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 7nm.

2. Optical spectroscopy

After fabrication the films are illuminated in areas by scanning a f-located 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 film.

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 film, a quantitative 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 and easily comparable between samples. By a sequential analysis we can isolate the many different plasmon peaks and easily comparable between 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.

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 pulse, we can construct a simple toy model of elongated resonant particles. We can imagine the three processes for their photothermal melting:

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

5. Polarization dependence

To visualize the particles responsible for the polarization responses observed in the transmission experiment (Fig. 3), we plot the integrated EELS data from the 1.90-2.00 eV range in which we see the transmission dip for the different 5nm samples.

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 microscopic images printed on a metal substrate. We then apply the EELS intensity to make a thickness map of the metal in the sample. 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. 5.

7. Simulation results

We perform simulations of plane wave excitations on our constructed geometry. This allows us to choose the perpendicular \( \vec{\alpha} \) and \( \vec{\beta} \) polarizations, aligned with the polarization used initially in the laser writing. We can then map the component of the excited field 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 also are strongly excited 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 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 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 open up new writing applications for plasmonic metal films.