Light emission statistics in correlated random photonic nanostructures

N. de Sousa[1], J. J. Saenz[1], A. García-Martín[2] and L. S. Froufe-Pérez[3]

[1] Departamento de Física de la Materia Condensada,
Universidad Autónoma de Madrid, 28049 Madrid, Spain

[2] Instituto de Microelectrónica de Madrid, CSIC,
Isaac Newton 8, Tres Cantos, 28760 Madrid, Spain

[3] Instituto de Ciencia de Materiales de Madrid, CSIC,
Campus de Cantoblanco, 28049 Madrid, Spain

nuno.teixieira@uam.es

The statistical properties of light transport and emission in disordered media has been a matter of intense research during the last century. Being the basis of coherent multiple scattering of waves well known, the phenomenon itself is not yet fully explored and understood. These multiple wave scattering effects are at the heart of emerging behaviors like Anderson localization of light and electrons, band structure in crystalline solids or photonic crystals (PhC), among many others.

Although the limits of perfectly ordered systems on the one hand, and uncorrelated and relatively weakly scattering systems on the other hand, are quite well understood. There is a gap between both limits which is largely unexplored. In particular, it has been shown in many different situations that disordered systems exhibiting certain structural correlations can share properties of both crystalline and fully disordered systems. For instance, the conductivity of liquid metals [1] or the cornea transparency [2] can be understood in the same footing: a disordered but correlated system can present spectral regions of high transparency for electron or light transport.

The effects of disorder in an initially ordered structure, such as a PhC, might lead to strong Anderson localization, as the scattering mean free path can be severely reduced in the band edges [3]. Also, strongly correlated charged colloids can scatter light in such a way that the transport mean free path presents a strong chromatic dispersion [4]. Even in the absence of practically any long range correlations, the structure of the scatterers itself can be used to modify the light emission and transport properties of a disordered system in such a way that transport parameters [5], or even the threshold of a random laser [6], can present resonances which can be tuned in advance.

The effect of correlations in a disordered structure regarding light emission properties of single fluorescent emitter has been a matter of much less intense research efforts. It is clear that the structure surrounding a single emitter can largely alter its emission dynamics [7]. In the last years, several groups considered such effects in a statistical way suitable for the description of disordered systems [7,8,9]. In particular, in ref.[9] it was shown that several structural properties near a phase transition can be accessed via fluorescence intensity fluctuations.

Is has been theoretically proven that near field scattering in random systems alters fluorescence dynamics in such a way that microscopic information about the surroundings of a single emitter can be obtained from lifetime fluctuations or from the shape of the statistical distribution tails [10,11].

In this presentation, we theoretically show how, in the previous context, fluorescence emission rate statistics are largely altered due to the appearance of structural correlations in a disordered system.

We have developed a model of point resonant interacting scatterers which are placed at random. Emission dynamics of a single emitter is calculated for each sample of an ensemble of structural realizations of the system.

While keeping constant the scattering properties of single scatterers, the global geometry, and scatterers density, the structural correlations are controlled changing the temperature of the interacting set of scatterers.

It is shown that fluorescence decay rate statistics of a the single emitter correlates with the structural phase transitions of the system. In the low temperature limit, the structure freezes in an face center cubic lattice. This structure presents a gap (frequency range of low photonic density of states) corresponding to a vanishing fluorescence decay rate. As usual, it also presents narrow frequency windows of high density of states, corresponding to band edges of the perfect infinite crystalline structure, leading to high decay rates.

At frequencies corresponding to both a band gap and a band edge, we perform decay rate statistics varying the temperature of the system. It is shown that, at low temperature, decay rates hardly fluctuates and its average value corresponds to the crystalline one. On temperature raising, fluctuations of decay rate grow, and the averaged values undergoes a relatively sharp transition to a different value. This transition can be identified with a structural phase transition in the system.

References

[1] N. W. Aschcroft and J. Lekner, Phys. Rev. **145** (1966) 84.
[2] R. W. Hart and R. A. Farrell, J. Opt. Soc. Am. **59** (1969) 766.
[3] Sajeev John, Phys. Rev. Lett. **58** (1987) 2486.
[4] L. F. Rojas-Ochoa, J. M. Mendez-Alcaraz, J. J. Sáenz, P. Schurtenberger, and F. Scheffold, Phys. Rev. Lett. **93** (2004) 073903.
[5] P. D. García, R. Sapienza, A. Blanco, and C. López, Adv. Mater. **19,** 2597 (2007); R. Sapienza, P.D. García, J. Bertolotti, M.D. Martín, A. Blanco, L. Viña and C. López, D.S. Wiersma, Phys. Rev. Lett. **99**, (2007) 233902.
[6] S. Gottardo, R. Sapienza, P.D. Garcia, A. Blanco, D. S. Wiersma and C. Lopez, Nat. Phot. **2** (2008) 429.
[7] Jordi Hernando, Erik M. H. P. van Dijk, Jacob P. Hoogenboom, Juan-José García-López, David N. Reinhoudt, Mercedes Crego-Calama, María F. García-Parajó, and Niek F. van Hulst, Phys. Rev. Lett. **97** (2006) 216403.
[8] H. Gersen, M. F. García-Parajó, L. Novotny, J. A. Veerman, L. Kuipers, and N. F. van Hulst, Phys. Rev. Lett. **85** (2000) 5312.
[9] R. A. L. Vallee, M. Van der Auweraer W. Paul and K. Binder, Phys. Rev. Lett. **97** (2006) 217801.
[10] L. S. Froufe-Pérez, R. Carminati and J. J. Sáenz, Phys. Rev. A **76** (2007) 013835.
[11] L. S. Froufe-Pérez and R. Carminati, Phys. Stat. Sol. (a) **205** (2008) 1258.