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Publication date: 2017

Document Version: Publisher's PDF, also known as Version of record


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Optical reconfiguration and polarization control in semi-continuous gold films close to the percolation threshold


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1. Introduction

In this work we have studied the intrinsic and reconfigured optical properties of semi-continuous gold films, fabricated via a simple metal evaporation technique. We have prepared three films of nominal thicknesses 5, 6, and 7nm.

After fabrication the films are illuminated in areas by scanning a fs-pulsed laser over the films (Fig. 1). This results in permanent morphological changes in the films observed in a scanning transmission electron microscope (STEM), see Fig. 2. The laser writing also introduces a polarized feature in the transmission spectra of the films.

We have performed electron energy-loss spectroscopy (EELS) measurements and extensive finite-element simulations of our sample morphologies to better understand the origin of this polarization effect as well as the distribution of plasmonic resonances with and without laser writing.

From this we see that a strong dip in transmission appears when aligning the light source parallel to the writing laser. From this we infer that a strong dip in transmission appears when aligning the light source parallel to the writing laser.

During the transmission experiment it is possible for us to polarize the light source illuminating the sample, and we observe different regions, see Fig. 3.

We also see that the wavelength position of this dip depends on the power and wavelength of laser light used.

2. Optical spectroscopy

After illuminating the gold films with different laser powers we performed bright-field transmission spectroscopy on the different regions, see Fig. 3.

During the transmission experiment it is possible for us to polarize the light source illuminating the sample, and we observe different regions, see Fig. 3. From this we see that a strong dip in transmission appears when aligning the light source parallel to the writing laser.

We also see that the wavelength position of this dip depends on the power and wavelength of laser light used.

3. Hyperspectral images

To elucidate the origin of the polarization effect observed in Fig. 3, we have recorded hyperspectral images of our different sample morphologies using EELS, see Fig. 5.

Because of the fractal and self-similar nature of the films, a statistical representation of the image data is more succinct and easily comparable between samples. By a sequential filtering routine we can isolate the many different plasmon peaks found in the samples. We then sort them by central energy and peak EELS-intensity in histograms and probability density functions (PDFs), see Fig. 6.

By comparing the individual histograms and PDFs, we see that the particles aligned with the experimental polarization we also strongly polarized in their response.

As their polarization and resonance energy fit the features observed in the optical experiment (Fig. 3), we suggest that the polarization response of the gold film after illumination comes from these resonant particles formed by the photothermal processes.

4. Toy model description

To understand how the individual clusters and gaps of gold in the film morphologies are altered by the photothermal process of the laser, we assume that the photothermal evaporation of the metal particles can be described using a simple toy model of elongated resonant particles. To understand how these three processes influence the resonance of the particles, we have performed a set of different finite element simulations where the aspect ratios of the particles are altered, but all other parameters are assumed constant.

This simulates the melting and reshaping processes of the metallic particles if we assume minimal metal evaporation.

5. Polarization dependence

To visualize the particles responsible for the polarization response observed in the transmission experiment (Fig. 3), we plot the integrated EELS data from the 1.90-2.00eV range in which we see the transmission dip for the different samples.

From these maps we see several elongated particles that show EELS intensity distributions consistent with a longitudinal dipole mode predominantly aligned along the polarization used in the laser reconfiguration.

Because EELS does not provide us with a polarized excitation source, we perform simulations to recover the polarization dependence of the plasmon excitations.

6. Simulation geometry

To simulate our structures we utilize the already available microscopic images containing particle locations, and compute EELS intensity to make a thickness map of the metal in the samples. We then use the average particle thickness within its outline to map the particles as straight planes with varying heights in the simulation geometry, see Fig. 8.

7. Simulation results

We perform simulations of plane wave excitations on our constructed geometry. This allows us to choose the perpendicular and parallel polarization, and perform simulations of the distribution of electric field vectors observed in a dark-field microscope. Red arrows indicate polarization of the writing laser, and black arrows indicate polarization of the writing laser and black arrows indicate polarization of the writing laser and black arrows indicate polarization of the writing laser.

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From these maps we see several elongated particles that show EELS intensity distributions consistent with a longitudinal dipole mode predominantly aligned along the polarization used in the laser reconfiguration.

8. Conclusions

- Semi-continuous gold films fabricated by simple metal evaporation techniques can be locally altered by fs-pulsed laser illumination.
- This laser illumination creates elongated resonant particles that are aligned with the polarization of the laser used.
- The resonance of these particles can be controlled by using different film thicknesses, laser power, and laser wavelength.
- By this illumination it is possible to perform ‘grayscale’ plasmonic image printing using the films as writing medium.
- Locally tuning the resonance properties of the films could also open up new area to the percolation threshold.