Direct determination of diffusion properties of random media from speckle contrast

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We present a simple scheme to determine the diffusion properties of a thin slab of strongly scattering material by measuring the speckle contrast resulting from the transmission of a femtosecond pulse with controlled bandwidth. In contrast with previous methods, our scheme does not require time measurements nor interferometry. It is well adapted to the characterization of samples for pulse shaping, nonlinear excitation through scattering media, and biological imaging.

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The propagation of optical waves in scattering media is both a fundamental and a very applied topic that requires a deep understanding of how waves propagate in the medium [1]. Upon multiple scattering, that is, when the dimension L of the system is much larger than the transport mean free path ℓ_t, the transport is diffusive, with a diffusion coefficient D.

Coherence plays an essential role in propagation. In the cw domain, characterized by a single pulsation ω_0, multiple scattering gives rise to a very universal random interference figure: the speckle [2]. For a polarized monochromatic wave, the spatial distribution of the speckle intensity I follows a negative exponential distribution, with a contrast of unity.

In a slab geometry, measuring scattering properties, in particular ℓ_t, D, and the energy velocity v_E, is by no means trivial. The angular width of the coherent backscattering cone [3] or, in the case of thin slab geometry, total transmission studies [4] can give an evaluation of ℓ_t. The spread of a short pulse [5,6], speckle correlations [4,7], interferometric measurements [8], or variation of the effective refractive index [9] can be used to extract the diffusion constant D. Simultaneous measurement of D and ℓ_t [10,11] give access to v_E.

The relevant time, in the dynamical case, is the Thouless time τ_d [12], with associated bandwidth $\Delta\nu_d \sim 1/\tau_d$. The characteristic time of diffusion $\tau_d \sim L^2/(\pi^2 D)$ is the time it takes for a photon to leave the medium, which is related to the diffuse traversal time $\tau_l \sim L^2/(6D)$ [8,13], which also takes into account the direction of the propagation of light ($\tau_l \geq \tau_d$). In essence, a short laser pulse of duration $\tau_p < \tau_l$ transmitted through the medium will be scattered throughout and its duration will be extended by approximately τ_l by multiple scattering.

Earlier works have studied how the speckle depends on the coherence or frequency of the source [14–16]. The temporal behavior of the speckle has been measured with a streak camera in the subnanosecond range [17] or via linear or nonlinear interferometry [18–20]. In this Letter, we introduce a simple method to determine the diffusion properties of a thin slab of strongly scattering material, in a regime where the other techniques are not easily applicable. The method is based on a simple contrast measurement on a CCD camera and illumination with a femtosecond laser of variable spectral bandwidth.

The investigated samples are solid three-dimensional scattering media with slab geometry made by sedimentation through vertical deposition, and subsequent drying, of a 5 wt. % water suspension of ZnO powder [21]. This technique allows the growth of homogeneous samples of the order of a few cm^2 with very flat surfaces, and filling fractions of around 50%. The particle size distribution is $230 \pm 70\,\mathrm{nm}$.

In the simple model of [2], the contrast of the speckle image is shown to be $C = 1/\sqrt{N}$, where N is the number of independent speckle patterns added incoherently. In the present case, the medium is illuminated by a pulse with a bandwidth $\Delta\nu_p$, larger than the bandwidth of the medium $\Delta\nu_m (\simeq 1/\tau_l)$. After propagation through the medium, each independent speckle pattern has a bandwidth limited by $\Delta\nu_m$. Thus $N \simeq \Delta\nu_p/\Delta\nu_m$, making the contrast $C \sim 1/(\tau_l \times \Delta\nu_p)^{1/2}$. This implies that a measurement of the contrast as a function of $\Delta\nu_p$ will give access to τ_l and, therefore, D.

The experimental setup is shown in Fig. 1. The light source is a mode-locked Ti:sapphire laser (Spectra Physics Mai Tai 90 fs pulse and 80 MHz repetition rate), with a central wavelength of 748 nm. The 2 mm collimated TEM_{00} output of the laser is sent in a zero dispersion line [22], used to select the spectral bandwidth of the light before the scattering medium. It consists of a series of three optical elements, a grating (1200 pitch per millimeter), a variable slit attached to a mirror, and a lens between the mirror and the grating at the focal distance $f = 50\,\mathrm{cm}$ from both elements. After the zero dispersion line, the beam is focused at the surface of the scattering medium with a $f = 50\,\mathrm{cm}$ lens. Transmitted light is collected with a microscope objective (Olympus 100×,

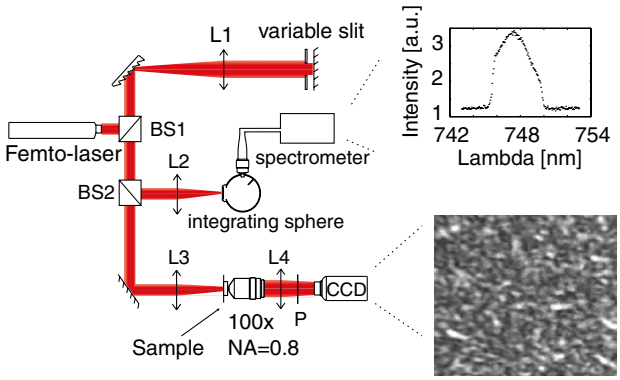

Fig. 1. (Color online) Experimental setup with variable slit to control the spectral bandwidth, typical measured spectrum, and speckle image on the CCD camera.

NA = 0.8) and the back focal plane of the objective is imaged with a $f = 20\,\mathrm{cm}$ lens on a CCD camera (AVT PIKE F-100B). The object plane of the microscope is moved by $\sim 10\,\mu\mathrm{m}$ from the sample surface in order to increase the size of the speckle grain on the CCD Camera. A linear polarizer is placed between the objective and the lens to ensure that a single polarization component is imaged. To monitor the input spectrum, the laser is partially deflected onto a spectrometer (Ocean Optics HR4000, resolution $\leq 0.1\,\mathrm{nm}$) by a beam splitter after the zero dispersion line. Spectrum and image are acquired simultaneously. The pulse bandwidth at the entrance of the medium is varied between the bandwidth of the laser $\Delta\lambda_{\max} = 8\,\mathrm{nm}$ and $\Delta\lambda_{\min} = 0.3\,\mathrm{nm}$, limited by diffraction on the slit.

A typical image on the CCD is shown in the inset of Fig. 1. The contrast of the speckle is calculated within a homogeneously illuminated region of the CCD. The region is statistically large and calculated either in the speckle image (speckle), or outside (dark). The contrast $C = \sigma(\text{speckle})/(\langle\text{speckle}\rangle - \langle\text{dark}\rangle)$, where $\langle\,\rangle$ and $\sigma(\,)$ are the mean and standard deviation of the pixel intensities within the defined region. The error of C is evaluated based on the standard deviation of the dark image.

Samples with different thicknesses were illuminated with light pulses of various bandwidths $\Delta\nu_p$ in order to investigate the link between the speckle contrast and D. We quantify the contrast modification with $\Delta\nu_p$ in a more complete formalism, by treating a thin strongly scattering sample using a slab geometry and assuming a nonabsorbing medium. This treatment, however, is general and can be extended to any geometry and medium, including those in which both scattering and absorption are significant. The first question is to determine the distribution of transit time through such a medium. A general treatment of this problem has been introduced in [23]. In the multiple scattering regime, the spatially integrated transmittance $T_L(t)$ as a function of time for negligible absorption reads (from Eq. 15 of [23])

$$T_L(t) = (4\pi D)^{-1/2} \times t^{-3/2} \times \left\{ (L - \ell_t) e^{-\frac{(L-\ell_t)^2}{4Dt}} - (L + \ell_t) e^{-\frac{(L+\ell_t)^2}{4Dt}} + (3L - \ell_t) e^{-\frac{(3L-\ell_t)^2}{4Dt}} - (3L + \ell_t) e^{-\frac{(3L+\ell_t)^2}{4Dt}} \right\}. \quad (1)$$

We neglect contributions from more than three reflections as they do not contribute significantly to T_L in the numerical calculation. This time-dependent transmission has to be compared to the pulse duration in order to determine the contrast. In [14], a formalism is introduced for this purpose, in the context of a cw laser propagating through a very thick medium, which is also valid in the case of a short pulse. A spatially coherent source of spectrum $S(\lambda)$ going through a scattering medium of time distribution $\tilde{T}_L(t) = T(t)/\int_0^\infty T(t)\mathrm{d}t$ produces a speckle with a contrast C given by the following integral:

$$C = \int_0^\infty \frac{1}{S(\lambda)} \left\{ \int_0^\infty \int_0^\infty S(\lambda) S(\lambda') \times |f(\lambda, \lambda', \tilde{T}_L(t))|^2 \mathrm{d}\lambda \mathrm{d}\lambda' \right\}^{1/2} \mathrm{d}\lambda, \quad (2)$$

and where f is, with a change of variable, a Fourier transform of $\tilde{T}_L(t)$, and reads

$$f = \int_0^\infty c\tilde{T}_L(t) \times \exp\left[-i2\pi ct\left(\frac{1}{\lambda} - \frac{1}{\lambda'}\right)\right] \mathrm{d}t. \quad (3)$$

It is important to note that all parameters of the problem are not fully independent as $T_L(t)$ and C depend in a nontrivial way on all the parameters L, ℓ_t, and D. As a consequence, it is not possible to infer all parameters independently from contrast measurements.

Experimentally, by changing the width of the variable slit, measurements of the contrast were made over a range of spectral bandwidths $\Delta\nu_p$ (Fig. 2). As expected for large values of the bandwidth, the contrast asymptotically decreases with increasing bandwidth in the form $C \sim 1/\sqrt{\Delta\nu_p}$ and decreases with $\Delta\nu_p$ faster for thicker material as the traveling time gets longer. The monochromatic limit where $C = 1$ is reached when $\Delta\nu_p < \Delta\nu_d$, i.e., when τ_p is identical to the confinement time of the medium [24].

Once $C(\Delta\nu_p)$ is known, the diffusion constant D can be obtained if the transport mean free path ℓ_t is measured. ℓ_t can be extracted from a total transmission measurement by employing the optical analogue of Ohm's law: the angular integrated total transmission T is inversely proportional to the thickness L of the sample [11].

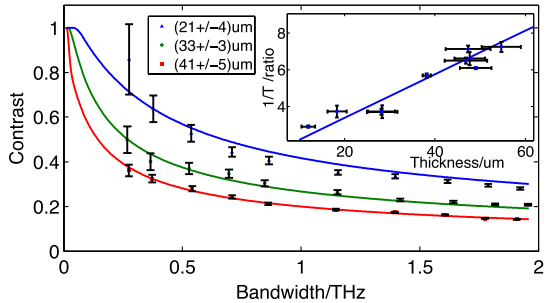

Fig. 2. (Color online) Contrast of the speckle for three different thicknesses of ZnO. Dots are the experimental measurements; solid curves are from calculation following the model developed by [23]. Inset: inverse of the transmission for ZnO layers of different thicknesses L in micrometers.

Total transmission as a function of sample thickness is measured by focusing a laser beam at 748 nm on a set of samples placed at the entrance of an integrating sphere. The integrated light transmitted at all angles is sent via an optical fiber to a spectrometer, which measures the total transmission. The inset in Fig. 2 shows a plot of the inverse of the transmission $T(L)$ as a function of the thickness for $\lambda = 748\,\mathrm{nm}$. Vertical error bars are based on the variation of transmission for different positions on the sample, while the horizontal ones are the standard deviation of the sample thickness upon repeated measurements. Fitting this data to Ohm's law gives $\ell_t = 2.1 \pm 0.2\,\mu\mathrm{m}$ and absorption length $\ell_a \geq 100\,\mu\mathrm{m}$ such that absorption is negligible.

$C(\Delta\nu_p)$, L, and ℓ_t were measured independently and, by fitting Eq. (2) to all data, D is found to be $(29 \pm 4)\,\mathrm{m^2 s^{-1}}$ for our ZnO samples. Fits are shown in Fig. 2 for three samples. This allows v_E to be calculated for our medium as $(0.47 \pm 0.04) \times 10^8\,\mathrm{ms^{-1}}$, i.e., $v_E \simeq 0.16c$, where c is the speed of light in vacuum.

Alternatively, the description of the speckle contrast in terms of interference of N independent spectral modes provides a straightforward estimate of the diffusion properties of the medium: using $C \sim 1/\sqrt{\tau_l \times \Delta\nu_p}$, we estimate the diffusion coefficient to be $(19 \pm 2)\,\mathrm{m^2 s^{-1}}$, close to the rigorous value previously calculated. Therefore, even if within this description, the geometry of the sample enters only through τ_l, this simple approach well describes the diffusion of a light pulse in a thin scattering slab.

In conclusion, using a CCD camera and a femtosecond light pulse, we demonstrate a simple setup to characterize the diffusion properties of a scattering medium. In order to avoid complex interferometry measurement of time and phase, we exploit the ability of a multiple scattering medium to mix spatial and spectral modes at the output [25,26]. In view of the recently realized spatiotemporal control of light pulses in scattering media [25–27], our concept applies for a wide range of Thouless time and, therefore, allows the characterization of the diffusion coefficient D for various scattering media, for photonics and biological applications.

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References

1. P. Sebbah, *Waves and Imaging through Complex Media* (Kluwer Academic, 2001).
2. J. W. Goodman, J. Opt. Soc. Am. **66**, 1145 (1976).
3. P. Wolf, G. Maret, E. Akkermans, and R. Maynard, J. Phys. **49**, 63 (1988).
4. A. Z. Genack, Phys. Rev. Lett. **58**, 2043 (1987).
5. D. S. Wiersma, A. Muzzi, M. Colocci, and R. Righini, Phys. Rev. E **62**, 6681 (2000).
6. J. M. Drake and A. Z. Genack, Phys. Rev. Lett. **63**, 259 (1989).
7. W. Cai, B. Das, F. Liu, M. Zevallos, M. Lax, and R. Alfano, Proc. Natl. Acad. Sci. USA **93**, 13561 (1996).
8. I. M. Vellekoop, P. Lodahl, and A. Lagendijk, Phys. Rev. E **71**, 056604 (2005).
9. S. Faez, P. M. Johnson, and A. Lagendijk, Phys. Rev. Lett. **103**, 053903 (2009).
10. M. P. van Albada, B. A. van Tiggelen, A. Lagendijk, and A. Tip, Phys. Rev. Lett. **66**, 3132 (1991).
11. R. Sapienza, P. D. García, J. Bertolotti, M. D. Martín, A. Blanco, L. Viña, C. López, and D. S. Wiersma, Phys. Rev. Lett. **99**, 233902 (2007).
12. D. J. Thouless, Phys. Rev. Lett. **39**, 1167 (1977).
13. R. Landauer and M. Buttiker, Phys. Rev. B **36**, 6255 (1987).
14. C. A. Thompson, K. J. Webb, and A. M. Weiner, Appl. Opt. **36**, 3726 (1997).
15. J. D. McKinney, M. A. Webster, K. J. Webb, and A. M. Weiner, Opt. Lett. **25**, 4 (2000).
16. J.-M. Tualle, H. L. Nghiêm, C. Schäfauer, P. Berthaud, E. Tinet, D. Ettori, and S. Avrillier, Opt. Lett. **30**, 50 (2005).
17. M. Tomita and T. Matsumoto, J. Opt. Soc. Am. B **12**, 170 (1995).
18. W.-L. Liu, R.-X. Gao, and S.-L. Qu, Chin. Phys. B **19**, 024204 (2010).
19. P. M. Johnson, A. Imhof, B. P. J. Bret, J. G. Rivas, and A. Lagendijk, Phys. Rev. E **68**, 016604 (2003).
20. E. Tal and Y. Silberberg, Opt. Lett. **31**, 3529 (2006).
21. P. García, R. Sapienza, Á. Blanco, and C. López, Adv. Mater. **19**, 2597 (2007).
22. A. Monmayrant, S. Weber, and B. Chatel, J. Phys. B **43**, 103001 (2010).
23. M. S. Patterson, B. Chance, and B. C. Wilson, Appl. Opt. **28**, 2331 (1989).
24. N. Cherroret and A. Buchleitner, Phys. Rev. A **83**, 033827 (2011).
25. J. Aulbach, B. Gjonaj, P. M. Johnson, A. P. Mosk, and A. Lagendijk, Phys. Rev. Lett. **106**, 103901 (2011).
26. O. Katz, Y. Bromberg, E. Small, and Y. Silberberg, Nat. Photon. **5**, 372 (2011).
27. D. J. McCabe, A. Tajalli, D. R. Austin, P. Bondareff, I. A. Walmsley, S. Gigan, and B. Chatel, "Shaping speckles: spatio-temporal focussing of an ultrafast pulse through a multiply scattering medium," arXiv:1101.0976v1 (2011).