Near-field interactions and nonuniversality in speckle patterns produced by a point source in a disordered medium

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A point source in a disordered scattering medium generates a speckle pattern with nonuniversal features, giving rise to the so-called C_0 correlation. We analyze theoretically the relationship between the C_0 correlation and the statistical fluctuations of the local density of states, based on simple arguments of energy conservation. This derivation leads to a clear physical interpretation of the C_0 correlation. Using exact numerical simulations, we show that C_0 is essentially a correlation resulting from near-field interactions. These interactions are responsible for the nonuniversality of C_0 that confers to this correlation a huge potential for sensing and imaging at the subwavelength scale in complex media.

I. INTRODUCTION

Waves propagating in a disordered medium generate a strongly fluctuating spatial distribution of intensity known as a speckle pattern [1]. The spatial structure of a speckle pattern is often characterized by the intensity spatial correlation function $\langle I(\mathbf{r})I(\mathbf{r}')\rangle$, or its angular counterpart $\langle I(\mathbf{u})I(\mathbf{u}')\rangle$, where the unit vector $\mathbf{u}$ defines an observation direction. In usual experiments the medium is illuminated by an external beam, and the speckle pattern is observed, e.g., in transmission. Short-range and long-range contributions can be identified in the intensity correlation function, which is written as a sum of three terms denoted by C_1 (short range), C_2, and C_3 (long range) [2]. These correlations have been widely studied since they are responsible for enhanced mesoscopic fluctuations [3] and their sensitivity to changes in the medium can be used for imaging in complex media [4]. Moreover, the possibility of controlling speckle patterns by wavefront shaping has generated new interest [5]. When the waves are generated by a point source placed inside the medium, a new type of spatial correlation appears, which has been denoted by C_0 [6]. This correlation has features that make it particularly interesting: It is of infinite range and can dominate the usual long-range correlations, and it is strongly dependent on the local environment of the source [6,7]. This nonuniversality makes C_0 a valuable quantity for sensing or imaging in complex media with a high sensitivity to the microscopic structure of the medium [7]. Moreover, it has been shown that the C_0 correlation and the fluctuations of the local density of states (LDOS) at the location of the point source are equal [8]. This means that C_0 could be obtained from measurements of LDOS fluctuations instead of an analysis of speckle patterns. In optics, LDOS fluctuations can be measured from the fluorescence lifetime of nanoscopic emitters [9–12]. Note that the full statistical distribution of the LDOS can be obtained experimentally by measuring temporal fluctuations of the lifetime of one emitter in a slowly moving dynamic medium [9] or by measuring the lifetime of a large number of emitters spread over one configuration of a disordered sample [10–12].

This statistical distribution is expected to provide information on the local environment of the emitter [13,14] and on the photon transport regime [15–19]. In this article, we revisit the C_0 correlation concept and its connection to LDOS fluctuations in the framework of scattering of electromagnetic waves. We consider electromagnetic waves since the most recent relevant experiments have been performed in this context. First, we give a novel derivation of the equality between C_0 and the normalized variance of the LDOS based on energy conservation, showing that the relationship is valid in any regime, including strong localization [1]. This derivation leads for the first time to a clear physical interpretation of the C_0 correlation. Second, we analyze the nonuniversality of C_0 and show that it is due to a large extent to near-field interactions. The sensitivity to the degree of correlation of disorder around the location of the source is demonstrated based on numerical simulations on three-dimensional open systems.

II. ENERGY CONSERVATION, LDOS, AND C_0 CORRELATION

In this first part, we define the C_0 correlation and derive its relationship with LDOS fluctuations based on energy conservation. We consider a nonabsorbing disordered system of finite size, embedded in a sphere with radius R (this geometry is that used in the subsequent numerical simulations). The system is illuminated by a classical point dipole $\mathbf{p}$ radiating at frequency ω, placed at the center of the cluster at position $\mathbf{r}_s$. The medium is assumed statistically isotropic and homogeneous within the sphere of radius R. For a given configuration of disorder, the LDOS $\rho(\mathbf{r}_s,\omega)$ can be computed using the dyadic Green function that describes the response at point $\mathbf{r}$ to the dipole source through the relation $\mathbf{E}(\mathbf{r}) = \mu_0\,\omega^2\,\mathbf{G}(\mathbf{r},\mathbf{r}_s,\omega)\mathbf{p}$. It is given by $\rho(\mathbf{r}_s,\omega) = 2\omega/(\pi c^2)\,\mathrm{Im}[\mathrm{Tr}\,\mathbf{G}(\mathbf{r}_s,\mathbf{r}_s,\omega)]$, where Tr denotes the trace of a tensor and c is the speed of light in vacuum [20]. In terms of the LDOS, the power radiated outside the system, and averaged over the orientation of the dipole source, reads $P = \pi\omega^2/(12\epsilon_0)\,|\mathbf{p}|^2\,\rho(\mathbf{r}_s,\omega)$. Denoting by ρ_0 and P_0 the LDOS and the radiated power in vacuum, respectively, we obtain the simple equality

$$\frac{\rho(\mathbf{r}_s,\omega)}{\rho_0(\omega)} = \frac{P}{P_0}. \tag{1}$$

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As a characterization of the far-field speckle produced by the point source, we consider the angular intensity correlation function

$$C(\mathbf{u},\mathbf{u}') = \frac{\langle I(\mathbf{u})I(\mathbf{u}')\rangle}{\langle I(\mathbf{u})\rangle\langle I(\mathbf{u}')\rangle} - 1, \qquad (2)$$

where $I(\mathbf{u})$ is the directional radiated power in direction $\mathbf{u}$, such that $\int_{4\pi} I(\mathbf{u})d\mathbf{u} = P$. From Eqs. (1) and (2), the fluctuations of the normalized LDOS can be written

$$\left\langle\frac{\rho^2(\mathbf{r}_s,\omega)}{\rho_0^2(\omega)}\right\rangle = \frac{1}{P_0^2}\int\int\langle I(\mathbf{u})I(\mathbf{u}')\rangle\,d\mathbf{u}\,d\mathbf{u}'$$

$$= \frac{1}{P_0^2}\int\int\langle I(\mathbf{u})\rangle\langle I(\mathbf{u}')\rangle[1 + C(\mathbf{u},\mathbf{u}')]\,d\mathbf{u}\,d\mathbf{u}'. \qquad (3)$$

Using the hypothesis of statistical isotropy, the averaged directional radiated power reduces to $\langle I(\mathbf{u})\rangle = (P_0/4\pi)\langle\rho(\mathbf{r}_0,\omega)\rangle/\rho_0(\omega)$, and the angular intensity correlation becomes a function of $x = \mathbf{u}\cdot\mathbf{u}'$. This allows us to simplify Eq. (3) into

$$\frac{\langle\rho^2(\mathbf{r}_s,\omega)\rangle}{\langle\rho(\mathbf{r}_s,\omega)\rangle^2} = 1 + \frac{1}{16\pi^2}\int\int C(\mathbf{u}\cdot\mathbf{u}')\,d\mathbf{u}\,d\mathbf{u}'$$

$$= 1 + \frac{1}{2}\int_{-1}^{+1} C(x)\,dx. \qquad (4)$$

The correlation function can be expanded on the basis of Legendre polynomials in the form $C(x) = \sum_{n=0}^{\infty} a_n P_n(x)$. Since $P_0(x) = 1$, the first term is constant and corresponds to an infinite range correlation. To be consistent with initial considerations of the C_0 correlation [6,8], we define the C_0 contribution as that given by the constant term, such that $a_0 = C_0$. The integral in Eq. (4) is performed by writing $C(x) = \sum_{n=0}^{\infty} a_n P_0(x)P_n(x)$ and using the orthogonality condition of Legendre polynomials $\int_{-1}^{+1} P_n(x)P_m(x)dx = 2\delta_{nm}/(2n+1)$ where δ_{nm} is the Kronecker delta. We finally obtain

$$C_0 = \frac{\langle\rho^2(\mathbf{r}_s,\omega)\rangle}{\langle\rho(\mathbf{r}_s,\omega)\rangle^2} - 1 = \frac{\text{Var}[\rho(\mathbf{r}_s,\omega)]}{\langle\rho(\mathbf{r}_s,\omega)\rangle^2}. \qquad (5)$$

Equation (5) shows that the C_0 speckle correlation and the normalized variance of the LDOS at the position of the emitter are the same, a result that was first derived in Ref. [8] based on a diagrammatic approach. Our derivation relies only on considerations of energy conservation and the assumption of a nonabsorbing and statistically isotropic medium. In particular, Eq. (5) holds in all wave transport regimes, from weakly scattering to strongly scattering, including Anderson localization. Another feature of our derivation is that it leads to a simple interpretation of the C_0 correlation. For a classical point dipole source, changes in the LDOS correspond to changes in the power transferred to the environment (for a quantum emitter, this corresponds to a change in the spontaneous decay rate). In a nonabsorbing medium, this power coincides with the total power radiated in the far field, whose angular (or spatial) variations generate the speckle pattern. Therefore LDOS fluctuations are transferred into global fluctuations of the speckle patterns, which themselves are encoded into angular (or spatial) correlations.

III. NEAR-FIELD INTERACTIONS AND INFLUENCE OF DISORDER CORRELATIONS

We have carried out numerical simulations in order to analyze the dependance of C_0 on the local environment of the emitter. The scattering medium is a three-dimensional cluster of N resonant point scatterers randomly distributed inside a sphere with radius R. A dipole emitter is placed at the center of the cluster (position $\mathbf{r}_s$) and is surrounded by a small exclusion volume with radius R_0. The geometry is shown in the inset in Fig. 1. In the generation of the random configurations of the disorder (i.e., of the positions of the N scatterers), a minimum distance d_0 is forced between the scatterers. This permits to induce a degree of correlation in the disorder, since this amounts to simulating an effective hard-sphere potential between scatterers. One can define a fictitious volume fraction $f = N(d_0/2)^3/(R^3 - R_0^3)$ that can be taken as a measure of the degree of correlation of the disorder (f will be denoted by "correlation parameter" in the following). For large f, the hard-sphere potential is long range (d_0 is large), so there is a weak probability of getting two closely separated scatterers. Small values of f correspond to nonoverlapping point scatterers (delta-correlated disorder). The scatterers are described by their electric polarizability $\alpha(\omega) = -3\pi c^3\gamma/[\omega^3(\omega - \omega_0 + i\gamma/2)]$, where ω_0 is the resonance frequency and γ is the linewidth. This corresponds to the polarizability of a resonant nonabsorbing point scatterer (which is similar to that of a two-level atom far from saturation). The scattering cross section is $\sigma_s(\omega) = (k^4/6\pi)|\alpha(\omega)|^2$, with $k = \omega/c$. The parameter $k\ell_B$ measures the scattering strength, where $\ell_B = [\rho\sigma_s(\omega)]^{-1}$ is the independent-scattering (or Boltzmann) mean-free path, $\rho = N/V$ being the density of scatterers. In all the numerical computations that follow, we have taken the following set
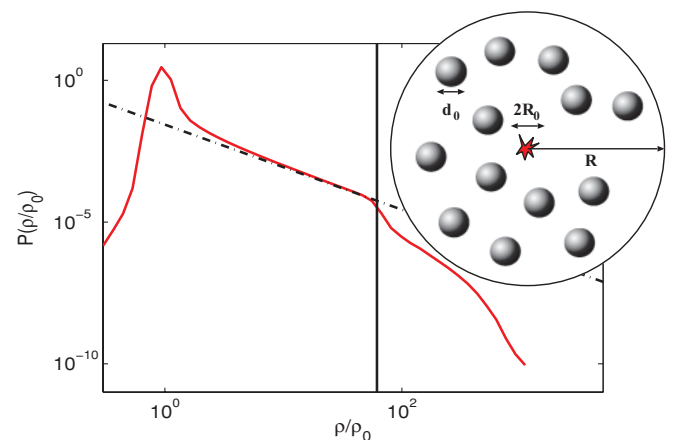

FIG. 1. (Color online) Statistical distribution of the normalized LDOS $\rho(\mathbf{r}_s,\omega)/\rho_0(\omega)$ for a uncorrelated system with correlation parameter (effective volume fraction) $f = 0.001\%$ in a double logarithmic plot. Others parameters are $N = 100$, $\omega_0 = 3 \times 10^{15}$ Hz, $\omega - \omega_0 = 1 \times 10^9$ Hz (wavelength $\lambda = 0.63\,\mu$m), $\gamma = 1 \times 10^9$ Hz, $R = 1.2\,\mu$m, and $R_0 = 0.05\,\mu$m. The calculations are performed with 3×10^8 configurations. This large number of configurations is necessary to correctly describe the tail of the distribution. The dashed line indicates a power-law behavior $P(\rho/\rho_0) \propto (\rho/\rho_0)^{-3/2}$. The vertical solid line indicates the one-particle cutoff. (Inset) Schematic view of the system.

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The calculation of the statistical distribution of the LDOS, from which C_0 can be deduced, amounts to calculating the Green function $\mathbf{G}(\mathbf{r},\mathbf{r}_s,\omega) = \mathbf{G}_0(\mathbf{r},\mathbf{r}_s,\omega) + \mathbf{S}(\mathbf{r},\mathbf{r}_s,\omega)$ for an ensemble of realizations of the scattering medium. Since the free-space Green function $\mathbf{G}_0$ is known analytically, we only need to calculate the Green function $\mathbf{S}(\mathbf{r},\mathbf{r}_s,\omega_0)$ which corresponds to the scattered field. To proceed, we perform a coupled-dipole numerical computation. The field exciting scatterer number j is given by the contribution of the dipole source and of all other scatterers, leading to a set of $3N$ self-consistent equations [21]:

$$\mathbf{E}_j = \mu_0 \omega^2 \mathbf{G}_0(\mathbf{r}_j,\mathbf{r}_s)\mathbf{p} + \alpha(\omega)k^2 \sum_{\substack{k=1 \\ k \neq j}}^{N} \mathbf{G}_0(\mathbf{r}_j,\mathbf{r}_k)\mathbf{E}_k, \qquad (6)$$

where $\mathbf{r}_j$ is the position of scatterer number j and the dependence of the Green functions on ω have been omitted. This linear system is solved numerically for each configuration of the disordered medium. Once the exciting electric field on each scatterer is known, it is possible to compute the scattered field at the source position $\mathbf{r}_s$ and deduce the Green dyadic, from which the LDOS ρ is readily obtained. In this numerical approach, near-field and far-field dipole-dipole interactions and multiple scattering are taken into account exactly. We stress that for the computation of near-field interactions, the vectorial nature of the field has to be taken into account, as is well known in near-field optics [22]. In particular, dipole-dipole interactions through near (quasistatic) fields cannot be described with a scalar model.

We show in Fig. 1 the statistical distribution of the normalized LDOS $\rho(\mathbf{r}_s,\omega)/\rho_0(\omega)$ in the case of a correlation parameter $f = 0.001\%$ (minimum interparticle distance $d_0 = 7.5$ nm) which corresponds to an almost uncorrelated medium. The curve exhibits a broad distribution, with values of ρ/ρ_0 ranging from 0.2 to 1000. This corresponds to much larger fluctuations than those observed in the single scattering regime, where ρ/ρ_0 slightly deviates from unity [13]. The analysis of the line shape allows us to distinguish three regimes. First, the curve covers a zone corresponding to $\rho/\rho_0 < 1$, which means that some configurations lead to a reduction of the LDOS compared to that in free space. This effect has been analyzed previously and is due to collective interactions in the multiple-scattering regime [19]. Recent measurements of the fluorescence lifetime of emitters at the surface of a volume scattering disordered medium seem to provide evidence of this regime [10]. Second, in the region $\rho/\rho_0 > 1$, a power-law decay is observed, with a statistical distribution behaving as $P(\rho) \propto \rho^{-3/2}$ (the power law is indicated by the dashed line in Fig. 1). As originally discussed in Ref. [14], this behavior is due to near-field interactions between the emitter and the nearest scatterer, in the regime of dipole-dipole interactions, and in the absence of absorption. Evidence of this regime, and of its
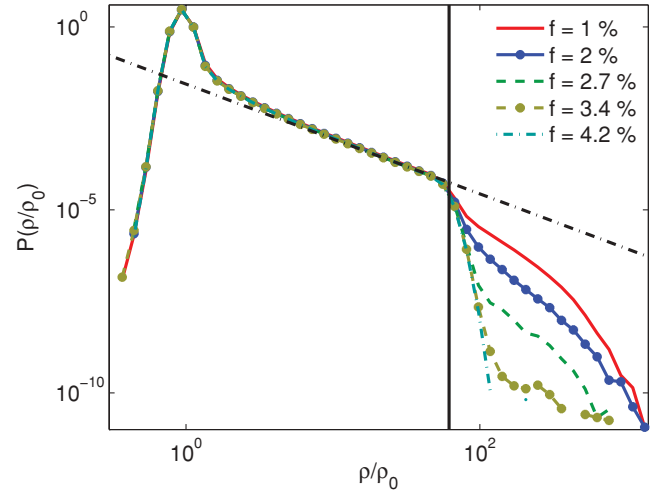

FIG. 2. (Color online) Same as Fig. 1, with different values of the correlation parameter f. As a result of near-field interactions, the tail of the distribution is the signature of the local environment of the emitter.

influence on the C_0 correlation, have been reported recently [11]. Third, in the region $\rho/\rho_0 \gg 1$, the tail of the distribution deviates from the power law $\rho^{-3/2}$. The onset of the deviation depends on the exclusion volume with radius R_0 that encloses the emitter. Indeed, in the single-scattering limit (the emitter interacts chiefly with the nearest scatterer), one would observe a sharp cutoff indicated by the vertical solid line in Fig. 1. This corresponds to the value of ρ/ρ_0 given by the near-field dipole-dipole interaction between the emitter and one scatterer located at a distance R_0 ($\rho/\rho_0 = 62$ in the present case) [14]. The observation of a tail beyond this single-scattering cutoff is the evidence of near-field interactions with more than one scatterer. As we shall see below, this tail also contains the information on the local environment of the emitter, and in particular on the degree of correlation of disorder.

We show in Fig. 2 the statistical distribution of ρ/ρ_0 for different values of the correlation parameter, ranging from 1% to 4.2% (i.e., d_0 ranging from 111 to 180 nm). Increasing f amounts to increasing the level of correlation in the positions of the scatterers (one imposes an effective hard-sphere potential with a longer range). The tail of the distribution is substantially affected by the level of correlations in the system. Also note that the part of the distribution corresponding to ρ/ρ_0 smaller than the single-scatterer cutoff remains unchanged. This means that the sensitivity of C_0 to the local environment of the emitter is driven by the near-field interactions with the surrounding scatterers, this information being encoded in the tail of the statistical distribution of the LDOS. Although this tail corresponds to events with a low probability, it is at the core of the C_0 correlation concept.

In order to visualize the influence of the correlation of disorder directly on C_0, we have plotted in Fig. 3 the values of C_0 versus the correlation parameter f obtained from numerical simulations (for the same system as in Figs. 1 and 2). A sharp transition is visible at $f \simeq 2\%$, the value of C_0 dropping by a factor of 2. In order to give a physical interpretation of this behavior, we have also plotted in Fig. 3 the values of
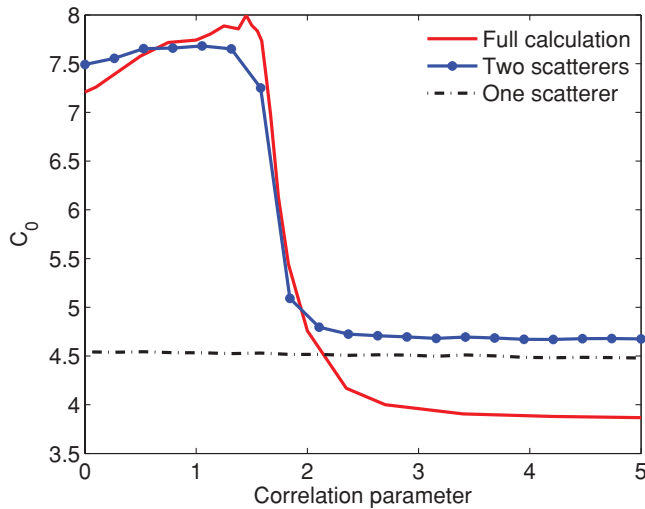
FIG. 3. (Color online) C_0 speckle correlation versus the correlation parameter f. Red solid line: Full numerical simulation with the same parameters as in Fig. 1. Blue solid line with markers: Calculation considering only the two nearest scatterers (double scattering). Black dashed line: Calculation considering only the nearest scatterer (single scattering).

C_0 computed by considering the interaction with the nearest scatterer only (black dashed curve), and with the two nearest scatterers (blue line with markers). For large f, the emitter essentially interacts with one particle (the red solid line and the black dashed curve have a similar behavior) and the C_0 correlation can be understood in simple terms. This is the regime found experimentally in Ref. [11]. We stress here that this regime results from a near-field interaction, so the value

of C_0 depends on local microscopic parameters (it cannot be described with the scattering or transport mean free path as a single parameter). For small f, the probability of getting more than one scatterer in the vicinity of the emitter becomes non-negligible, and the behavior of C_0 cannot be explained (even qualitatively) with a single-scattering model. One sees that by including the interaction with the two nearest scatterers (blue curve with markers), one reproduces nicely the behavior of the transition. This result demonstrates the high sensitivity of C_0 to the level of correlation of the disorder and the key role of near-field interactions [23]. These interactions are responsible for the nonuniversality of C_0.

IV. CONCLUSION

In summary, we have derived the relation between the C_0 speckle correlation and the LDOS fluctuations using arguments of energy conservation. This simple and exact derivation leads for the first time to an interpretation of C_0 based on the fluctuations of the energy delivered by a classical dipole source to a disordered environment. Using exact numerical simulations, we have shown that C_0 is essentially a correlation resulting from near-field interactions. These interactions give C_0 its nonuniversal character, which is reflected in its high sensitivity to the level of correlation of disorder. This nonuniversality confers to C_0 a potential for imaging at the submicron scale in complex media.

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