Effect of long range spatial correlations on the lifetime statistics of an emitter in a two-dimensional disordered lattice

N. de Sousa, J. J. Sáenz, A. García-Martín, L. S. Froufe-Pérez, and M. I. Marqués

1Departamento de Física de la Materia Condensada, Universidad Autónoma de Madrid, 28049, Madrid, Spain.
2Condensed Matter Physics Center (IFIMAC) and Instituto “Nicolás Cabrera”, Universidad Autónoma de Madrid, 28049, Madrid, Spain.
3Donostia International Physics Center (DIPC), Paseo Manuel de Lardizabal 5, Donostia-San Sebastian 20018, Spain.
4IMM-Instituto de Microelectrónica de Madrid (CNM-CSIC), Isaac Newton 8, PTM, Tres Cantos, E-28760 Madrid, Spain.
5Instituto de Estructura de la Materia, (IEM-CSIC), Serrano 121, 28006 Madrid, Spain.
6Departamento de Física de Materiales, Universidad Autónoma de Madrid, 28049, Madrid, Spain.

The effect of spatial correlations on the Purcell effect in a bidimensional dispersion of resonant nanoparticles is analyzed. We perform extensive calculations of the fluorescence decay rate of a point emitter embedded in a system of nanoparticles statistically distributed according to a simple 2D lattice-gas model near the critical point. For short range correlations (high temperature thermalization) the Purcell factors present a non-Gaussian long-tailed statistics which evolves towards a bimodal distribution as approaching the critical point where the spatial correlation length diverges. Our results suggest long range correlations as a possible origin of the large fluctuations of experimental decay rates in disordered metal films.

Since Purcell work [1], it is known that lifetime of an excited atomic state is a combination of the atom properties and the environment where it is embedded. Changes in the emission decay rate in regular structures have been reported for emitters placed close to planar interfaces [2–5], cavities [6], photonic crystals [7], plasmonic [8–9] and magnetoplasmonic structures [10], among others. More recently, the possibility of creating nanostructured materials stimulated the interest of waves propagation through disordered media. Examples like backscattering enhancement [11–12], photon localization [13] random lasing [14, 15], or photonic membranes [16] can be found in the literature. In complex systems like liquids, colloids, granular or biological materials, the dynamical modification of the environment or the movement of the emitter imply the need for a statistical study of the decay rate [17, 18]. In these random systems the decay rate exhibits a non Gaussian, long tailed distribution, where large enhancements of the Purcell factor are attributed to strong fluctuations of the local density of states induced by the near field scattering [14-21]. These rare events create optical modes confined in a small volume around the source.

In most cases previously studied disorder was produced in a random way, that is, scatterers were distributed throughout the lattice randomly. Real systems, however, can be realized with other kinds of disorder, where the locations of the dipoles are correlated. In particular, structural disorder with long range correlation (LRC) has been found for instance in X-ray and Neutron Critical Scattering experiments in systems undergoing magnetic and structural phase transition [22, 23]. This correlation effect in magnetic systems maybe modeled by assuming a spatial distribution of critical temperatures with a correlation function obeying a slow power law decay [24, 25]. Spatial correlations in disordered scattering materials has been shown to dramatically modify the light transport properties, in particular, light scattering mean free path presents strong chromatic dispersion [26, 29].

In the present paper, long range correlated distributions of scatterers are produced using a thermal order-disorder distribution governed by a characteristic ordering temperature (θ) [31, 32]. In particular, the quenched randomness is implemented using a lattice-gas model equivalent to a thermally diluted ferromagnetic two dimensional (2D) Ising lattice at a temperature (θ). We perform extensive calculations of the fluorescence decay rate of a point emitter embedded in a system of nanoparticles statistically distributed according to a 2D lattice-gas model near the critical point. As we will show, for short range correlations (high temperature thermalization) the Purcell factors present a non-Gaussian long-tailed statistics where events with large Purcell factors are extremely rare. Interestingly, as we approach the critical point where the spatial correlation range diverges, the statistics evolves towards a bimodal distribution with a well defined peak at high enhancement factors. Our numerical results strongly resemble those obtained experimentally in resonant metallic thin films near percolation [20], which suggest long range correlations as a possible origin of the large fluctuations of experimental decay rates in disordered metal films.

Let us consider the lattice system sketched in Fig. 1. The nodes of this lattice are taken as the possible location points of the optical dipoles by taking into account the following mechanism: After thermalization of the pure

* manuel.marques@uam.es
and oriented out of plane (see Fig. 1). The emitter is placed in the central position of the lattice and the particles in the system are scatterers in resonance, meaning that $\alpha(\omega) = \frac{i\omega}{\omega^2 + \Gamma^2}$, where $\omega$ is the angular frequency of the incident wave. In the presence of a dipole emitter $\mathbf{p}(r)$ the electric field at some position $r'$ can be obtained by operating the Green tensor over the dipole positioned at $r$. Mathematically, this is expressed as:

$$E(r') = \frac{k^2}{\epsilon_0} G_0(r', r) \cdot \mathbf{p}(r).$$  \hspace{1cm} (2)

The Green tensor is given by $[33]$:

$$G_0(r, r') = \frac{e^{ikR}}{4\pi R} \left[ \left( 1 + i\frac{kR - 1}{k^2 R^2} \right) \mathbf{I} + \left( \frac{3 - 3i kR - k^2 R^2}{k^2 R^2} \right) \mathbf{R} \otimes \mathbf{R} \right],$$  \hspace{1cm} (3)

where $R$ is the modulus of the vector $\mathbf{R} = r - r'$ and $\mathbf{R} \otimes \mathbf{R}$ denotes the outer product of $\mathbf{R} = \mathbf{R}/R$ by itself.

When the emitter is in the presence of $N$ dipole scatterers, the scattering field at position $r'$ is given by:

$$E(r') = \frac{k^2}{\epsilon_0} G_0(r, r') \mathbf{p}(r) + \frac{k^2}{\epsilon_0} \sum_{m=1}^{N} G_0(r, r_m) \mathbf{p}_m,$$  \hspace{1cm} (4)

where $r_m$ is the position of the $m$th scatterer and $\mathbf{p}_m = \epsilon_0 \mathbf{E}(\mathbf{r}_m)$ is the value of the induced dipole located at $\mathbf{r}_m$. To obtain the value of all induced dipoles we should solve the electric fields in Eq. 4 by considering the coupled dipole method $[33]$. The second term on the right-hand side of Eq. 4 represents the modification of the free space dyadic Green function due to the presence of the scatterers. The scattering field is then given by:

$$\mathbf{E}_s(r') = \frac{k^2 \alpha}{\epsilon_0} \sum_{m=1}^{N} G_0(r', r_m) \mathbf{E}(\mathbf{r}_m).$$  \hspace{1cm} (5)

Once the scattering field is known, the normalized spontaneous decay rate $\Gamma$ of a dipole $\mathbf{p}(r')$, in the weak coupling regime, is given by $[34]$

$$\frac{\Gamma}{\Gamma_0} = 1 + \frac{6\pi \epsilon_0}{|\mathbf{p}|^2 k^3} \Im[\mathbf{p}^* \cdot \mathbf{E}_s(r)],$$  \hspace{1cm} (6)

being $\Gamma_0$ the decay rate of the emitter in free space.

First, we analyse the full crystalline configuration and we calculate the normalized decay rate of the emitter, positioned in the center of the structure, as a function of the ratio between the lattice parameter $a$ and the resonance wavelength considered $\lambda = \frac{a}{\alpha}$. We fix the wavelength of the emitter and we vary the particles position in the lattice.

Results presented in Fig. 2 for a system with lateral dimension $D = 23$ show how it is possible to identify a maximum at $\frac{a}{\lambda} = 0.44$ with value $\Gamma/\Gamma_0 \sim 7$.

FIG. 1. Schematic representation of a crystalline structure with an edge $(D)$ of 7 particles. The transparent spheres represent the removed particles. The dipole emitter is represented by the blue sphere and the scatterers by red spheres.
FIG. 2. Normalized decay rate of a crystalline structure for a system with edge of 23 particles (total system with 528 particles). The maximum can be found at $\frac{\theta}{T_c} = 0.44$.

Next, we fix the lattice parameter to $a = 0.44\lambda$ and we analyze the decay rate distribution for disordered systems generated at different ordering temperatures ranging from $\theta >> T_c$ ($\xi \to 0$) corresponding with a short range correlated disorder to $\theta = T_c$ ($\xi \to \infty$), corresponding with a long range correlated distribution of the vacancies. Results, for $10^6$ different configurations, are shown in Fig. 3.

When the ordering temperature is large ($\theta \sim 2T_c$) we obtain a long tailed distribution centred at $\Gamma/\Gamma_0 \sim 1.3$, where some rare events are detected for values high as $\Gamma/\Gamma_0 \sim 7$. Similar results have been previously reported [18, 21]. However, as the ordering temperature decreases towards $T_c$, the correlation function between vacancies changes from an exponential decay to a power law and the decay rate distribution re-shapes dramatically. For $\theta = 1.001T_c$, there is no longer any long tailed behavior, but a bimodal distribution where the previously reported rare events increase considerably to build a new maximum centred at $\Gamma/\Gamma_0 \sim 7$.

We have also analysed the possible presence of finite size effects by considering different lateral sizes ranging from $D = 13$ to $D = 63$ for $\theta = T_c$. Results are shown in Fig. 4. Note how all secondary maxima between $\Gamma/\Gamma_0 \sim 1$ and $\Gamma/\Gamma_0 \sim 7$ tend to smear out, while maxima at $\Gamma/\Gamma_0 \sim 1$ and $\Gamma/\Gamma_0 \sim 7$ are reinforced when the lateral size increases.

Large Purcell factors are due to optical modes confined around the source and are sustained by near field interactions [21] so, systems with strong correlations, like the ones reported in this paper, should promote an increase on the number of configurations where the emitting dipole is surrounded by clusters of dipoles allowing for field confinement. To further analyze this idea we have plotted the histogram of the frequency of occupation for each position normalized to the number of configurations, for different values of the decay rate. We have focused our attention in $\Gamma/\Gamma_0 = 1.31 \pm 0.02$, corresponding to the maximum of the distribution for $\theta = 2T_c$ and $\Gamma_0 = 0.98 \pm 0.02$, $6.88 \pm 0.02$, corresponding to the two main maxima of the distribution for $\theta = T_c$. Results are shown in Fig. 5.

For $\Gamma_0 = 1.31$ we detect no special patterns, and dipoles are distributed almost randomly. However, for $\Gamma_0 = 6.88$ (corresponding to the second maxima for $\theta = T_c$) the dipoles distribution changes markedly. In this case the emitting particle is clearly surrounded by scattering dipoles, allowing for a confinement of the optical modes. This is in agreement with the attribution proposed in ref. [21] and clearly shows how large Purcell factors are boosted by long range correlations in the disordered sample. Interestingly, for the other maxima located at $\Gamma_0 = 0.98$ the situation is just the opposite and the emitting dipole is, in average, surrounded by vacancies where no field confinement is possible, entailing a dipole’s response similar to the one found in vacuum. This last effect is also fostered by the existence of long
range correlations when $\theta = T_c$.

In order to understand these effects it is important to take into account that, at criticality, clusters of all sizes, containing either dipoles or vacancies, exist on the system and the response due to larger clusters resembles the one found for the crystalline structure $\Gamma_0 \sim 7$. However, when a non-correlated distribution of vacancies is considered, the correlation length and the cluster’s sizes are very small, and the detection of events with a large Purcell factor turns out to be very unlikely. The surface structure of the clusters in the Ising model for long range correlation, i.e. at criticality, is known to be fractal and scale invariant [36], like the clusters obtained for high filling factors in semicontinuous metal films experiments [20]. These structures are responsible for surface plasmon localization leading to a large increase in the decay rates.

In conclusion, the normalized fluorescent decay rate distribution has been analyzed in a thermally disordered two dimensional diluted dipole lattice where the correlation between vacancies may be tuned at will. When the ordering temperature is far from criticality, the correlation length is small, and the decay rate distribution has the typical long tailed shape where events with large Purcell factors are extremely rare. However, when the ordering temperature is close to criticality, the correlation length tends to infinity, turning the decay rate function into a bimodal distribution where large Purcell factor events are much more probable. Our analysis shows how diluted systems, where vacancies are distributed in a long range manner, enclose clusters of dipoles of all sizes, allowing for the existence of many configurations with optical modes confined around the source.
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