Journal of Microscopy, Vol. 209, Pt 3 March 2003, pp. 173–176
Received 10 August 2002; accepted 25 October 2002

Near-field measurement of short-range correlation in optical waves transmitted through random media

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Key words. Near-field, random media, multiple scattering, light diffusion, correlation function.

Summary

Two-dimensional near-field images of speckle patterns formed by optical waves transmitted through a disordered porous silica glass sample are measured. The corresponding 2D intensity correlation function, *C*, is extracted. The subwavelength spatial resolution of near-field microscopy allows us to resolve in the spatial distribution of *C* the expected subwavelength oscillations and to follow their dependence on the excitation wavelength. Finally, we deduce the effective refractive index of the material by fitting the theoretical spatial dependence of *C* to our experimental results.

Introduction

Light wave propagation in disordered dielectric structures is a very promising and interesting research subject. It shows many similarities with the propagation of electrons in disordered metals and various phenomena related to electron transport have now been identified in the propagation of light waves in disordered media. Important examples are the photonic Hall effect (van Tiggeln 1995), universal conductance fluctuations (Scheffold & Marat, 1998) and Anderson localization (John, 1984). Important applications of light diffusion include medical imaging (Yodh & Chance, 1995) and diffusing wave spectroscopy (Pine *et al.*, 1988).

In optically thick disordered dielectric structures light is diffusely scattered via complicated multiple scattering processes (Ping Sheng, 1995). Interference effects can survive these random events and the spatial and angular distributions of the intensity pattern contain a large amount of information about the particular realization which produced them. The surviving interference gives rise, for example, to the memory effect (Freund *et al.*, 1988), to coherent backscattering (van Albada & Langebdjik, 1985; Wolf & Maret, 1985), and to short- and long-range correlations in the intensity distribution (Shapiro, 1986; Stephen & Cwilich, 1987; Feng *et al.*, 1988; Genack *et al.*, 1990). It is now well known that the diffusely transmitted intensity pattern, also referred to as the bulk speckle pattern, shows a highly irregular light distribution governed by statistical properties. These are well described by the correlation function $C = \langle \delta I \delta I' \rangle$, where $\delta I = I - \langle I \rangle$ is the fluctuation of the intensity with respect to its average.

In the weak scattering limit ($kl >> 1$ where $k = 2\pi/\lambda$ and l is the free mean path), the main contribution to C is the short-range correlation term which is given by the square of the field correlation function: $C_1 = |\langle EE'^* \rangle|$. The spatial dependence of C_1 for a monochromatic source has been calculated by Shapiro (1986) and is given by $(\sin k\Delta r/k\Delta r)^2 \exp(-\Delta r/l)$. This expression shows that the correlation in a speckle pattern decreases exponentially with the mean free path, modulated by an oscillating feature on the scale of the wavelength. By defining a correlation length δr as the first zero of the fine structure, the short-range correlation term shows that in the weak scattering regime there will always exist a lower limit to the correlation length, given by the probe wavelength.

While the far-field properties, such as the memory effect or coherent backscattering, have been demonstrated for optical waves (van Albada & Lanendijk, 1985; Wolf & Maret, 1985; Freund *et al.*, 1988), the spatial oscillations in the short-range correlation, which are an intrinsic property of the near-field intensity distribution, have so far been observed only for microwave radiation (Sebbah *et al.*, 2002).

Here, we report on the two-dimensional (2D) short-range correlation of the intensity distribution of the transmitted light waves that have been repetitively scattered through a

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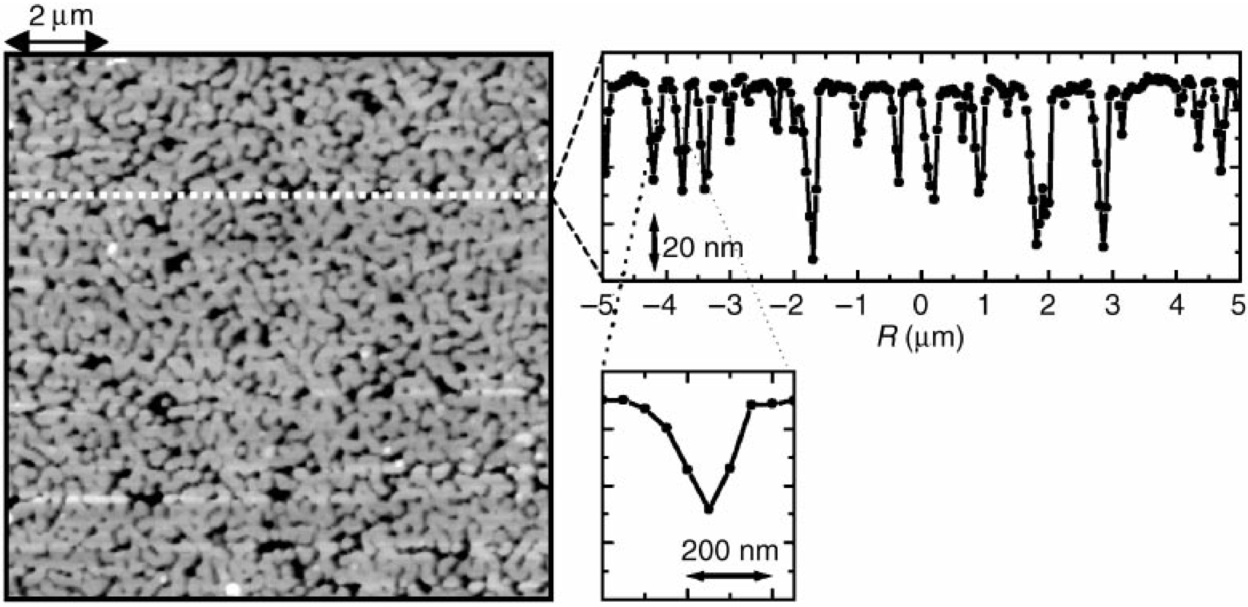

Fig. 1. Two-dimensional shear force image of the sample surface; cross-section through the image indicates an average pore size of ≈ 100 nm.

disordered medium. By using a scanning near-field optical microscope, we collect images of the intensity patterns for different excitation wavelengths. The autocorrelation of these images unmistakably shows a 2D subwavelength modulation as predicted by theory. By fitting the theoretical spatial dependence of C_1 to our data we were able to deduce the effective refractive index of disordered samples.

The sample studied is a disordered dielectric structure of macroporous silica glass with randomly orientated and interconnected pores 200 nm in size. The thickness of the sample is ≈ 1.7 mm, the surface area is ≈ 4×6 mm^2.

Spatially resolved near-field images of the speckle patterns are collected using a commercial (Twinsnom, OMICRON) scanning near-field optical microscope in the illumination mode. Light from a He–Ne (632 nm) and a diode laser (780 nm) is transmitted through a pulled coated near-field probe and the spot of the diffusely transmitted light through the sample is imaged onto the entrance of a GaAs photomultiplier. The solid angle of collection is 0.36 sr.

Near-field optical maps are recorded by scanning the sample with respect to the near-field probe and by acquiring, for each position, the corresponding integrated intensity of the diffusely transmitted spot. The wavelength dependence is obtained by coupling a different excitation laser into the fibre and taking care that exactly the same sample region is scanned. The mean free path is derived from the diffusion constant as measured in a time-resolved transmission experiment, following the configuration of Wiersma *et al.* (2001). A short (1 ps) pulse is incident on the front sample interface and the diffused light in transmission is monitored using a streak camera.

Figure 1 shows a 10×10 µm^2 shear force image of the sample surface. A cross-section through the image (inset) reveals an average pore size of 100–200 nm and a volume fraction of roughly 70%. Using the geometrical average, we can estimate an average refraction index $n_{av} = 1.3$. This value is expected to be ≈ 10–15% higher than the actual effective refractive index (den Outer & Lagendijk, 1993).

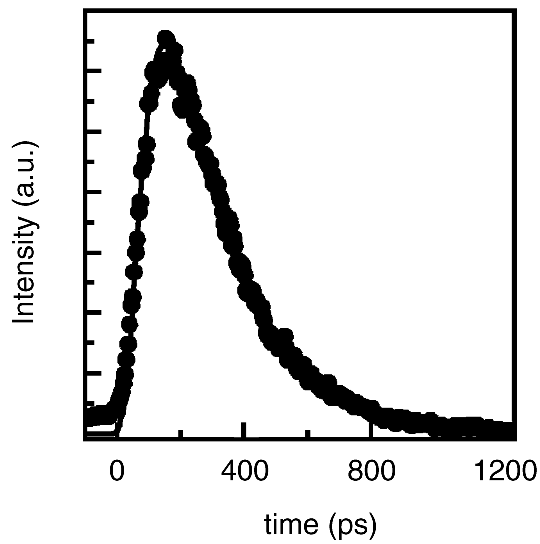

Fig. 2. Time-resolved transmission through the sample. The solid line is a fit from diffusion theory (Wiersma *et al.*, 2001). The resulting diffusion constant is $D = 9.6 \times 10^2 \pm 5\%$ m^2 s^{-1}.

Figure 2 shows the time-resolved transmission measured in the slab geometry where a short laser pulse is incident on the front sample interface, and the time evolution of the transmitted

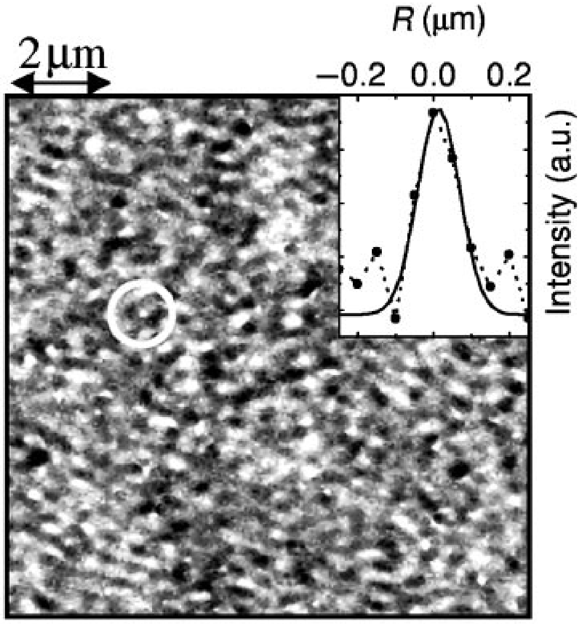

Fig. 3. Two-dimensional near-field optical image of the integrated intensity of the transmitted spot as a function of the tip position. Inset: cross-section through the white circle in the image. Incident wavelength = 780 nm.

diffused light is recorded. The diffusion constant is found by comparing the transmitted light curve (Fig. 2: dots) with the solution of a diffusion equation for a slab geometry (Fig. 2: solid line) (Wiersma *et al.*, 2001). From the fit to the data we find a diffusion constant of $D = 9.6 \times 10^2 \pm 5\%\ \mathrm{m^2\ s^{-1}}$.

The value of l can be obtained via $l = 3D/v_e$ with $v_e = c_0/n$ and n the average refractive index of the sample (see below). In this way we find for l a value of 11 μm ± 5%.

Figure 3 shows the 2D map of the transmitted intensity $I(\vec{R})$, as a function of the tip position. The wavelength is 780 nm. Dark and bright spots are resolved with a spatial resolution of ≈ 150 nm (inset cross-section through the white circle in the image). Similar images have been recorded for excitation wavelength at 632 nm. From the 2D near-field map of the intensity pattern, we extract the 2D intensity correlation function, $C(\Delta\vec{R}) = \langle \delta I(\vec{R})\delta I(\vec{R} - \Delta\vec{R})\rangle$. Figure 4 shows the map of $C(\Delta\vec{R})$ for 632 and 780 nm incident wavelengths, respectively. The images show a peak in the centre, which corresponds to $\Delta\vec{R} = 0$. Radially from the centre, a series of maxima and minima indicate the subwavelength oscillatory components of the correlation function. As noted in the figure, their spatial distribution scales with the excitation wavelength.

It is also evident that the correlation function does not have a full radial symmetry as expected from the theory. The reason for this may be related to an asymmetry in the porous orientation or in the tip aperture. Measurements are in progress in order to address this point.

The normalized intensity correlation function is defined as

$$C(\Delta r, \Delta R) = \langle \delta I(\vec{r}, \vec{R})\, \delta I'(\vec{r}\,', \vec{R}')\rangle / \langle I(\vec{r}, \vec{R})\rangle \langle I(\vec{r}\,', \vec{R}')\rangle, \qquad (1)$$

where δI is the deviation of the intensity from its ensemble average value ($\delta I = \langle I - I\rangle$), $\Delta r = |\vec{r} - \vec{r}\,'|$ and $\Delta R = |\vec{R} - \vec{R}'|$ are the radial displacements across the output and input surfaces, respectively, and the brackets ⟨ … ⟩ represent the ensemble average.

The normalized intensity correlation function can be written as the sum of three terms: C_1, C_2 and C_3, describing short-, long- and infinitive-range correlations, respectively. The dominating contribution to C in our system is given by the short-range correlation term C_1.

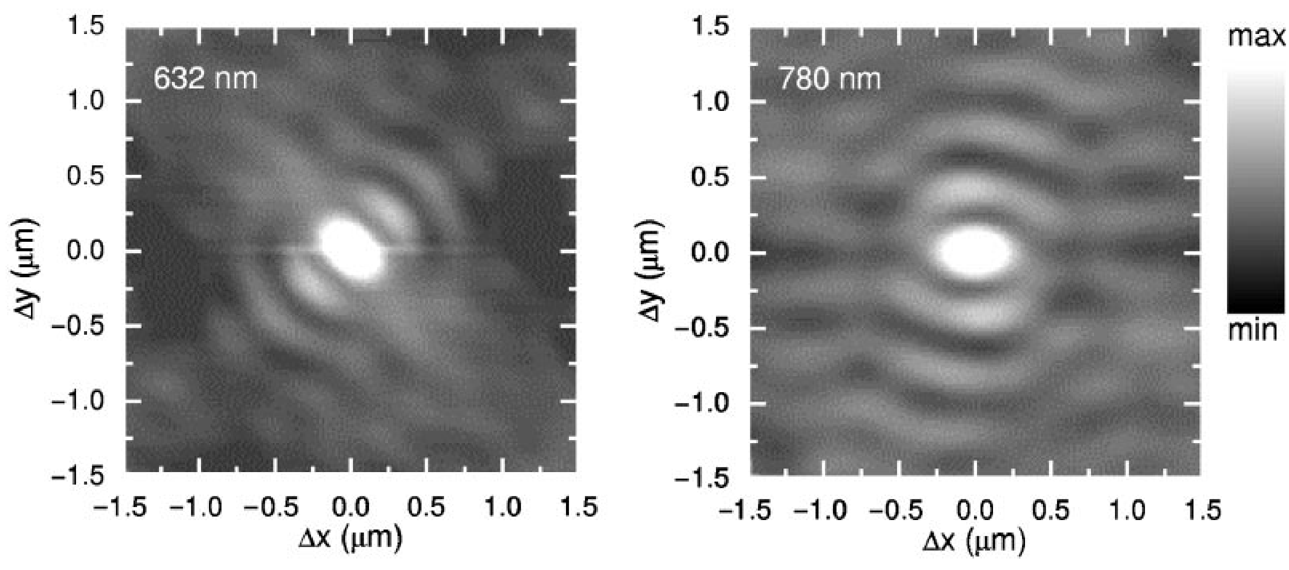

Fig. 4. Two-dimensional spatial dependence of the intensity correlation function $C(\Delta R)$ for 632 and 780 nm excitation wavelengths.

It has recently been shown (Sebbah *et al.*, 2002) that $C(\Delta r,0)$ and $C_1(0,\Delta R)$ are equivalent as required by reciprocity. In our experimental configuration, the sample is moved with respect to the source and the transmitted light is collected by a fixed detector with a solid angle of collection of 0.36 sr. The total number of independent speckle spots is given by $N = 2\pi W/\lambda^2$ (Feng *et al.*, 1988; van Albada *et al.*, 1990), with W the illuminated area on the output surface of the sample. For our geometry this yields $N \approx 3.6 \times 10^7$ at 632 nm wavelength and $N \approx 2.3 \times 10^7$ at 780 nm wavelength. At our solid angle of collection we collect a fraction of 0.057 of these independent speckle spots, which is $N_c \approx 2.0 \times 10^6$ at 632 nm and $N_c \approx 1.3 \times 10^6$ at 780 nm. Hence our correlation function is normalized by a factor $A = 1/\sqrt{N_c}$, with N_c as given above. This means that we actually measure the function $AC_1(0,\Delta R) = A \times F(\Delta R)$, with $A \approx 7.0 \times 10^{-4}$ at 632 nm and $A \approx 8.6 \times 10^{-4}$ at 780 nm wavelength.

For the case of a point source in an elastic scattering medium the function $F(\Delta R)$ has been calculated by factorizing the fields (Shapiro, 1986)

$$F(\Delta R) = |\langle E(\vec{r},\vec{R})E^*(\vec{r}',\vec{R}')\rangle|^2/\langle I(\vec{r},\vec{R})\rangle\langle I(\vec{r}',\vec{R}')\rangle = \left(\frac{sin(k\Delta R)}{k\Delta R}\right)^2 exp(-\Delta R/\ell). \quad (2)$$

Equation 2 predicts that the short-range correlation term is dominated by a rapidly varying structure determined by the wavelength of the incident light and the refractive index of the material.

For the sample under investigation, the measured l is ≈ 10 µm so that the exponential term can be neglected as the spatial region where the oscillations are experimentally observed (2 µm) is much shorter than l. The radial section through the images of Fig. 3 are fitted by the expression: $AC_1(\Delta R) = A \times \left(\frac{sin(k\Delta R)}{k\Delta R}\right)^2$, where A is expected to be of the order of $1/\sqrt{N_c}$, and the scaling factor A and the effective refractive index n are the only fitting parameters. We found good agreement between theory and experimental data by taking for n a value of 1.1 ± 0.1, which is somewhat lower than the geometrical average value of 1.3 as expected (den Outer & Lagendijk, 1993).

In conclusion, near-field optical images of the speckle patterns formed by optical waves transmitted through an optically thick random medium have been measured. From the 2D images a map of the intensity correlation function has been extracted. The predicted existence of short-range correlation in the transmitted pattern and its dependence on the incident wavelength was studied experimentally. Comparing the theory and our data, the effective refractive index of the medium is extracted. The good agreement between experimental results and theoretical prediction provides the first experimental evidence for the spatial dependence of C_1 for optical waves in the diffusive transport regime.

Acknowledgements

We thank M. Gurioli, Patrick Sebbah, and Azriel Genack for helpful discussions. This work was supported by the European Community under contract number HPRI-CT1999-00111, and by the INFM project PHOTONIC.

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