Optical reconfiguration and polarization control in semicontinuous gold films close to the percolation threshold

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Publication date: 2017

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

Citation (APA):
Frydendahl, C., Repän, T., Geisler, M., Novikov, S. M., Beermann, J., Lavrinenko, A., ... Stenger, N. (2017). Optical reconfiguration and polarization control in semicontinuous gold films close to the percolation threshold. Poster session presented at 8th International Conference on Surface Plasmon Photonics, Taipei, Taiwan, Province of China.
Continuous gold films close to the percolation threshold

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

1. Introduction

In this work we have studied the intrinsic and reconfigurable 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 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.

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 orient the light source illuminating the area where the light can align either parallel or perpendicular to the polarization of the laser beam. We can use this to reconfigure the gold films.

From this we 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 particles. We can imagine the three processes for their photothermal reshaping:

- Particle shortening/merging periphery.
- 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 is altered, but the 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 1.90-2.00eV range in which we see a transmission dip for the different 5nm 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.

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 image of the particles, and compare the individual field components, we see that the particles aligned with the experimental polarization are also highlighted 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 and parallel polarizations, aligned with the polarization used initially in the laser writing. We can then map the 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 simulated theoretical fields and the EELS data. When comparing the individual field components, we see that the particles aligned with the experimental polarization are also strongly enhanced 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.

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 nominal film thicknesses, laser power, and laser wavelength.
- By this illumination it is possible to perform ‘grey-scale’ plasmonic image printing using the films as writing medium.
- Locally tuning the resonance properties of the films could also open up new writing applications for periodic metal films.

Work funded by:
- ERC, 341054 (PLAQNAP)
- KNAP, Grant Agreement 716716 (PLAQNAP)
- Aarhus University Foundation, Virtuosus scholarship
- DTU’s Foundation

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