Spatial Correlations in the Near Field of Random Media

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(Received 19 November 2002; published 26 August 2003)

The conventional coherence theory suggests that the fields radiated by statistically homogeneous sources correlate over spatial regions of the order of the wavelength irrespective of the distance from the surface of the source. Contrary to these predictions, we show that the spatial correlations of optical fields in close proximity of highly scattering, randomly inhomogeneous media depend on this distance and, moreover, their extent can be significantly smaller than the wavelength. The contribution of evanescent fields is experimentally demonstrated and the coherence length in the near field is shown to relate to the coherence properties at the surface which are, in turn, determined by the structural characteristics of the random media.

DOI: 10.1103/PhysRevLett.91.093901 PACS numbers: 42.25.Kb, 42.25.Dd

When optical radiation propagates through a randomly inhomogeneous medium, the multiple scattering events randomize the direction and the phase of the wave. The radiation originating from such a system is commonly regarded as spatially incoherent. It was a long-time belief that multiple scattering washes out all the structural information and that such an inhomogeneous medium would behave as a perfect lambertian diffuser.

The classical example of an incoherent source is a thermal source. The optical coherence theory predicts that the quasihomogeneous lambertian sources of radiation are not completely spatially incoherent; at a given frequency ω, the field correlates over regions whose spatial dimensions are of the order of the wavelength $\lambda = 2\pi c/\omega$ [1]. This fundamental result has been obtained by neglecting the contribution of short-range evanescent waves and it has been successful in describing the far-field properties of thermal emission [2]. However, recent advances in the experimental capabilities and an increasing interest in nanoscale phenomena has also raised new questions regarding the emission of optical radiation and, in general, the coherence properties in the near field [3–6]. Very recently, it has been suggested that the spatial coherence length of the field close to the surface of a thermal source may be either much larger or much smaller than the wavelength of the light depending on the dominant contribution to the cross-spectral density tensor [3,7]. To date, no experimental evidence exists for spatial near-field coherence properties.

So far, near-field scanning techniques have been used to investigate different aspects of electromagnetic fields in close proximity of interfaces aiming primarily at extending the spatial resolution of various optical microscopies. In this Letter we report direct measurements of coherence effects in the near field of highly inhomogeneous media which are known to produce significant field fluctuations and a radiant intensity typical to incoherent sources.

The interaction between optical waves and random media has been systematically investigated and it is now well understood that the familiar appearance of speckle patterns is due to the short-range correlations of the waves transmitted or reflected from the random medium. The field correlations $C(\mathbf{R}) = \langle E(\mathbf{r})^* E(\mathbf{r} + \mathbf{R})\rangle$ in the bulk of a random medium have been under scrutiny for quite some time [8]. Using diagrammatic calculations and assuming a constant photon density, Shapiro found that the optical field at points separated by a distance R in a highly random volume correlates like [8]

$$C(R) = [\sin(kR)/(kR)]\exp(-R/2l), \tag{1}$$

where l is the scattering mean free path and $k = 2\pi/\lambda$. In the weakly scattering regime, $kl > 1$, the speckle size in the bulk of a multiple scattering medium is of the order of λ. Based on photon diffusion arguments, Freund finds that the field correlation at the surface of a random medium is given by [9]

$$C(R) = 2[\Delta \sin(kR)/(kR) + J_1(kR)/(kR)]/(1 + 2\Delta), \tag{2}$$

with Δ being a factor of the order of unity. Again, the wavelength of light determines the size of the field correlations and it is worth mentioning that a similar conclusion is reached when the coherence properties of two-dimensional statistically homogeneous sources are evaluated [1]. It would appear therefore that the extension of the spatial correlations is limited to the value of the wavelength. Of course, this treatment does not account for the existence of significant evanescent contributions to the field distribution at the surface and, therefore, fails to explain the coherence effects in the near field of a random medium.

An alternative description can be developed which considers the surface of the random medium as being equivalent to a homogeneous, planar, statistically stationary source of optical radiation [1]. This source, located at $z = 0$, is characterized by a cross-spectral density function [10]

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$$W^{(0)}(\boldsymbol{\rho}_1, \boldsymbol{\rho}_2, \omega) = F^{(0)}(\boldsymbol{\rho}_2 - \boldsymbol{\rho}_1, \omega), \quad (3)$$

where $\boldsymbol{\rho}_1$ and $\boldsymbol{\rho}_2$ are two-dimensional position vectors in the plane of the source and ω is the radiation frequency.

The source generates, in the half space $z > 0$, a field with the cross-spectral density function given by [10]

$$W(\mathbf{r}_1, \mathbf{r}_2, \omega) = k^2 \iint_{-\infty}^{\infty} \tilde{F}(ks_x, ks_y, \omega) e^{ik(\mathbf{s}\mathbf{r}_1 - \mathbf{s}^*\mathbf{r}_2)} ds_x ds_y, \quad (4)$$

where $\tilde{F}(ks_x, ks_y, \omega)$ is the two-dimensional Fourier transform of cross-spectral density at $z = 0$, $F^{(0)}(\boldsymbol{\rho}_1 - \boldsymbol{\rho}_2, \omega)$.

In general, one can decompose the cross-spectral density function in low- and high-frequency components:

$$W(\delta x, \delta y, z, \omega) = W_{\mathrm{hom}}(\delta x, \delta y, \omega) + W_{ev}(\delta x, \delta y, \omega), \quad (5)$$

where the homogeneous component of the cross-spectral density function is

$$W_{\mathrm{hom}}(\delta x, \delta y, \omega) = k^2 \iint_{s_x^2 + s_y^2 \leqslant 1} \tilde{F}(ks_x, ks_y, \omega) e^{ik(s_x \delta x + s_y \delta y)} ds_x ds_y, \quad (6)$$

and the inhomogeneous one is evaluated like

$$W_{ev}(\delta x, \delta y, z, \omega) = k^2 \iint_{s_x^2 + s_y^2 > 1} \tilde{F}(ks_x, ks_y, \omega) e^{[ik(s_x \delta x + s_y \delta y)]} e^{(-2kz\sqrt{s_x^2 + s_y^2 - 1})} ds_x ds_y, \quad (7)$$

where $r_2 - r_1 = (\delta x, \delta y, 0)$ and $s = (s_x, s_y, \sqrt{1 - s_x^2 - s_y^2})$. It is worth mentioning that the previous treatment of the spatial coherence properties of planar sources has been conducted by neglecting the high-frequency components of the cross-spectral density function which are associated with the evanescent field [10].

As can be seen, the coherence function $W$is practically determined by the spectral degree of coherence in the source plane at $z = 0$:

$$\mu^{(0)}(\boldsymbol{\rho}_2 - \boldsymbol{\rho}_1, \omega) = \frac{F^{(0)}(\boldsymbol{\rho}_2 - \boldsymbol{\rho}_1, \omega)}{S^{(0)}(\omega)}, \quad (8)$$

where $S^{(0)}(\omega) = F^{(0)}(0, \omega)$ is the spectrum of the light in the plane of the source. We consider a Gaussian correlated source with its spectral degree of coherence of the form

$$\mu^{(0)}(\boldsymbol{\rho}_2 - \boldsymbol{\rho}_1, \omega) = \exp[-(\boldsymbol{\rho}_2 - \boldsymbol{\rho}_1)^2/2\sigma^2], \quad (9)$$

where σ is the field correlation length in the plane of the source. Straightforward algebra leads to

$$W_{\mathrm{hom}}(r, \omega) = \pi k^2 \sigma^2 S^{(0)}(\omega) \int_0^1 \rho e^{-(k^2\sigma^2\rho^2/2)} J_0(k\rho r) d\rho \quad (10)$$

and

$$W_{ev}(r, z, \omega) = \pi k^2 \sigma^2 S^{(0)}(\omega) \int_1^{\infty} \rho e^{-(k^2\sigma^2\rho^2/2)} J_0(k\rho r) e^{-2kz\sqrt{\rho^2 - 1}} d\rho, \quad (11)$$

where $\delta x = r\cos\theta$, $\delta y = r\sin\theta$, and $s_x = \rho\cos\phi$, $s_y = \rho\sin\phi$.

Using the results in Eqs. (5), (10), and (11) we calculate the cross-spectral density as a function of the distance z for different values of the field correlation length σ at $z = 0$. In Fig. 1, the values of the field coherence length σ_μ defined as the $1/e$ values of the cross-spectral density are plotted as a function of distance z from the surface. The calculations are for $\lambda = 488$ nm and account for both homogeneous and inhomogeneous contributions. One can see that the field coherence length increases with distance z and that larger values of σ_μ are obtained when the field correlation length σ in the plane of the source is increased.

We measured the spatial coherence properties near the surface of various random, highly scattering media which were illuminated in a transmission geometry. Here we report data collected on compact slabs made of calcium carbonate and kaolin microparticles with an average diameter of 300 nm and an average refractive index 1.46. Their scattering properties are characterized by the correlation length of the refractive index fluctuation and by the rms height of the surface which is typically in the range from $\lambda/4$ to $\lambda/2$. When optical radiation propagates through a highly random medium, the multiple scattering events randomize completely the direction of the wave. As a result, the propagation vectors of the waves reaching the surface together with the topological characteristics of the surface determine both propagating and evanescent waves. A collection mode near-field scanning optical microscope (Nanonics NSOM-100) was used to obtain simultaneously near- or far-field optical images as well as the corresponding atomic force microscopy (AFM) topography [11]. We refer here as "near field" to images collected at distances z smaller than the wavelength, and as "far field" for those distances z of 5–20 wavelengths from the interface. An

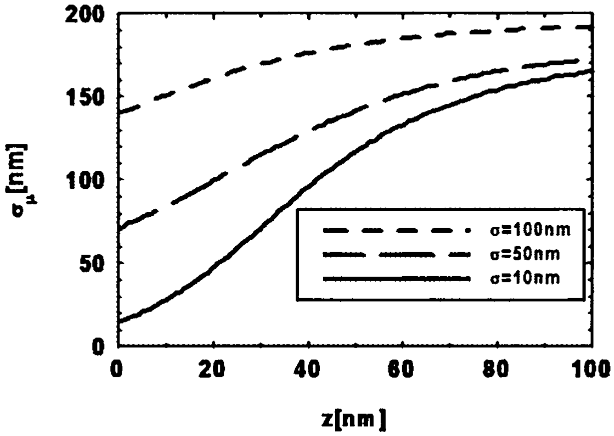

FIG. 1. Field coherence length σ_μ as a function of distance z from the surface and calculated for different values of the source correlation length σ as indicated. The calculations account for both homogeneous and inhomogeneous contributions.

Ar + ion laser ($\lambda = 488$ nm) is used to illuminate the random medium and the transmitted field is coupled to the cantilevered near-field optical force sensor which is scanned along the sample. The scattered light is detected using a photomultiplier tube (PMT) operating in the photon counting mode. The Al-coated fused silica fiber tip with an aperture of 150 nm provides the possibility to scan in constant distance (for the near-field images) or in constant height mode (for the far-field images).

In the close vicinity of the surface of a highly inhomogeneous medium, the NSOM probe collects both the homogeneous and inhomogeneous components of the transmitted field. When the intensity is averaged over an area of 5×5 μm, a negative exponential dependence on z is observed, proving that the detected signal contains a significant contribution of inhomogeneous components. As expected, this contribution vanishes for distances z larger than the wavelength of light, where only the homogeneous components are contributing. From the intensity collected at constant z, we have also obtained the first-order intensity statistics and noticed that the probability density function of intensity manifests a longer tail in the near-field than in the far-field zone. This can be interpreted as an intensity enhancement due to the evanescent contributions at distances z smaller than the wavelength.

The spatial distribution of the detected intensity resembles the typical spatial variations present in a classical speckle pattern. At a given observation point in the speckle pattern, the field is a superposition of contributions originating from different locations within the bulk of the random medium and we can consider that the amplitude and the phase of the elementary phasors satisfy circular Gaussian statistics. This assumption is supported by the first-order statistics of intensities determined in both near and far field.

The second-order intensity-intensity correlation can also be evaluated from the intensity distributions measured at a given z. Using standard properties of Gaussian random variables, the field correlation function can then be expressed in terms of intensity correlation function. We evaluated numerically the field correlation functions and estimated the average radius of the cross section area where the field correlation decreases at $1/e$ of its maximum. We consider the width of the field correlation as a measure of the field coherence length σ_μ.

Typical results of far-field measurements are presented in Fig. 2. When estimating the field coherence lengths, the data were corrected with the point spread function of the probe. The error bars denote the average over different scanning measurements at the same location across the sample. As can be seen the spatial coherence properties do not depend on the distance z from the surface. This is to be expected from Eqs. (5) and (10) when the contribution from homogeneous components of the field is considered and when the extension of the source is practically infinite. Note that classical measurements of spatial coherence properties are performed at much larger distances z where the limited size of the source cannot be ignored. In that case, the size of the coherence area is actually determined by the extent of the source as described by the Van Cittert–Zernike theorem.

As opposed to the far-field situation, the field coherence length σ_μ measured in the near field of a highly inhomogeneous medium has a significant dependence on the distance z. In addition, we found that σ_μ is always smaller than the far-field saturation value. This is predicted by our model of optical field radiated by a planar source generating both propagating and evanescent waves. We would like to emphasize that a nontrivial z dependence is also predicted for both first and second-order statistics of the emitted field. Therefore, a direct

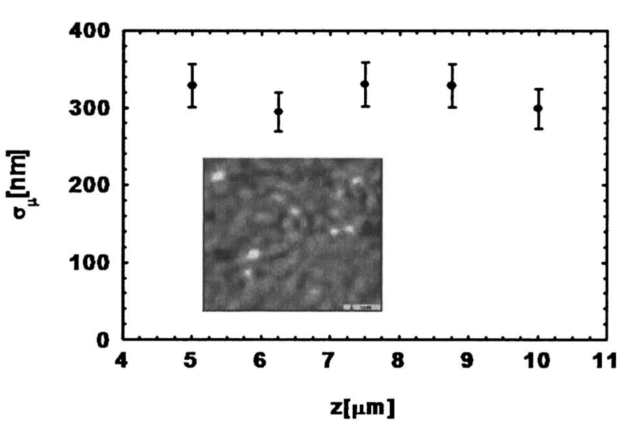

FIG. 2. Measured far-field coherence length as a function of the distance z from the surface. The inset shows a typical far-field intensity distribution.

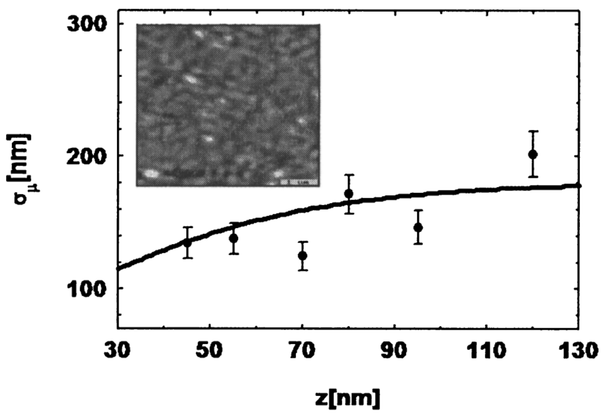

FIG. 3. Measured near-field coherence length as a function of the distance z from the surface. The inset shows a typical near-field intensity distribution.

comparison with experimental data is presented in Fig. 3 where the near-field values of σ_μ are plotted as a function of distance z. A fair agreement is obtained with the model described before. The calculation based on Eqs. (5), (10), and (11) is shown with a continuous line in Fig. 3. We used a value of $\sigma = 50$ nm for the field correlation length in the plane of the source. As can be seen, this simple model of a planar source qualitatively describes both first- and second-order statistical properties of the field emitted at the surface of a highly random medium. It also represents a direct relationship between the measurable statistical properties of the optical near field and the statistical properties of an equivalent planar source of radiation which, in turn, are determined by the physical properties of the random medium. Now, if it would be to relate the equivalent source model to physical properties of the random medium, one should bear in mind that the model assumes a planar source and, therefore, the same spectral degree of coherence in the source plane at $z = 0$ for both homogeneous and inhomogeneous contributions. However, the real scattering media have rough interfaces which may induce different surface coherence effects for low- and high-frequency components of the cross-spectral density function. A more refined model would require a full three-dimensional treatment of the field in the neighborhood of an interface that has transverse spatial frequencies [12].

In conclusion, we found that the spatial coherence properties of the field measured in the far field, i.e., several wavelengths away from the surface, do not change with the distance from the interface. At even larger distances, the coherence properties are dominated by the finite size of the scattering media. In the near field, however, we found that the spatial coherence properties have a nontrivial dependence on the distance from the physical interface and, therefore, on the near field average intensity. This is explained by the fact that coherence properties of random media at subwavelength scale are determined by both propagating and evanescent waves. Contrary to the predictions of conventional coherence theory, we have shown that the near-field coherence length is z dependent and that it can have values smaller than the wavelength of radiation.

Our results demonstrate that subtle coherence properties of light scattered by random media can be examined using optical near-field approaches. Based on the coherence properties at subwavelength scales, new possibilities could be suggested for surface and subsurface diagnostics of randomly inhomogeneous media.

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