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Nanoantenna structures for strong coupling studies of surface plasmon polaritons and quantum dots

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ABSTRACT

We present measurement and simulation results of local surface plasmon resonances on silver nanoantenna structures, fabricated with electron beam lithography. Such structures offer interesting possibilities to study strong coupling phenomena between surface plasmon polaritons (SPP) and, e.g., quantum dots, along the lines of our previous work on vacuum Rabi splitting for SPP and dye molecules.

Keywords: nanoantenna, surface plasmon resonance, transmittance, extinction cross section, strong coupling, quantum dot

1. INTRODUCTION

Nanoantenna structures, showing local surface plasmon resonances in the visible regime, have been a research topic of great interest in recent years. Many theoretical studies¹⁻⁴ and experimental results^{5,6} are published in which the resonances of nanoantennas with different shapes and configurations have been considered. In this report, we present experimental results of resonances in arrays of rectangular silver nanoantennas and compare the experimental results with simulation results. Moreover, we consider the use of these nanoantenna structures to investigate strong coupling phenomena between surface plasmon polaritons (SPP) and quantum dots, related to our previous work on vacuum Rabi splitting for SPP and dye molecules.⁷⁻⁹ This report consists of three main parts. First, the experimental and simulation methods are discussed, including the fabrication of such nanoantenna structures. Second, the measurement results are shown for six different samples with differently sized nanoantennas, and two of these measurements are compared to results obtained by simulating models of our structures. Finally, we consider the improvements that are still necessary in order to use these nanoantenna structures for the study of strong coupling phenomena.

2. EXPERIMENTAL METHODS

2.1 Sample fabrication

Nanoantenna structures were fabricated in a clean room environment with a standard electron beam lithography procedure. Glass slides with indium tin oxide (ITO) coating were used as substrates. Glass was chosen as the substrate material because of its transparency that makes excitation and detection versatile from both sides of the sample. The conductive and transparent ITO layer is needed for electron beam exposure to avoid charge accumulation on the substrate. First, the glass substrates were sonicated for 20 minutes in acetone, followed by another 20 minutes of sonication in de-ionized water. After drying with nitrogen, they were further cleaned by a Reactive Ion Etching step with an oxygen plasma for one minute (an oxygen flow of 45 sccm, an argon flow of 5 sccm and a pressure of 200 mTorr). Second, a positive double layer resist was prepared by first spin coating a copolymer MMA (methyl-methacrylate) layer of approximately 500 nm on the substrate and then spin coating a 950PMMA (poly-methyl-methacrylate) layer of approximately 100 nm on top of the first layer. A double layer resist was used for a convenient lift off process applied later. Third, an electron beam exposure was applied and the designed pattern of nanoantennas was written in the resist. The structures of nanoantennas

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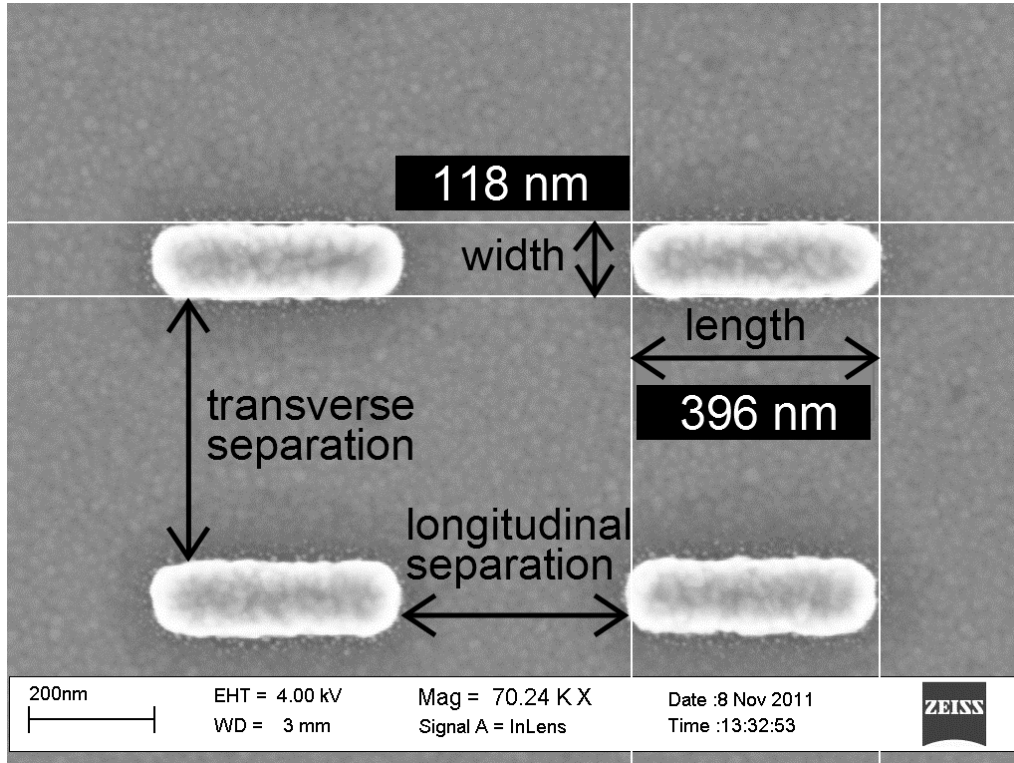


Figure 1. A zoom-in onto the sample surface, such that only four single nanoantennas are shown in the image. The dimensions and the transverse and longitudinal separation of the nanoantennas are indicated in the image. The measured transmittance for this sample is shown in Fig. 2(d). (SEM image)

that were used in this work are discussed below. After exposure, the resist was developed in 1:3 MIBK(methyl-isobutyl-ketone):IPA(isopropanol) and IPA solutions, 30 seconds in each. Next, a layer of 50 nm of silver was evaporated onto the substrates with an e-beam evaporator followed by a lift off process in heated acetone. Finally, the samples were imaged and the exact dimensions of nanoantennas were measured with a scanning electron microscope (SEM).

2.2 Nanoantenna structure

The nanoantenna structures used in the measurements were arrays of $50 \times 50 \mu\text{m}$ of equal-sized equidistant rectangular silver nanoantennas. The rectangular nanoantennas had a length of 200 to 550 nm and a width of 100 to 140 nm, and the separation was varied from 200 to 1350 nm in the transverse and longitudinal direction with respect to the orientation of the antennas (Fig. 1). A transverse separation of about 500 nm and a longitudinal separation of the length of a single antenna were found suitable for a decent signal and low interaction between the antennas, based on the change of resonant wavelengths as a function of separation distance. Longitudinal and transverse coupling in nanoantenna arrays has been studied with respect to separation distances.¹⁰ Furthermore, a separation of 100–300 μm was found to be appropriate for $50 \times 50 \mu\text{m}$ arrays, because they were then sparse enough to ensure that only a single array of nanoantennas was measured at a time, and on the other hand, they were dense enough to facilitate the lift off process around the patterned area.

2.3 Experimental setup and measurements

An inverted optical microscope (Nikon Eclipse TE2000) was used for transmission measurements of the fabricated nanoantenna samples. In the microscope, the sample was illuminated from above with white light and the transmitted light was gathered with an objective (10x magnification, 0.25 NA) below the sample. The gathered light was guided into an optical fibre with a core size of 200 μm . The fibre was aligned using a pin hole on the sample stage such that the input signal was maximized while the pinhole was in the middle of the crosshair on the eyepiece. The spectrum of the light coupled into the fibre was measured with an Acton SP2500i series spectrometer. Due to the 200 μm core fibre and the objective with 10x magnification, a circular area with an effective diameter of 20 μm was measured at a time.

The reference spectrum was measured through a plain area of the glass substrate, and the sample spectra were measured through the nanoantenna arrays, placing them in the middle of the crosshair one by one. The measured sample spectra were divided by the reference in order to get the transmittance of the nanoantenna arrays. Light absorption and scattering are the strongest at the resonance wavelengths, and the light scattered beyond the acceptance angle of the objective is missing from the transmission spectrum, such as absorbed light. Therefore, the resonance wavelengths show as clear dips in the transmission spectrum of the corresponding nanoantenna structure.

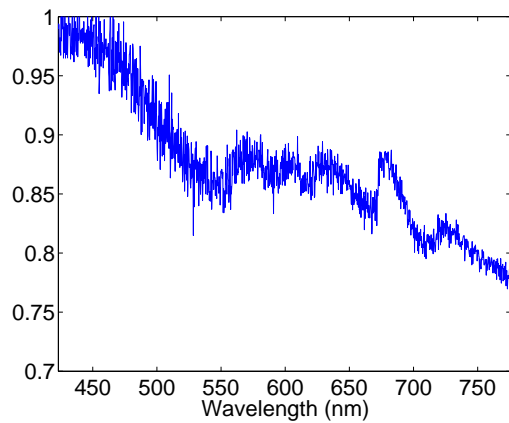
2.4 Simulation methods

The simulations were performed with a commercial software package called FDTD Solutions by Lumerical, which employs the finite-difference time-domain (FDTD) method to calculate the electric field at discrete spatial lattice points. In this study, we calculated the extinction cross section of nanoantennas as a function of wavelength, where the extinction cross section is the sum of the absorption cross section and the scattering cross section. The resonance peaks in the extinction cross section spectrum, which can have various line shapes,¹¹ correspond to local surface plasmon resonances of the antennas.

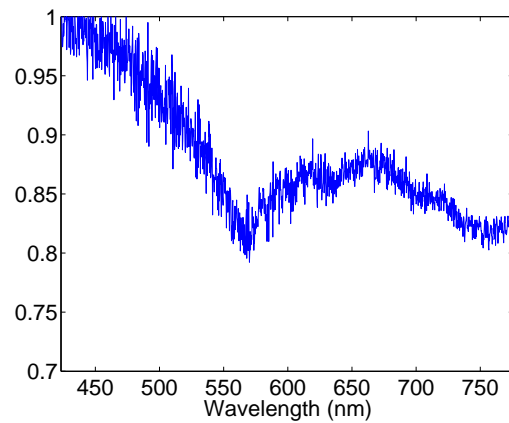
We used a broadband plane wave with a Gaussian spectrum as a source for excitation, and simulated both single nanoantennas and infinite arrays of nanoantennas with periodic boundaries, located on an ITO surface in air. The dimensions and spacing of the antennas were the actual ones as measured with the scanning electron microscope for each sample. We used a spatial resolution of 2 nm, the model for silver was based on the tabulated data as found in Palik,¹² and a constant refractive index of 2 was used for the ITO layer.

3. RESULTS

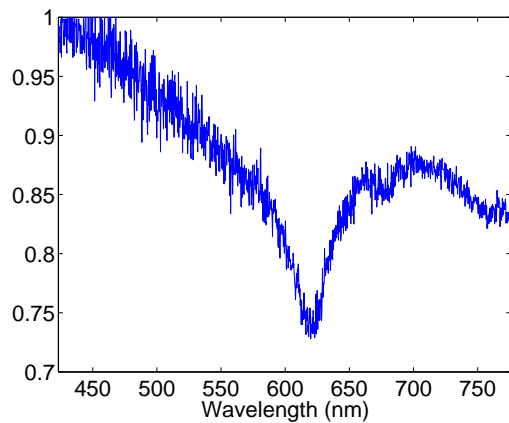
The measurement results are presented in Fig. 2 for six different nanoantenna samples. Many higher order modes show, especially for the sample with the shortest nanoantennas, and one of the modes becomes dominant with increasing length of the antennas. In addition, the spectra clearly show the resonance wavelengths moving towards the infrared with increasing antenna length, as can be expected based on classical antenna theory and effective wavelength studies for optical antennas.¹ Furthermore, the wavelength of the dominant resonance mode seems to have a linear relation with antenna length. The dimensions of nanoantennas were measured with a scanning electron microscope with a measurement error of ± 5 nm.



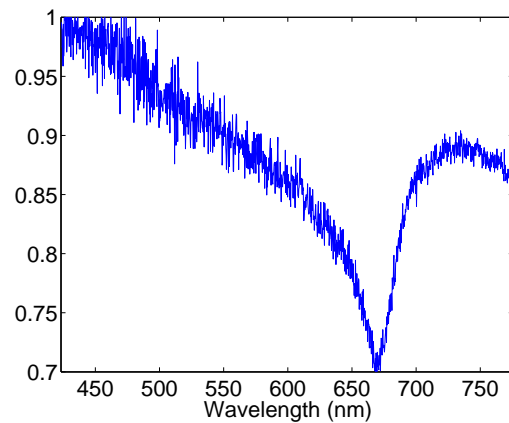
(a) 245x128 nm



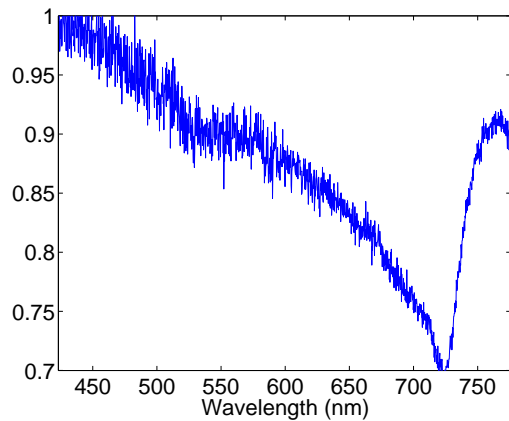
(b) 297x126 nm



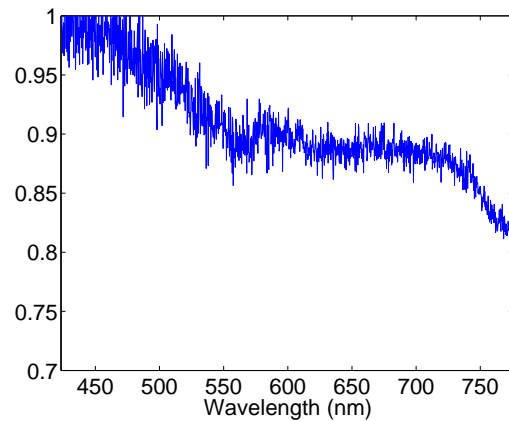
(c) 354x125 nm



(d) 396x118 nm



(e) 452x118 nm



(f) 511x116 nm

Figure 2. Transmission spectra for rectangular silver nanoantennas of different length, ranging from 245 nm to 511 nm with approximately 50 nm length steps. Every subfigure is labeled with the dimensions (length x width) of the corresponding nanoantennas. All the antennas have a height of 50 nm. In general, the spectra clearly show that the resonance wavelengths move towards the infrared with increasing length of the antennas.

246x128 nm

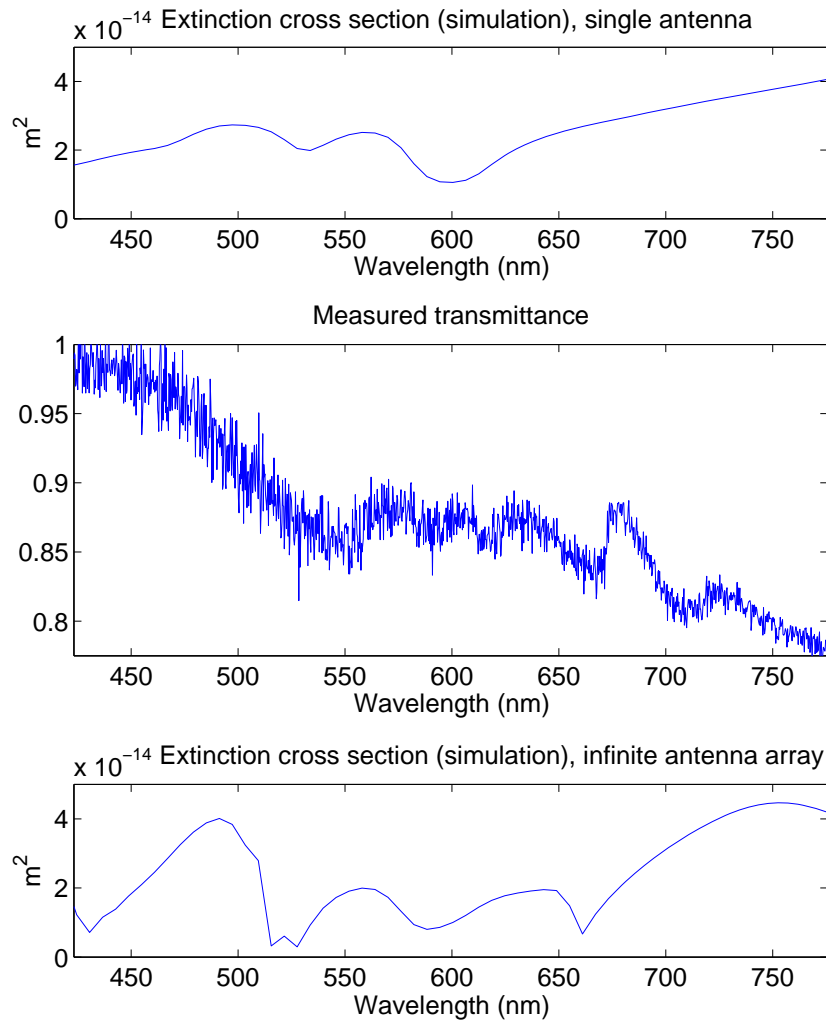


Figure 3. Simulated extinction cross section and measured transmittance for rectangular silver nanoantennas having a length of 246 nm, a width of 128 nm and a height of 50 nm. On the top: the simulation result for a single nanoantenna. In the middle: the measured transmittance. On the bottom: the simulation result for an infinite array of nanoantennas. The transmission spectrum and the extinction cross section spectrum are inversely related, such that peaks in the extinction cross section should correspond to dips in the measured transmittance. Note the different scales and units on the y-axes, as the transmittance is normalized and the cross section is reported in units of area. The simulation for the infinite array shows much better correspondence with the measurement results than the simulated response of a single antenna. However, there is a slight shift to the left, to shorter wavelengths.

The comparison of measurement results and simulations for a single nanoantenna and an infinite antenna array, for 246x128 and 452x118 nm antennas, are shown in Fig. 3 and 4, respectively. The transmission spectrum and the extinction cross section spectrum are roughly each other's inverse, such that peaks in the extinction cross section should correspond to dips in the measured transmittance. The curves obtained through simulation show a slight shift with respect to the measured ones, and for 245x128 nm antennas, some peaks are lacking. For 246x128 nm antennas, the infinite antenna array simulation shows significantly better correspondence with the measurement results than the single antenna model. However, the curve is slightly shifted to the left. In contrast, for longer 452x118 nm antennas, the single antenna simulation shows a better correspondence, even though the curve is slightly shifted to the right.

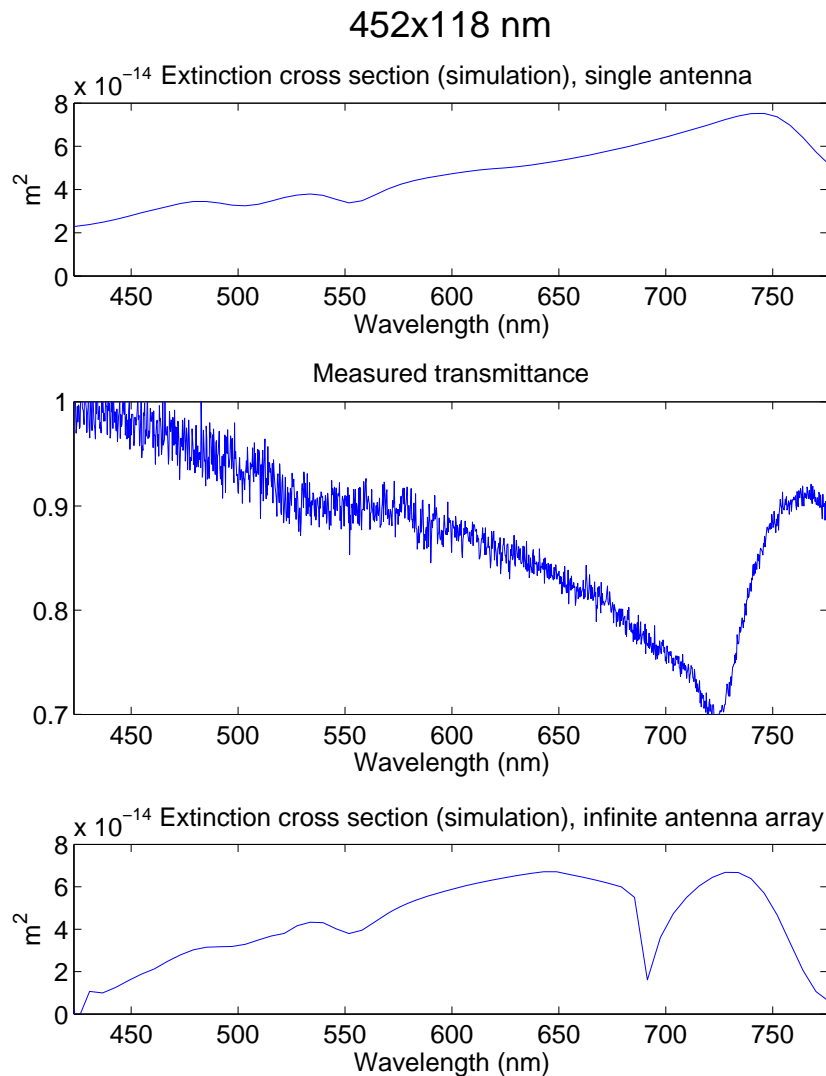


Figure 4. Simulated extinction cross section and measured transmittance for rectangular silver nanoantennas having a length of 452 nm, a width of 118 nm and a height of 50 nm. On the top: the simulation result for a single nanoantenna. In the middle: the measured transmittance. On the bottom: the simulation result for an infinite array of nanoantennas. The transmission spectrum and the extinction cross section spectrum are inversely related, such that peaks in the extinction cross section should correspond to dips in the measured transmittance. Note the different scales and units on the y-axes, as the transmittance is normalized and the cross section is reported in units of area. The single antenna simulation shows better correspondence with the measurement results, even though the curve is slightly shifted to the right, to longer wavelengths. A small peak in both simulation curves can be seen between 500 and 550 nm in accordance with the small dip in the measured transmittance.

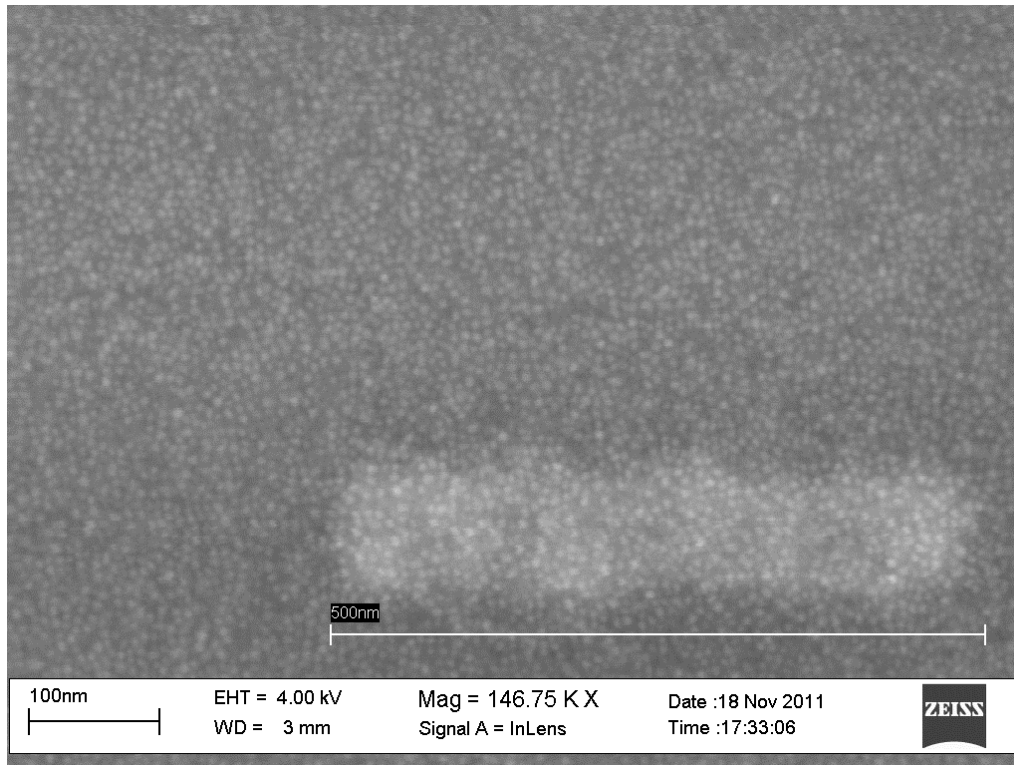


Figure 5. A uniform quantum dot layer on top of a nanoantenna structure. A single nanoantenna can be seen on the bottom of the image with lighter shade of grey. (SEM image)

4. TOWARDS STRONG COUPLING STUDIES

We added a quantum dot (QD) layer on top of the nanoantennas in order to study how the quantum dots change the resonance properties of the antennas, due to strong coupling between surface plasmon polaritons and the quantum dots. Fig. 5 shows a uniform QD layer applied onto the surface of a nanoantenna structure. The QD layer was prepared by spin coating a QD solution on the substrate after the lift off process. The quantum dots were dissolved in toluene. Measurements and simulations including the quantum dot layer are in progress.

5. CONCLUSIONS

We have presented experimental results of local surface plasmon resonances of silver nanoantennas. They show many higher order modes and the resonance wavelengths move to the infrared with increasing length of the antennas. We have compared results obtained by simulation with the measurements, which show a significant correspondence, except for a slight shift in the resonance wavelengths. A quantum dot layer was deposited on top of the nanoantenna structures to proceed with work on strong coupling studies.

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