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Radiative and non-radiative local density of states on disordered plasmonic films

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Abstract

We present numerical calculations of the local density of optical states (LDOS) in the near field of disordered plasmonic films. The calculations are based on an integral volume method, that takes into account polarization and retardation effects, and allows us to discriminate radiative and non-radiative contributions to the LDOS. At short distance, the LDOS fluctuations are dominated by non-radiative channels, showing that changes in the spontaneous dynamics of dipole emitters are driven by non-radiative coupling to plasmon modes. Maps of radiative and non-radiative LDOS exhibit strong fluctuations, but with substantially different spatial distributions.

Keywords: Metallic films; Plasmons; Fractals; Numerical simulations; Local density of states; Disordered systems

1. Introduction

Disordered plasmonic films obtained by evaporating noble metals on a substrate are known to exhibit unusual optical properties [1]. Close to the percolation threshold, metallic clusters with fractal perimeters leads to the emergence of subwavelength areas supporting enhanced electric field, commonly called hot spots [2]. These randomly distributed localized fields turned out to be very promising for sensing [3,4], subwavelength focusing [5], or non-linear optics [6]. Although several theoretical and numerical works have been reported on the subject, the question of the local density of optical states (LDOS) has been hardly addressed.

It has been known for long that the decay rate of a fluorescent emitter depends on its electromagnetic environment [7,8], the dependence being described by the LDOS $\rho(\mathbf{r}, \omega)$, with $\mathbf{r}$ the location of the emitter and ω the emission frequency. Indeed, the lifetime τ of the excited state of a dipole emitter with transition dipole $\mathbf{p}$ is given in perturbation theory by $1/\tau = \pi\omega|\mathbf{p}|^2\rho(\mathbf{r}, \omega)/(3\epsilon_0 \hbar)$ where ϵ_0 is the vacuum permittivity and $\hbar$ the reduced Planck constant. Thus the LDOS can be directly probed experimentally by measuring τ. In a disordered medium, changes in the LDOS probe the local environment [9–11], the photon transport regime [12,13] or drive long-range correlations of speckle patterns [14,15]. Recently, LDOS statistics in the vicinity of disordered films have been studied experimentally [16]. Enhanced LDOS fluctuations have been observed close to the percolation threshold, in a regime where the film morphology is controlled by fractal clusters. These enhanced fluctuations have been qualitatively associated to localized plasmon modes.

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Theoretical and numerical studies of semi-continuous disordered metallic films are very often based on approximations, such as mean-field theories [17] or quasi-static calculations [6,18]. An exact numerical approach has been reported recently using a FDTD (finite-difference time-domain) scheme [19].

In this paper, we present numerical calculations of the LDOS in the vicinity of disordered metallic films based on an integral volume method. This exact formulation is limited only by the discretization of the films into finite size cells. The numerical algorithm is divided into two steps. Firstly, we use a Monte–Carlo algorithm to simulate the growth of a gold film under an evaporation/deposition process, and check that the geometrical properties of the film near the percolation threshold are in good agreement with experimental observations. Secondly, we solve Maxwell's equations in 3D, taking into account polarization and retardation effects, which allows us to compute maps and statistical distributions of the LDOS. The computations are in agreement with known experimental results. The approach allows us to split the LDOS into its radiative and non-radiative contributions, and to discuss their relative contributions to the spatial fluctuations of the LDOS, which is the main focus of this work.

2. Numerical approach

2.1. Generation of disordered films

Our first goal is to generate numerically disordered metallic films that have the same properties as the experimental evaporated metallic films. To do so, we use a kinetic Monte–Carlo algorithm, as proposed in [20]. The idea is to randomly deposit 5 nm large gold particles on a square grid via an iterative algorithm, and let the particles diffuse under the influence of an interaction potential until a stable geometry is reached. At every iteration of the algorithm, we randomly choose either to deposit a new particle (probability p_0) or to make a particle on the grid jump to a more stable neighbour site (probability $p_{i\to j}$ to scatter from site i to site j). Using the normalization $p_0 + \sum_{i,j\neq i} p_{i\to j} = 1$, we only need to pick a random number out of [0, 1] to determine the relative weight of each process. More precisely, the probability to deposit a particle reads $p_0 = NF$, where N is the number of particles that remains to be deposited in order to reach the prescribed filling fraction, and F is a constant (with dimension s^{-1}) modeling the experimental deposition rate. The probability for a particle located on site i to jump to the neighbor site j reads $p_{i\to j} = \exp[-\Delta E_{i\to j}/(k_B T)]$, where k_B is the Boltzmann constant, T the temperature of the surface and $\Delta E_{i\to j}$ the activation energy barrier. Computing $\Delta E_{i\to j}$ is a complex issue for atoms [21,22], and is not possible from first principles for nanometer size particles. In the present approach, we have chosen to deal with a rescaled atomic potential that renormalizes the energy barrier in order to apply to a nanoparticle. We assume that $\Delta E_{i\to j} = \alpha(E_i - E_j)$, where α is a positive dimensionless adjustable parameter taking into account the influence of the substrate and the scaling. E_i is the rescaled ''atomic'' potential of a particle located on site i, which is allowed to jump to the neighbor site j if $E_i > E_j$. This potential is given by the following expression based on a tight-binding second moment method [23]:

$$E_i = A\sum_{i\neq j}\exp[-p(r_{ij}/r_0 - 1)] - B\left\{\sum_{i\neq j}\exp[-2q(r_{ij}/r_0 - 1)]\right\}^{1/2}. \quad (1)$$

In this expression, r_0 is the size of one particle, r_{ij} the distance between two sites i and j and A, B, p and q are constants that were tabulated for atoms [23]. The iterative deposition process is stopped when all particles have been deposited (so that the prescribed filling fraction has been reached) and no particle can move to a more stable site.

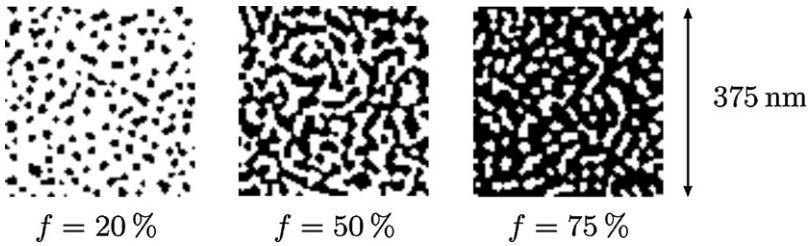

Fig. 1. Numerically generated gold films for three different filling fractions f (gold is represented in dark). The parameters for the computation are: $T = 300$ K, $\alpha = 2.58 \cdot 10^{-2}$, $F = 10^{14}$ s^{-1}, $A = 0.2061$ eV, $B = 1.79$ eV, $p = 10.229$, $q = 4.036$.