible to experimentally realize using today's available fabrication techniques.

In this thesis, we consider a different approach, that circumvents these limitations, in which the effective refractive index of the metamaterial is controlled by the coupling of surface plasmon polaritons (SPPs) on thin metal-dielectric-metal (MDM) waveguides. In particular, we consider a polarization-independent geometry consisting of a hexagonal array of dielectric coaxial waveguides, embedded in metal. The dispersion of the metamaterial can be controlled by tuning the geometry of the individual coaxes and designing the coupling with neighboring coaxial waveguides. In this thesis we describe the fabrication process and determine the optical properties of such a three-dimensional coaxial plasmonic metamaterial.

In Chapter 2, we describe the many-step fabrication process of the coaxial metamaterial in full detail. The fabrication process is a combination of electron beam lithography, reactive ion etching, metal infilling and polishing of the surface using a focused ion beam. We demonstrate successful fabrication of Si and hydrogen silsesquioxane (HSQ) hollow cylinders embedded in either silver or gold. We realize cylinders with an outer diameter in the range of 100 nm to 1  $\mu\text{m}$ , and various dielectric channel widths, of which the smallest was  $\sim 7$  nm.

In Chapter 3 we explore the optical properties of the coaxial metamaterial, both theoretically and experimentally. We calculate the dispersion of a single coaxial plasmonic waveguide for different material and geometry choices, and we find that the dispersion is particularly sensitive to the dielectric channel width. With these

insights we design an optimal geometry for which a negative mode index can be achieved, consisting of Si cylinders with an outer diameter of 150–200 nm and a dielectric channel thickness of 10–30 nm, embedded in Ag. Furthermore, we study the coupling of the coaxial waveguides placed inside a hexagonal array. We find that the transmission spectrum changes at a ring-to-ring distance smaller than 75 nm, while the dispersion is significantly altered for distances smaller than 50 nm. To support our theoretical findings, we measure the Fabry-Pérot resonances inside a 100 nm thick metamaterial slab with cathodoluminescence spectroscopy.

In Chapter 4 we perform interferometry measurements on a coaxial metamaterial in the visible spectral range. We show that both the measured and numerically simulated data are consistent with a negative mode index in the range of  $\lambda = 440\text{--}500$  nm. Furthermore, we experimentally demonstrate that the metamaterial has a polarization independent optical response.

Chapter 5 demonstrates how the plasmonic coaxial structures can be used as nano-apertures to trap small particles. The strong confinement of light inside the coaxial structures results in large optical trapping forces and geometrically confined potential wells in the near fields of the coaxial apertures. We numerically investigate how the optical forces are influenced by the geometry of the apertures, and find that 10 nm diameter dielectric particles with  $n = 2$  can be stably trapped at the surface of the apertures, using a transmitted power of only  $\sim 6$  mW. Furthermore, we show that the trapping forces are very similar if Au is used instead of Ag. Arranging single apertures in a hexagonal array with a center-to-center distance of 300 nm leads to similar trapping characteristics as for single coaxes. We describe how the optical forces can be measured using an AFM, and demonstrate the first experimental results.

We investigate the geometrical resonances supported by hollow Si cylinders in Chapter 6. We show that introducing a gap in the center of a 100–250 nm outer diameter silicon cylinder results in relatively small deviations from the well known electric dipole (ED) and magnetic dipole (MD) resonances of solid cylinders. The nature of the MD mode is preserved up to gap widths as large as 190 nm for a wall thickness of just 40 nm. We fabricate hollow nanocylinders with an outer diameter in the range of 108–251 nm using electron beam lithography and reactive ion etching, and measure their cathodoluminescence and dark field scattering spectra. Furthermore, we systematically explore the trends of the scattering cross-section numerically by changing the cylinder geometry. The gap provides an interesting additional parameter to further tune the resonant wavelengths and the radiation pattern in the far field. We find good agreement between theory and experiment.

Overall, this thesis provides insight in the optical properties of coaxial plasmonic metamaterials, both theoretically and experimentally. We show that this metamaterial can be designed to have a negative refractive index in the visible spectral range and explore the use of coaxial plasmonic structures as optical traps for nanoscale particles.





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