Optical reconfiguration and polarization control in semicontinuous 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 7 nm. After fabrication the films are illuminated by scattering a f-pulsed laser over the film (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 two-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.

2. Optical spectroscopy

After illuminating the gold films with different laser powers we performed long-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 can align it either parallel or perpendicular to the polarization of the laser. This is usually used to reconfigure the gold films.

From this film we can see that the 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 successful 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.

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 illumination, we can construct a simple toy model of elongated resonant particle. We can imagine these processes for their photothermal evaporation:

- Particle shortening/polarization
  - Gap opening.
  - Gap closing/particle welding.

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 ratio of the particles are altered, but their volume is conserved. This simulates the melting and reshaping processes of the metal 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 1.90-2.00 eV range for which we see the transmission dip for the different 5 nm samples.

From these maps we can 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.

6. Simulation geometry

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

To simulate our structures we utilize the already available microscope images produced particle positions, and scan the 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 lines with varying heights in the simulation geometry, see Fig. 6.

7. Simulation results

We perform simulations of plane wave excitations on our constructed geometry. This allows us to choose the perpendicular z- and x-polarizations, aligned with the polarization used initially in the laser writing. We can then map the x-component of the excited fields from either of these excitations, or their sum.

For the two cases in Fig. 10 we get good agreement between the summed theoretical fields and the EELS data. When comparing the individual field components, 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 in the gold films after illumination comes from these resonant particles formed by the photothermal processes.

8. Conclusions

- Semi-continuous gold films fabricated by simple metal evaporation techniques can be locally altered by f-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 metal film thicknesses, laser power, and laser wavelength.
- By 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 processing applications for meta-material films.

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