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

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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.

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 these processes for their photothermal nanomechanics:

- Particle shortening/localization
- 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 as 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 1.90-2.01eV 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.

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 printing the local particle density, and can print EELS intensity to make a thickness map of the metal in the sample. We then see the average particle thickness within its outline to map the particles as straight prisms 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 y- and x-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 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 polarized 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 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 metal film thicknesses, laser power, and laser wavelength.
- By this illumination it is possible to perform ‘graphical’ plasmonic image printing using the films as writing medium.
- Locally tuning the resonance properties of the films could also open up new promising applications for percolation metal films.

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