Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

DisSci-2020-00005: Quantum Fields on Driven Plasmonic Nanostructures Yr2020 Review

Project Team: Patrick O'Rourke, Lindsay Roy, Simona Murph, Robert Lascola, Michael Thomas

Introduction

Energy absorbed by a system is eventually distributed throughout the system according the laws of thermodynamics. However thermodynamics does not address the mechanisms whereby a non-equilibrium state approaches equilibrium. Understanding the quantum mechanical mechanisms responsible for energy redistribution are complicated by the huge number of kinetic and electromagnetic interactions involved.

It may be possible to separate kinetic and electromagnetic interactions responsible for energy redistribution in plasmonic arrays because the array elements are fixed in space and only interact electromagnetically. In addition the array elements can be arranged in one- and two-dimensional geometries greatly simplifying simulations and experimental interpretation.

This work explores electromagnetic radiation coupling to plasmonic nano-particles, and its redistribution throughout a symmetric mesoscale nanostructure of the plasmonic particles. We hope to identify coherent EM oscillations associated with groups of nano-particles that could be interpreted as quasi-particles, and seek to stabilize the quasi-particles by introducing asymmetric 'traps' in the structure.

Methods

Computer simulations:
Time dependent density functional (TD-DFT) and time-dependent density functional tight binding (TD-DFTB) calculations to simulate EM field interactions with individual Au19 and Au67 nano-particles.

Finite-Difference Time-Domain (FDTD) to simulate EM near-field dynamics arising from 1D chains of Au NPs with 100 nm diameter and interparticle distance of 15 nm.

Experiments:
Focused ion beam milling of nanostructures in gold and silver films deposited on prism surfaces.

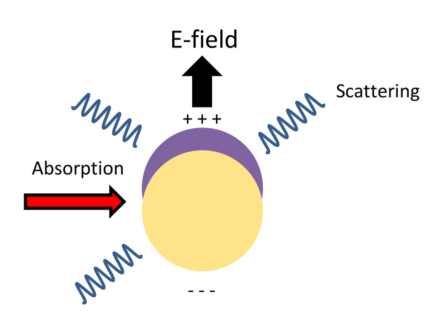
Evanescent wave coupling of EM fields to nano-structures.

Observation of EM absorbance and scattering from nano-structures.
- Polarization effects
- Propagation vector
- Surface enhanced scattering
- Tip enhanced scattering

Acknowledgement

Text or graphics

Simulations indicate anomalous optical resonances for nano-particles aligned with beam propagation



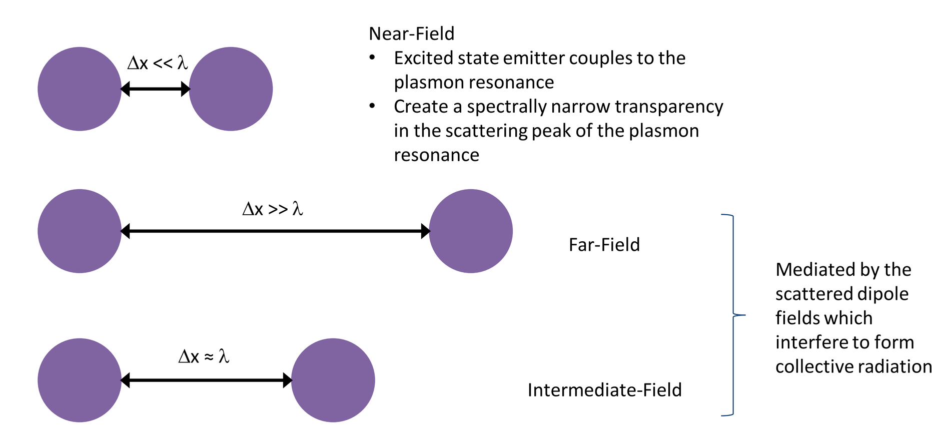
Coherent collective oscillation of electrons of a metallic nanoparticle

For a finite-sized particle, the near-field at a distance *r* from the particle (along a direction parallel to the incident light polarization) is given by an expansion of all possible multipolar modes:

$$E_{nf} = \frac{2\alpha E_0}{4\pi\varepsilon_0 r^3} + \frac{3\beta \dot{E}_0}{4\pi\varepsilon_0 r^4} + \frac{4\gamma \ddot{E}_0}{4\pi\varepsilon_0 r^5}$$

Where α, β, γ,…are, respectively, the dipole, quadrupole, octupole,…. Polarizability tensors of the particle. The smaller the distance r (relative to the particle size) where the field needs to be estimated, the higher is the number of terms that needs to be included in the near-field expansion.

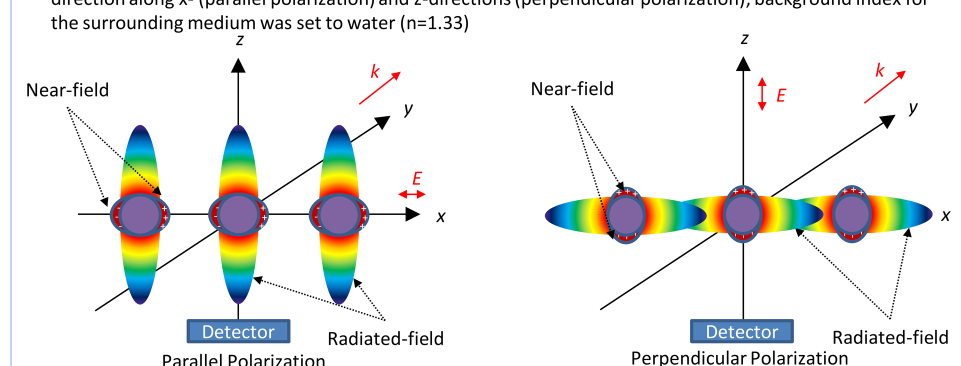
Interparticle interactions between metal nanoparticles



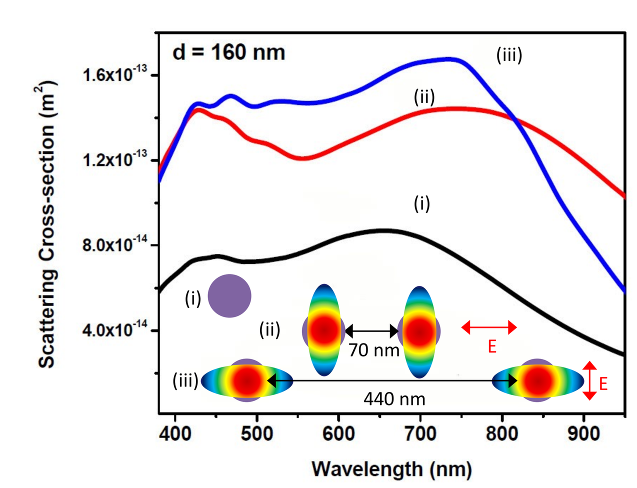
Question: Can you tune plasmonic properties of arrays of AuNPs by exploiting dipolar interactions at the intermediate-scale regime?

Electrodynamic Simulations

Software: MEEP – Open source finite-difference time-domain (FDTD) simulation software package developed at MIT
- Dielectric function for gold was determined using a two-term Drude-Lorentz model fit to the dielectric function measured by Johnson and Christy (Phys. Rev. B, 6, 4370, 1972).
- Method: 1D lattice illuminated by a linearly polarized plane wave with λ = 400-950 nm and polarization direction along x- (parallel polarization) and z-directions (perpendicular polarization); background index for the surrounding medium was set to water (n=1.33)

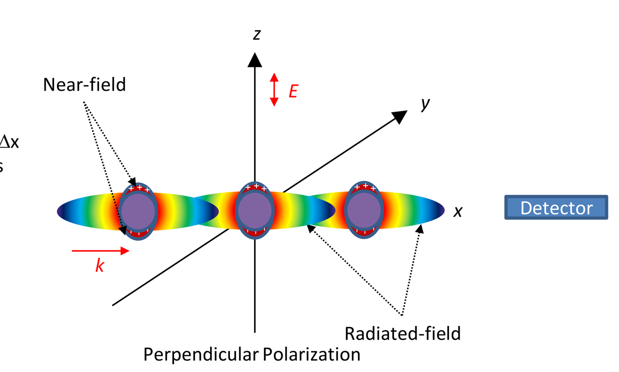


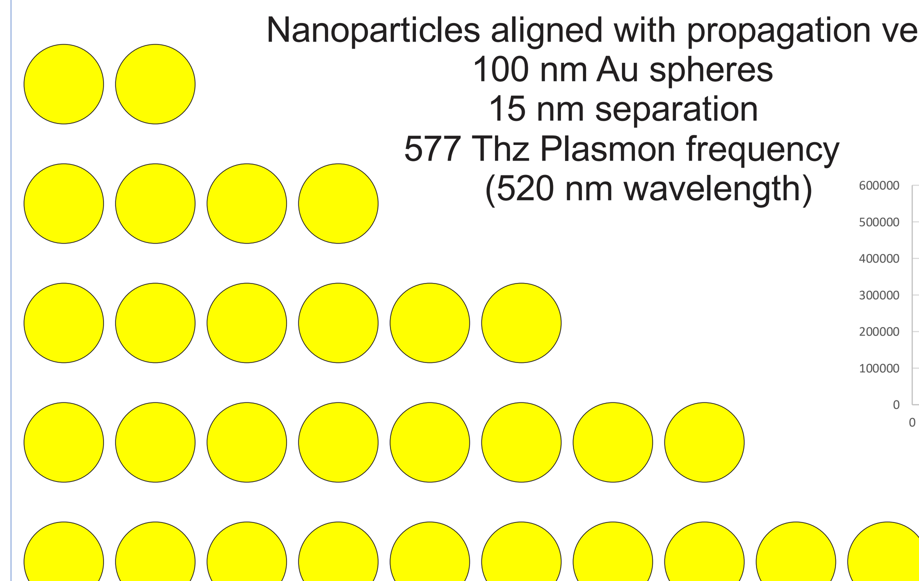
(Very) Preliminary Data



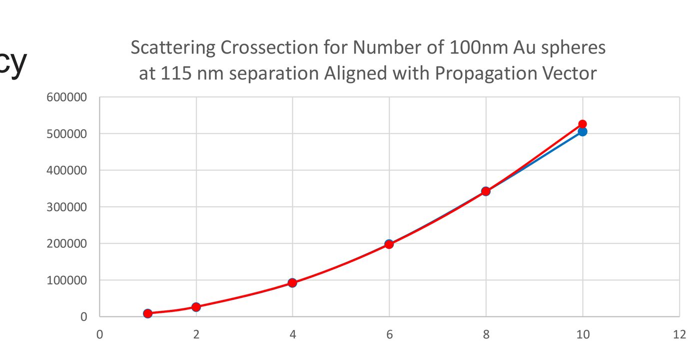
The magnitude of the red-shift for Δx = 230 nm for parallel polarization is equivalent to the red-shift for Δx = 600 nm for perpendicular polarization

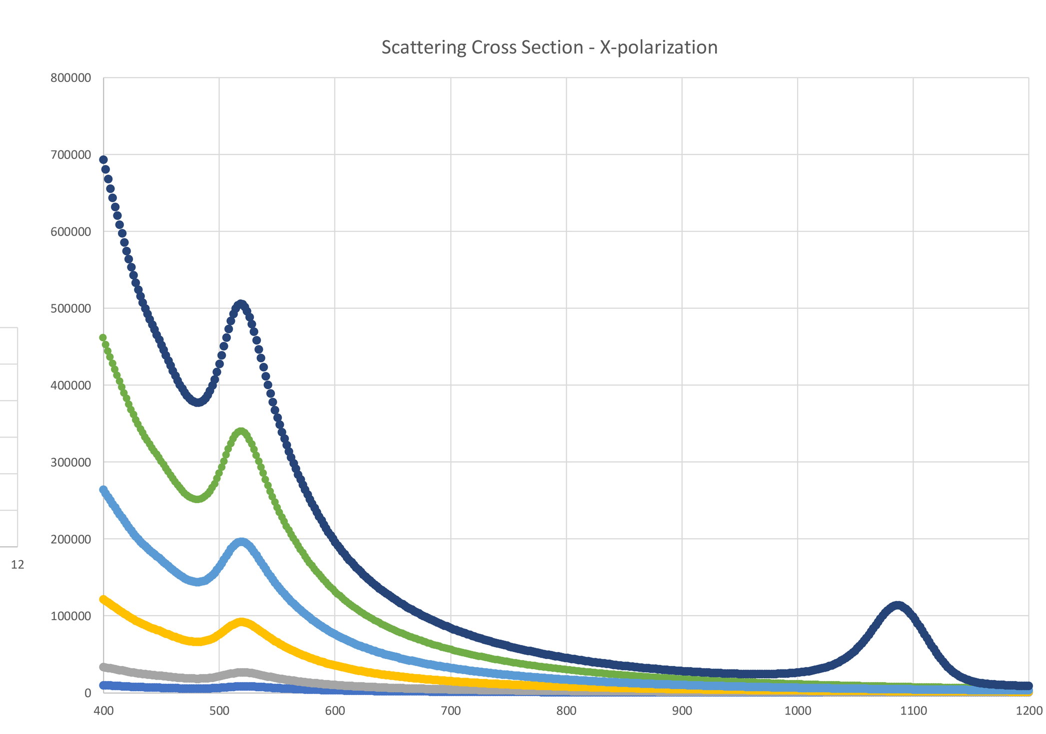
Change Propagation Direction to colinear with nanoparticle structure





Nanoparticles aligned with propagation vector
100 nm Au spheres
15 nm separation
577 Thz Plasmon frequency
(520 nm wavelength)

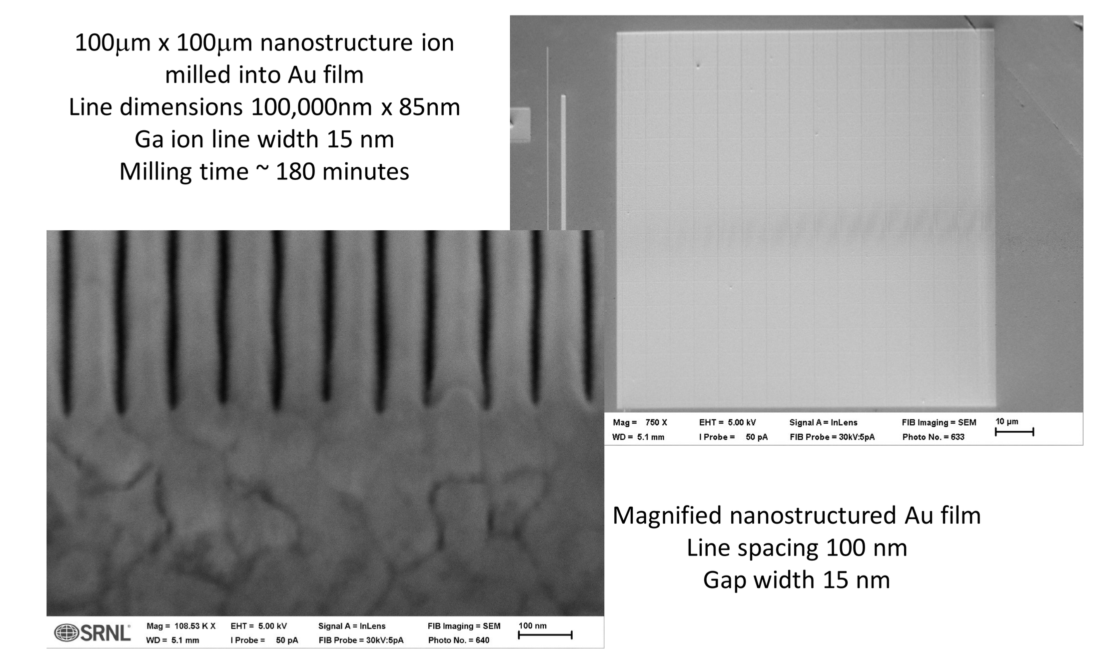




Results

Ga Ion Milled Nanostructure in Au Film

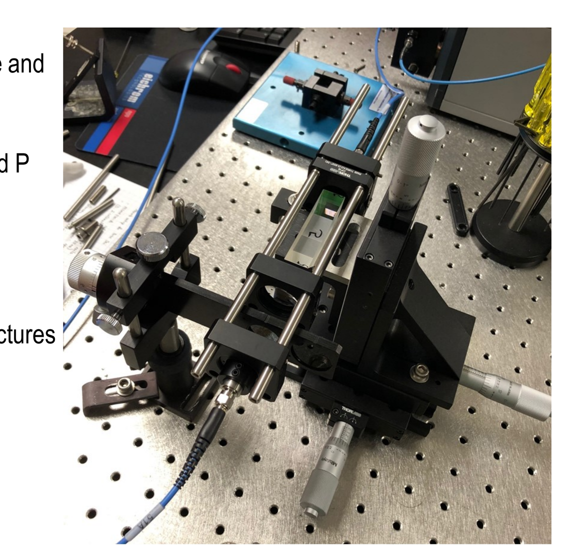
100µm x 100µm nanostructure ion milled into Au film
Line dimensions 100,000nm x 85nm
Ga ion line width 15 nm
Milling time ~ 180 minutes



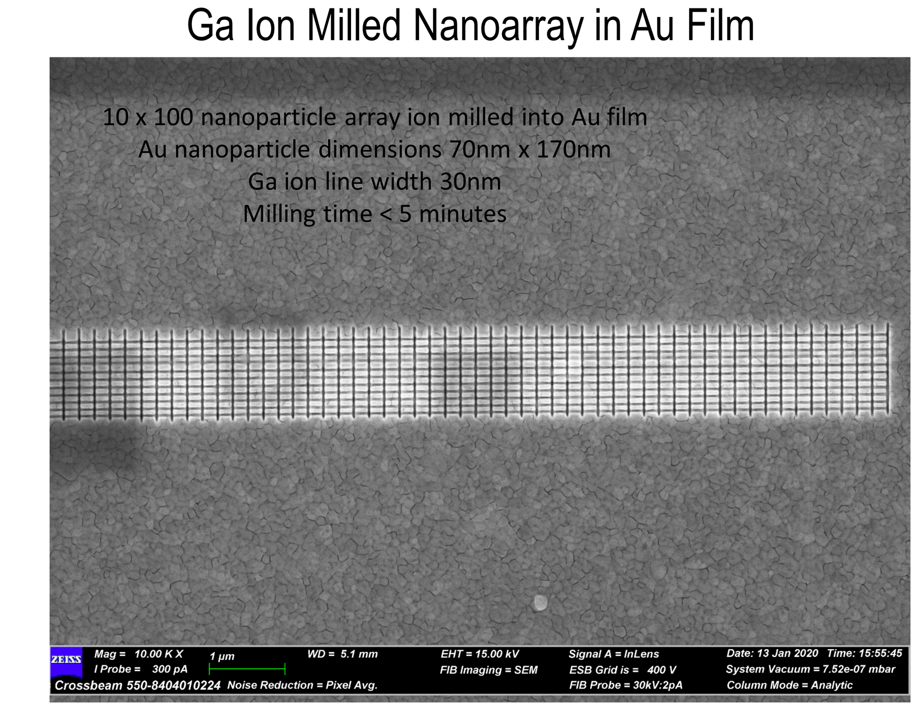
Magnified nanostructured Au film
Line spacing 100 nm
Gap width 15 nm

Optical assembly to Investigate 2-D Nanostructures

- Dove prism used to control interaction between metal-dielectric nano-structure and optical electro-magnetic fields
- Prism orientation selects between S and P polarization
- Beam angle selects incident condition
- Prism can support 5 separate nanostructures
- Can observe reflected light and angle resolved scattered light
- CW and pulsed optical sources
- High-resolution wide-range spectrophotometer



Ga Ion Milled Nanoarray in Au Film



Find out more about this research.