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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.

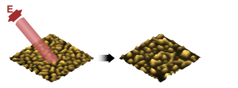

Fig. 1: Schematic overview of the laser illumination process.

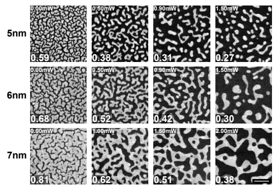

Fig. 2: STEM dark-field images of the different sample morphologies achieved with different laser powers. The scale bar is 150nm.

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 can align it either parallel or perpendicular to the polarization of the laser originally used to reconfigure the gold films.

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.

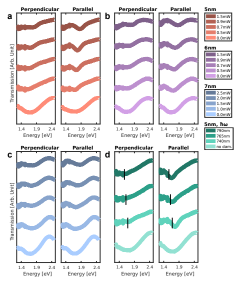

Fig. 3: Bright-field transmission spectra recorded through the different sample morphologies. (a-c) 740 nm laser used, (d) see legend, laser energy indicated by vertical line.

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 succint and easily comparable between samples. By a sequential fitting rutine 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.

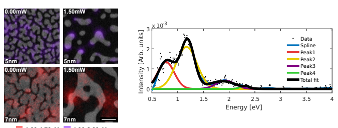

Fig. 5: Example EELS maps and example EEL spectrum and fits for statistical analysis. Scale bar is 75nm.

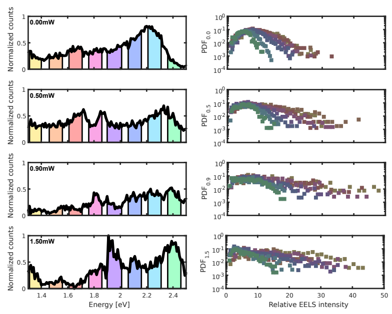

Fig. 6: Histograms of plasmon central energies from the 5nm samples, and PDFs of the EELS intensities from the same samples.

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 three processes for their photothermal reshaping:

- Particle shortening/spherification.
- 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 ratios of the particles are altered, but their volumes are conserved. This simulates the melting and reshaping processes of the metal particles if we assume minimal metal evaporation.

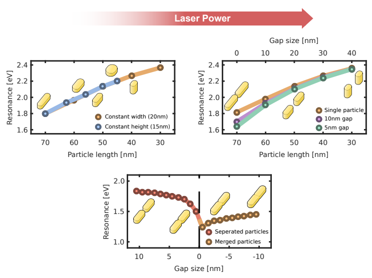

Fig. 7: Finite-element method simulations of the longitudinal plasmon modes for different regimes of particle pertubation from photothermal reshaping.

5. Polarization dependence

To visualize the particles responsible for the polarized response observed in the transmission experiment (Fig. 3), we plot the integrated EELS data from the 1.80-2.00eV range in which we see the 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.

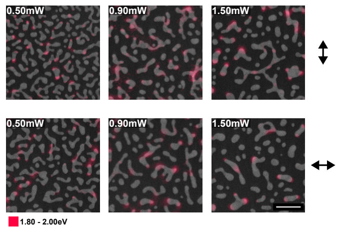

Fig. 8: Integrated EELS data mapped on top of the STEM dark-field images of the film morphologies, showing elongated particles with resonances near the features in the transmission spectra. The scale bar is 150nm.

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 to construct particle outlines, and use 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 prisms with varying heights in the simulation geometry, see Fig. 9.

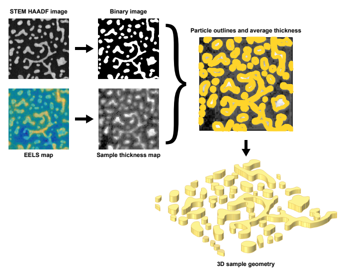

Fig. 9: Schematic overview of how the simulation geometry was generated from the experimental images.

7. Simulation results

We perform simulations of plane wave excitations on our constructed geometry. This allows us to choose the perpendicular *x*- and *y*-polarizations, aligned with the polarization used initially in the laser writing. We can then map the *z*-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 are 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 polarized response of the gold film after illumination comes from these resonant particles formed by the photothermal processes.

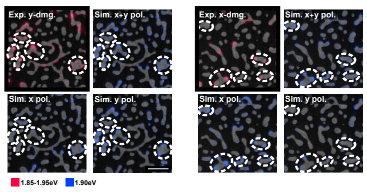

Fig. 10: Comparison between norm of simulated electric field *z*-components and EELS intensity for two different regions of the 5nm sample.

8. Conclusions

- Semi-continuous gold films fabricated by simple metal evaporation techniques can be locally altered via 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 initial 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 sensing applications for percolation metal films.

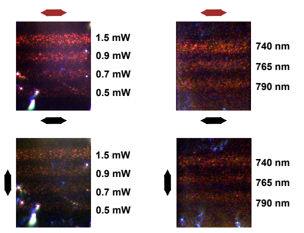

Fig. 11: Lines written in the 5nm gold film with different power and wavelength observed in a dark-field microscope. Red arrows indicate polarization of the writing laser, and black arrows the polarization of light used to record the image.

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