Cross Density Of States and spatial coherence in complex plasmonic systems

Alexandre Cazé, Romain Pierrat and Rémi Carminati

Institut Langevin, ESPCI Paristech, CNRS 1, rue Jussieu, 75001 Paris, France alexandre.caze@espci.fr

Abstract

The Local Density Of States (LDOS) is a fundamental quantity for the understanding of light-matter interaction. It can be accessed experimentally by measuring the spontaneous decay rate of a fluorescent emitter. A good picture to understand this connection is that the stronger the modes available at the emitter position, the faster the latter decays.

Theoretically, the LDOS can be defined independently of the mode structure, using the Green function ${\bf G}$

$$\rho(\mathbf{r},\omega) = \frac{2\omega}{\pi c^2} \operatorname{Im} \left[\operatorname{Tr} \mathbf{G}(\mathbf{r},\mathbf{r},\omega) \right].$$

In this theoretical and numerical work, we introduce a new quantity that we call Cross Density Of States (CDOS), defined by analogy with the LDOS as

$$\rho(\mathbf{r}, \mathbf{r}', \omega) = \frac{2\omega}{\pi c^2} \operatorname{Im} \left[\operatorname{Tr} \mathbf{G}(\mathbf{r}, \mathbf{r}', \omega) \right].$$

Whereas the LDOS contains information on the average strength of the modes at one position \mathbf{r} , the CDOS counts the average strength of all modes that *coexist* at both positions \mathbf{r} and \mathbf{r} '. As a consequence, the CDOS contains an information on the spatial coherence due to the mode structure. More precisely, it allows us to define rigorously an intrinsic coherence length at any point \mathbf{r} of a complex structure.

To illustrate the interest of this new quantity, we have focused on the case of disordered semicontinuous gold films. These structures are known to support unconventional plasmon modes that, in particular, can localize optical energy in subwavelength areas (hot spots) [1]. It has been previously shown experimentally in our group that light localization on these systems could be addressed considering the statistical properties of the local density of optical states (LDOS) [2]. A correlation between enhanced fluctuations of the LDOS near the percolation threshold of the film and a reduction of the spatial extent of plasmon modes has been established.

We have developed a numerical method based on the Lippmann-Schwinger equation that allows us to compute with no approximation the Green function in the near-field of three-dimensional disordered semi-continuous films. Our calculations take into account polarization, retardation and near-field interactions. They give us access to both the LDOS and the CDOS in the near field of these films.

Our numerical results exhibit the same enhanced LDOS fluctuations as in experiments [3]. Moreover, we are able to compute CDOS maps and to observe rigorously an overall squeezing of the optical modes near the percolation threshold (Fig. 1) [4]. This clarifies a basic issue in plasmonics concerning the description of the optical properties of these films. This illustrates the relevance of the CDOS in the study of spatial coherence in photonics and plasmonics systems, and more generally in wave physics.

References

- [1] S. Grésillon et al. Phys. Rev. Lett. 82 (1999) 4520.
- [2] V. Krachmalnicoff, E. Castanié, Y. De Wilde and R. Carminati, Phys. Rev. Lett. 105 (2012) 183901.
- [3] A. Cazé, R. Pierrat and R. Carminati, Photon. Nanostr. Fundam. Appl. 10 (2012) 339.
- [4] A. Cazé, R. Pierrat and R. Carminati, Phys. Rev. Lett, 110 (2013) 063903.

Figures



Fig. 1 - (a,b) Two numerical realizations of disordered gold films, with respective filling fractions f=20% and f=50%; (c,d) LDOS maps at distance d=40nm over the top of films (a) and (b); (e,f) CDOS maps on the same areas with one spatial dependance **r** fixed at the center of the film. Both LDOS and CDOS maps are normalized by the LDOS in vacuum. Wavelength is 780nm.