Spatial squeezing of plasmonic modes on disordered metallic films

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Decoding the title

• Scattering of visible (or near IR) light by disordered metallic samples

• Peculiar optical response due to interplay between material resonances (plasmons) and multiple scattering

• Address the spatial localization of modes
  LDOS fluctuations
  Spatial coherence
The colors of gold

Semi-continuous gold films on a glass substrate

\[
f = 30\% \quad 49\% \quad 67\% \quad 79\% \quad 82\% \quad 89\% \quad 99\%
\]

V.M. Shalaev, Nonlinear Optics of Random Media (Springer, 2000)
Low surface fraction

$f = 30\%$

Particle surface-plasmon resonance

$\text{Re} \varepsilon(\omega_{sp}) = -2$
Continuous film

Extended surface-plasmon

f = 99%
Intermediate regime: Giant intensity fluctuations

Localized and delocalized modes

Localized Luminous
$F_i = 0.07$

Delocalized Luminous
$F_i = 0.2$

Delocalized Dark
$F_i \approx 10^{-9}$

Localized Dark
$F_i \approx 10^{-9}$

« Inhomogeneous localization »

Stockman, Faleev, Bergman, PRL 87, 167401 (2001)
Near-field speckle correlations show different regimes

SNOM measurements of near-field intensity
Seal, Sarychev, Noh, Genov, Yamilov, Shalaev, Ying, Cao, PRL 95, 226101 (2005)
Outline

- LDOS fluctuations and localized plasmonic modes
- Radiative versus non-radiative modes
- Spatial coherence and the extent of plasmonic modes
LDOS fluctuations and localized plasmonic modes

Radiative versus non-radiative modes

Spatial coherence and the extent of plasmonic modes
Probing LDOS in optics: Fluorescence decay rate

Probability of being excited at time $t$ 

$P(t) \approx \exp(-\Gamma t)$

Spontaneous decay rate 

$\Gamma = \frac{1}{\tau}$

Perturbation theory

$\Gamma = \frac{2}{\hbar} \mu_0 \omega^2 \left| \mathbf{p}_{ge} \right|^2 \text{Im} \left[ \mathbf{u} \cdot \mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega) \mathbf{u} \right]$ 


Particular case: VACUUM

$\Gamma_0 = \frac{\omega^3}{3\pi \varepsilon_0 \hbar c^3} \left| \mathbf{p}_{ge} \right|^2$ 

(Einstein coefficient for spontaneous emission)
Decay rate and LDOS

\[ \Gamma = \frac{2}{\hbar} \mu_0 \omega^2 \left| p_{ge} \right|^2 \text{Im} \left[ \mathbf{u} \cdot \mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega) \mathbf{u} \right] \]

is also very often written as
(Fermi’s golden rule)

\[ \Gamma = \frac{\pi \omega}{\varepsilon_0 \hbar} \left| p_{ge} \right|^2 \rho_u(\mathbf{r}_0, \omega) \]

Local Density of States (LDOS)

\[ \rho_u(\mathbf{r}_0, \omega) = \frac{2 \omega}{\pi c^2} \text{Im} \left[ \mathbf{u} \cdot \mathbf{G}(\mathbf{r}_0, \mathbf{r}_0, \omega) \mathbf{u} \right] \]

\[ \frac{\Gamma}{\Gamma_0} = \text{change in the LDOS} \]

Changes in spontaneous decay rate (lifetime)
probe changes in the LDOS
Probing LDOS distributions on disordered metal films

Statistical distributions of $\Gamma$ (LDOS)

- $f = 30\%$
- $f = 82\%$

$\lambda = 605$ nm

Decay rate $\Gamma$ (ns$^{-1}$)

Occurrences

Fluorescence decay

$t$ (ns)

$\Gamma = 0.40$ ns$^{-1}$
$\Gamma = 0.24$ ns$^{-1}$
LDOS fluctuations

\[
\frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1
\]

Fractal and Euclidian clusters

\( f = 82\% \)
The peak reveals spatially localized modes

\[ \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} - 1 \]

The peak in the LDOS reveals spatially localized plasmon modes

Qualitative analysis (inverse participation ratio)

Mode extent \( \xi \)

\[ \frac{1}{S} \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} \approx \frac{1}{\xi^2} \]

Krachmalnicoff, Castanié, De Wilde, Carminati, PRL 105, 183901 (2010)
LDOS fluctuations and localized plasmonic modes

Radiative versus non-radiative modes

Spatial coherence and the extent of plasmonic modes
Numerical simulations (1)

Generating synthetic films

$f = 20\%$

$f = 50\%$

$f = 75\%$

Euclidian and fractal clusters

Numerical simulations (2)

Calculated LDOS maps (volume integral method)

Gold
\( \lambda = 780 \text{ nm} \)

\[
E(r) = E_0(r) + \frac{\omega^2}{c^2} \int [\epsilon(r', \omega) - 1] G_0(r, r', \omega) E(r') d^3 r'
\]

Distance dependence of LDOS distributions

**Experiment**

\[ \frac{\Gamma}{\Gamma_0} \]

**Numerical simulations**

\[ \text{Occurrences} \]

**Normalized decay rate**

\[ \lambda = 607 \text{ nm} \]
Non-radiative modes dominate at short distance

\[ \Gamma = \Gamma_{NR} + \Gamma_R \]

\[ \langle \Gamma/\Gamma_0 \rangle \]

\[ \text{Var}(\Gamma/\Gamma_0) \]

This is confirmed by calculated LDOS maps

$$\rho = \rho_{NR} + \rho_{R}$$
Estimating the size of hot spots

\[ K \approx \frac{2\pi}{\Delta x} \gg \frac{2\pi}{\lambda} \]

Evanescent modes \( \exp(-Kz) \)

Decay length \( 1/K \approx 10 \text{ nm} \)
\( \Delta x \approx 60 \text{ nm} \)

LDOS fluctuations and localized plasmonic modes

Radiative versus non-radiative modes

Spatial coherence and the extent of plasmonic modes
Beyond LDOS - spatial coherence

- The LDOS describes the number of modes contributing at a given point.
- How could we describe the connection between two points?

LDOS $\rho/\rho_0$

Cross Density Of States (CDOS)

- Density Of States (DOS)
  \[ \rho(\omega) = \sum_n \delta(\omega - \omega_n) \]

- Local Density Of States (LDOS)
  \[ \rho(r, \omega) = \sum_n |e_n(r)|^2 \delta(\omega - \omega_n) \]
  \[ \rho(r, \omega) = \frac{2\omega}{\pi c^2} \text{Im} [\text{Tr} G(r, r, \omega)] \]

- Cross Density Of States (CDOS)
  - Connecting two points at frequency \( \omega \)
  \[ \rho(r, r', \omega) = \sum_n e_n(r) \cdot e^*_n(r') \delta(\omega - \omega_n) \]

- CDOS in terms of Green's function
  \[ \rho(r, r', \omega) = \frac{2\omega}{\pi c^2} \text{Im} [\text{Tr} G(r, r', \omega)] \]
Ex: spatial coherence of thermal surface plasmons

Field spatial correlation

\[ \rho = |\mathbf{r} - \mathbf{r}'| \]

\[ \left\langle E_i(\mathbf{r}, \omega) E_j^*(\mathbf{r}', \omega) \right\rangle \propto \text{Im} G_{ij}(\mathbf{r}, \mathbf{r}', \omega) \]

Rytov, Kravtsov and Tatarskii, 

CDOS describes spatial squeezing of plasmonic modes

Topography

LDOS

CDOS

Gold $\lambda = 780$ nm

The width of the CDOS defines the spatial coherence length.

The coherence length describes the overall spatial squeezing of eigenmodes.
• Disordered (fractal) metallic films combine interesting physics and ease of fabrication

• Spatial coherence approach describes the overall spatial squeezing of plasmonic modes in the resonant regime (consistent with "inhomogeneous localization" picture)

• Wideband resonant system
  Large (> µm) or reduced (<100 nm) spatial coherence
  Radiative and non-radiative modes
  Large Purcell factors

  2D platform for basic nanophotonics experiments
Near-field speckle fluctuations

Near-field intensity fluctuations around the percolation threshold

Seal et al., PRL 97, 206103 (2006)

\[
\frac{\langle I^2 \rangle}{\langle I \rangle^2} - 1
\]

\[\lambda = 633 \text{ nm}\]

\[\Delta p = p - p_c\]
Near-field intensity: A strongly fluctuating pattern

Photo Electron Emission Microscopy (PEEM)

Qualitative analysis - Inverse participation ratio

Inverse participation ratio measures spatial extent of eigenmodes

\[ R_{IP} = \frac{\int |E(r)|^4 \, d^2r}{\left( \int |E(r)|^2 \, d^2r \right)^2} \]

Example: Localized mode on area A with constant amplitude

\[ R_{IP} = \frac{1}{A} \]

Assumption: At a given point and given frequency, only one mode contributes

\[ \rho(\omega, r) \propto \sum_n |E_n(r)|^2 \delta(\omega - \omega_n) \approx \frac{|E(r)|^2}{\Delta \omega} \]

LDOS fluctuations measure \( R_{IP} \)
and therefore the spatial extent of plasmon modes

\[ \frac{1}{S} \frac{\langle \rho^2 \rangle}{\langle \rho \rangle^2} = R_{IP} \approx \frac{1}{\xi^2} \]
Thermal-radiation STM maps plasmon interferences

Y. De Wilde (ESPCI)