Spontaneous decay rate of a dipole emitter in a strongly scattering disordered environment

R. Pierrat* and R. Carminati†
Institut Langevin, ESPCI ParisTech, CNRS UMR 7587, 10 rue Vauquelin, F-75231 Paris Cedex 05, France
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We study the statistics of the fluorescence decay rate of a dipole emitter embedded in a strongly scattering medium. In the multiple-scattering regime, the probability of observing a decrease in the decay rate is substantial, as predicted by exact numerical simulations. The decrease originates from a reduction of the local density of optical states and is due to collective interactions and interferences. In the strong-scattering regime, signatures of recurrent scattering are visible in the behavior of the average decay rate.

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I. INTRODUCTION

Optics in disordered media is a field of growing interest, stimulated by the development of imaging techniques in turbid media [1], by the possibility of studying fundamental questions in mesoscopic physics using optical waves [2], and by the emergence of randomly structured photonic materials with unconventional properties [3–5]. The local density of optical states (LDOS) is a fundamental quantity that drives the spontaneous emission of light as well as macroscopic transport properties [6–10]. LDOS fluctuations are also connected to intensity correlations in speckle patterns through the so-called C_0 correlation [11]. Understanding the (statistical) behavior of the LDOS in disordered systems is important; for example, to engineer photonic materials controlling light emission and propagation by multiple scattering [3–5,8,10], or to improve fluorescence lifetime imaging techniques [12].

In this paper, we study the spontaneous decay rate Γ of a dipole emitter (e.g., an atom, molecule, or quantum dot) considered as a probe of the LDOS in a three-dimensional (3D) disordered cluster of localized scatterers. The decay-rate distributions in the single-scattering regime were studied previously [13,14], and the fluctuations were explained in terms of near-field dipole-dipole interactions. Here, we compute the statistical distribution $P(\Gamma)$, using exact 3D numerical simulations, in the (strong) multiple-scattering regime. We show that the distributions are much broader and exhibit a different line shape. In particular, we show that the probability of observing decay rates smaller than the free-space decay rate Γ_0 is substantial. This corresponds to inhibition of spontaneous emission (reduction of the LDOS) by multiple scattering.

II. THEORETICAL MODEL

We consider a 3D cluster of N resonant point scatterers randomly distributed inside a sphere with radius R, with a dipole emitter located at the center (position $\mathbf{r}_0$). The geometry is shown in the inset of Fig. 1. It has been chosen, without loss of generality, to be spherically symmetric on average so that averaged quantities can be calculated analytically. The emitter is surrounded by an exclusion volume with radius R_0. A minimum distance d_0 is forced between the point scatterers, so that one can define an effective volume fraction $f = N(d_0/2)^3/(R^3 - R_0^3)$. In the numerical simulations, we have kept $f \leqslant 2\%$ so that correlations in the position of the scatterers remain negligible. The scatterers are described by their polarizability $\alpha(\omega) = -3\pi c^3\gamma/[\omega^3(\omega - \omega_0 + i\gamma/2)]$ where ω_0 is the resonance frequency, γ is the linewidth, and c is the speed of light in vacuum. This corresponds to the polarizability of a resonant nonabsorbing point scatterer, as a two-level atom far from saturation. In order to reach the multiple-scattering regime with a dilute system and a relatively small number of scatterers ($N \leqslant 10^3$), we assume an emission frequency ω_0, so that the scatterers are on resonance. The scattering cross section is $\sigma_s(\omega_0) = (k_0^4/6\pi)|\alpha(\omega_0)|^2 \sim \lambda_0^2$, where λ_0 is the wavelength in vacuum and $k_0 = \omega_0/c = 2\pi/\lambda_0$. The parameter $k_0\ell_B$ measures the scattering strength, where $\ell_B = [\rho\sigma_s(\omega_0)]^{-1}$ is the independent-scattering (or Boltzmann) mean-free path and $\rho = N/V$ is the density of scatterers. On resonance, one has $k_0\ell_B = [\rho|\alpha(\omega_0)|]^{-1}$. In all the numerical computations that follow, we have taken the following set of parameters: $\omega_0 = 10^{15}$ Hz, $\lambda_0 = 1.88$ μm, $R_0 = 0.36$ μm, and $d_0 = 0.12$ μm. The density $\rho = N/V$ is the adjustable parameter that allows us to vary $k_0\ell_B$ in the range $1 \lesssim k_0\ell_B \simeq 10$.

In the weak-coupling regime, the spontaneous decay rate of a dipole emitter takes the form

$$\Gamma_{\mathbf{u}} = \frac{2}{\hbar}\mu_0\omega_0^2|\mathbf{p}|^2 \mathrm{I}m[\mathbf{u}\cdot\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)\cdot\mathbf{u}], \quad (1)$$

where $\mathbf{p} = p\mathbf{u}$ is the transition dipole and $\mathbf{u}$ is a unit vector [15]. The dyadic Green function $\mathbf{G}$ connects the electric dipole at position $\mathbf{r}_0$ to the radiated electric field at position $\mathbf{r}$ through the relation $\mathbf{E}(\mathbf{r}) = \mu_0\omega_0^2\mathbf{G}(\mathbf{r},\mathbf{r}_0,\omega_0)\cdot\mathbf{p}$. In free space, the decay rate is obtained from the vacuum Green function $\mathbf{G}_0$, and reads $\Gamma_0 = \omega^3|\mathbf{p}|^2/(3\pi\epsilon_0\hbar c^3)$. In the present study, we consider the decay rate Γ averaged over the transition dipole orientation $\mathbf{u}$:

$$\Gamma = \frac{2}{3\hbar}\mu_0\omega_0^2|\mathbf{p}|^2 \mathrm{Im}[\mathrm{Tr}\ \mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)], \quad (2)$$

where Tr denotes the trace of a tensor. Note that the statistical properties of Γ may be very different from that of $\Gamma_{\mathbf{u}}$ [14] since the decay rate is strongly orientation dependent [16]. The decay rate Γ is proportional to the electric-field contribution to the LDOS [17]:

$$\rho(\omega_0,\mathbf{r}_0) = \frac{2\omega_0}{\pi c^2}\mathrm{Im}[\mathrm{Tr}\ \mathbf{G}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)], \quad (3)$$

so that measurements of Γ gives direct access to the LDOS.

*romain.pierrat@espci.fr
†remi.carminati@espci.fr

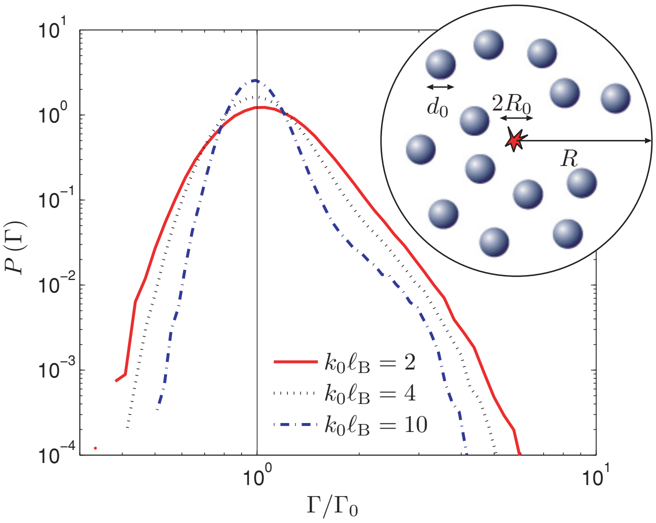

FIG. 1. (Color online) Statistical distribution of the decay rate Γ for three different scattering strengths. For $k_0\ell_B = 2$ (dense systems), $R = 4.96$ μm and $\ell_B = 0.6$ μm. For $k_0\ell_B = 4$, $R = 6.24$ μm and $\ell_B = 1.2$ μm. For $k_0\ell_B = 10$ (dilute systems), $R = 8.47$ μm and $\ell_B = 3.0$ μm. The radius R in each case is large enough to avoid finite-size effects. The calculations are performed with $N = 500$ scatterers and 1 080 000 configurations. Inset: schematic view of the system.

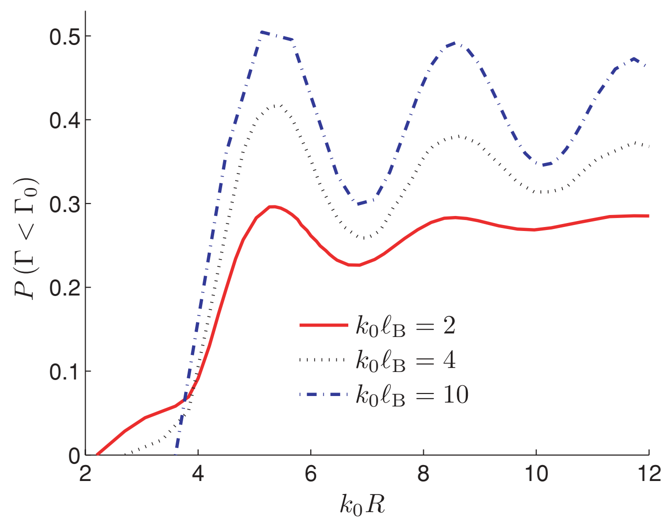

FIG. 2. (Color online) Probability density $P(\Gamma < \Gamma_0)$ versus the normalized size $k_0 R$ of the cluster, deduced from distributions similar to those in Fig. 1, computed for different sizes R of the cluster. The end point $k_0 R = 12$ corresponds to $R/\ell_B = \{6,3,1.2\}$ in the three situations $k_0\ell_B = \{2,4,10\}$, respectively.

The calculation of the statistical distribution of Γ amounts to calculating the Green function $\mathbf{G}(\mathbf{r},\mathbf{r}_0,\omega_0) = \mathbf{G}_0(\mathbf{r},\mathbf{r}_0,\omega_0) + \mathbf{S}(\mathbf{r},\mathbf{r}_0,\omega_0)$ for an ensemble of realizations of the scattering medium. Since $\mathbf{G}_0$ is known analytically, we only need to calculate the Green function $\mathbf{S}(\mathbf{r},\mathbf{r}_0,\omega_0)$ which corresponds to the scattered field. To proceed, we perform a coupled-dipole numerical computation. The field-exciting scatterer j is given by the contribution of the dipole source and of all other scatterers, leading to a set of $3N$ self-consistent equations [18]:

$$\mathbf{E}_j = \mu_0\omega_0^2\mathbf{G}_0(\mathbf{r}_j,\mathbf{r}_0)\mathrm{p} + \alpha(\omega_0)k_0^2\sum_{\substack{k=1\\k\neq j}}^{N}\mathbf{G}_0(\mathbf{r}_j,\mathbf{r}_k)\mathbf{E}_k, \qquad (4)$$

where $\mathbf{r}_j$ is the position of scatterer j and the dependence of the Green functions on ω_0 have been omitted. This linear system is solved numerically for each configuration of the disordered medium. Once the exciting electric field at each scatterer is known, it is possible to compute the scattered field at the source position $\mathbf{r}_0$ and deduce the Green dyadic $\mathbf{S}(\mathbf{r}_0,\mathbf{r}_0,\omega_0)$, from which Γ is readily obtained. In this numerical approach, near-field and far-field dipole-dipole interactions and multiple scattering are rigorously taken into account.

III. NUMERICAL RESULTS

We show in Fig. 1 the statistical distribution of the decay rate $P(\Gamma)$ for three different densities of scatterers, corresponding to a multiple-scattering regime with $2 \leqslant k_0\ell_B \leqslant 10$. We first observe that the curves exhibit broad distributions, with values of Γ/Γ_0 ranging from 0.38 to 6 or even more. This corresponds to much larger fluctuations than those observed in the single-scattering regime, where Γ/Γ_0 deviates only slightly from unity [13,14]. Interestingly, the computations also show that the probability of having $\Gamma < \Gamma_0$, denoted by $P(\Gamma < \Gamma_0)$ in the following, is not negligible. This means that a substantial number of configurations lead to a reduction of the LDOS compared with the free-space value. We checked that this reduction of the LDOS results from the interaction of the emitter with several scatterers (multiple scattering). Indeed, in the single-scattering regime, one has $P(\Gamma < \Gamma_0) \simeq 0$ [13,14]. A reduction of the decay rate of diamond color centers due to scattering in powders has been reported recently [19] and seems to support the results of these simulations.

In order to get more insight into the conditions leading to a reduction of the decay rate, we plot in Fig. 2 the probability $P(\Gamma < \Gamma_0)$ versus the size R of the cluster for the same values of $k_0\ell_B$ as in Fig. 1. Two points should be emphasized. First, if the thickness of the system is large enough, the probability of having a decay rate Γ smaller than the vacuum decay rate Γ_0 is substantial and can reach 50% for specific sets of parameters. Moreover, except for very small values of R/ℓ_B, this probability increases with $k_0\ell_B$. Secondly, we observe the presence of oscillations whose periodicity is, as we shall see, $\lambda_0/[2\mathrm{Re}(n_{\mathrm{eff}})]$ where n_{eff} is the effective refractive index describing the behavior of the averaged field. This suggests that the reduction of the decay rate results from a collective effect. Up to system sizes $R \sim 5\ell_B$, this collective effect is sensitive to the full size R of the finite-size system. For $R \gtrsim 5\ell_B$, the oscillations vanish and finite-size effects become negligible. This observation is also consistent with the fact that, in this range of parameters, small values of Γ cannot be obtained when the dipole source is interacting with only one scatterer [13].

IV. AVERAGED DECAY RATE AND EFFECTIVE MEDIUM APPROACH

The oscillating behavior in Fig. 2 is the combination of collective interactions and finite-size effects, which should

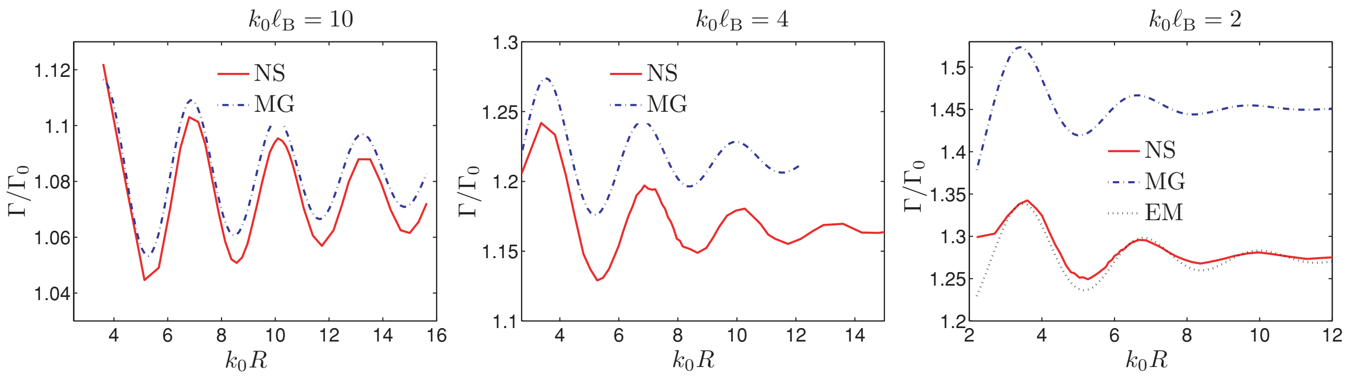

FIG. 3. (Color online) Averaged normalized spontaneous decay rates $\langle\Gamma\rangle/\Gamma_0$ versus the normalized system size k_0R, computed using the exact numerical simulation (NS), a Maxwell-Garnett mixing rule (MG), and a fit of the effective refractive index (EM). The parameters are the same as in Figs. 1 and 2.

also be observed in the averaged decay rate $\langle\Gamma\rangle$. In the absence of correlations in the scatterers positions, the averaged Green function can be described by an effective-medium theory [2], which relies on the computation of an effective dielectric function (or effective refractive index). For a system made of uncorrelated point dipoles, and in the absence of recurrent scattering, the Maxwell-Garnett (MG) mixing rule can give accurate results, even in the multiple-scattering regime [20]. In this approach, the effective dielectric function is given by

$$\epsilon_{MG}(\omega_0) = \frac{3 + 2\rho\alpha(\omega_0)}{3 - \rho\alpha(\omega_0)} \qquad (5)$$

and the effective refractive index is $n_{MG} = \sqrt{\epsilon_{MG}}$. Note that for a medium with identical scatterers, this expression is identical to that given by the Lorentz-Lorenz model [21]. From this value of the dielectric function and the expression of the Green function of a homogeneous sphere of radius R with a spherical cavity of radius R_0 at its center [22], one can compute the averaged normalized decay rate Γ_{MG}. This effective-medium result can be compared to the averaged decay rate $\langle\Gamma\rangle$ obtained from the numerical simulation. The results are plotted in Fig. 3 versus the system size R, for the same parameters as in Figs. 1 and 2. We observe the same oscillating behavior as in Fig. 2 for the probability $P(\Gamma < \Gamma_0)$. For $k_0\ell_B = 10$, the MG theory reproduces well the variations of $\langle\Gamma\rangle/\Gamma_0$. In particular, the period of the oscillations is half the effective wavelength $\lambda_{MG} = \lambda_0/\text{Re}(n_{MG})$, confirming an interference effect in the finite-size effective medium. The accuracy of the MG theory in this case results from the absence of correlations in positions between the scatterers (low effective volume fraction $f < 2\%$) and the absence of recurrent scattering [21]. For $k_0\ell_B = 4$ and $k_0\ell_B = 2$, the scattering strength becomes larger and recurrent scattering is no longer negligible. As expected, this severely limits the accuracy of the MG theory. Nevertheless, $\langle\Gamma\rangle$ can still be fitted using an effective refractive index $n_{eff} \neq n_{MG}$, as seen in Fig. 3 (right panel). The real part of n_{eff} is determined by the period of the oscillations $\lambda_0/[2\text{Re}(n_{eff})]$. The imaginary part is chosen to fit the averaged decay rate for large systems (for which finite-size effects vanish). For the parameters used in the simulation with $k_0\ell_B = 2$, the value is found to be $n_{eff} = 1.04 + 0.17i$. The real part is almost the same as in the MG theory whereas the imaginary part is rather different. The latter describes attenuation by scattering, with a scattering mean-free path $\ell = \lambda_0/[4\pi\,\text{Im}(n_{eff})]$ that is different from the Boltzmann mean free path ℓ_B. This difference is the signature of the existence of recurrent scattering in the disordered system; namely, repeated scattering by the same scatterer. Although the recurrent-scattering correction is expected to produce spatial dispersion (nonlocality) in the dielectric function, this remains negligible in the absence of spatial correlations in the positions of the scatterers and when the scattering process is dominated by far-field interactions [23]. This is observed in our calculations, where the averaged decay rate $\langle\Gamma\rangle$ can be described using an effective-medium model based on an isotropic and local refractive index, even in a strong-scattering regime ($k_0\ell_B = 2$). In these conditions, as shown in Ref. [9], one should be able to find a specific frequency range in which $\langle\Gamma\rangle$ is smaller than Γ_0. The frequency range depends on the dispersive behavior of the scattering medium. A numerical validation of this theoretical result is left for further studies.

Finally, let us discuss the possibility of measuring the LDOS in a disordered finite-size system made of resonant scatterers. The decay rate Γ is the inverse of the lifetime τ of the excited state of a dipole emitter, provided that this lifetime can be measured by usual techniques, assuming that the transit time of a photon from the emitter to the detector is negligible compared to τ. In the multiple-scattering regime, and for resonant point scatterers, this transit time is $\Delta t \sim R^2/D$, where $D = v_E\ell_B/3$ is the photon diffusion coefficient. The transport velocity is dominated by the dwell time (or Wigner time) due to the resonance of the scatterers, so that $v_E = \gamma\ell_B$ [6,24]. One ends up with $\Delta t \sim 3R^2/(\gamma\ell_B^2)$. The condition on the decay rate that enables a measurement through standard lifetime techniques is $\Gamma \ll \gamma/b_0^2$, where $b_0 = R/\ell_B$ is the optical thickness of the medium and γ is the linewidth of the resonant point scatterers.

V. CONCLUSION

In conclusion, we have studied the statistics of the spontaneous decay rate of a dipole emitter embedded in a strongly scattering medium on the basis of exact numerical simulations.

We have investigated the multiple-scattering regime in which the source interacts with more than one scatterer, with $1 \lesssim k_0\ell_B \simeq 10$. We have shown that a reduction of the spontaneous decay rate can be observed with a non-negligible probability, which corresponds to a reduction of the LDOS. This reduction of the LDOS is a consequence of collective interactions and interferences. In the absence of correlations in the positions of the scatterers, an effective-medium model can reproduce the behavior of the average decay rate. For $k_0\ell_B = 2$, the effective-medium model demonstrates the existence of recurrent scattering inside the medium. The approach described in this paper may be extended to study the behavior of the LDOS in random lasers based on resonant point scatterers [25,26] and in the presence of sub- or superradiant modes in atomic systems [27].

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