SCATTERING OF LIGHT PULSES

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Optical heterodyning study of the propagation dynamics of IR femtosecond laser pulses in a strongly scattering porous medium

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***Abstract*. A system is devised for optical heterodyning based on a femtosecond Cr : forsterite laser using a balance scheme for the laser noise compensation. The dependence of a heterodyne signal on the time delay is measured by detecting backscattered laser radiation from a strongly scattering porous object (a sheet of paper). It is found that the backscattered signal contains 'a long tail' with an exponential decay caused by multiple scattering. The exponent of the exponential is determined by the lifetime of photons in a scattering layer. The absorption and scattering coefficients for different types of paper are measured by the photon lifetime.**

Keywords: *optical heterodyning, femtosecond laser, porous media, multiple scattering.*

Porous media are very interesting physical objects, which have unique properties caused by their internal structure. Interest in the study of such objects is related not only to practical applications, for example, the investigation of phase substitution in porous media [1] but also to fundamental problems concerning the physical properties of low-dimensional media. Recently, the propagation of ultrashort light pulses in statistically inhomogeneous media has aroused considerable interest in connection with the diagnostics of biological tissues [2] and the determination of localisation of photons in disordered systems [3 – 5].

One of the spectacular examples of a disordered system is paper. Being a strongly scattering medium, paper can be studied, in particular, for practical applications by well-developed methods of light scattering. These methods include optical coherent tomography based on optical heterodyning [2], speckle-correlation tomography [6], and time-resolved reflectometry using an image converter [7].

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The basic feature of the propagation of laser radiation in a strongly scattering medium is the fast decorrelation of photons and the appearance of an intense diffusion component due to multiple scattering. On the one hand, this restricts the possibilities of methods based on the detection of ballistic photons, but on the other hand, the diffusion component can serve as an additional source of information, which is used in diffusion spectroscopy and tomography [6].

The efficient method for studying the structural features of strongly scattering media is optical coherent tomography (OCT), which is based on the detection of backscattered photons using the heterodyne scheme to record the useful signal. Most of the papers on OCT are devoted to imaging biological tissues, and they, as a rule, ignore the question about the possible influence of multiply scattered photons on the imaging process [2]. However, the selection of photons in the OCT method is performed not by the scattering multiplicity but by the optical path length, which can result in a considerable distortion of images of strongly scattering objects due to the influence of multiple scattering of photons.

The aim of this paper was to demonstrate the application of optical coherent heterodyning to study the propagation dynamics of ultrashort light pulses in a strongly scattering porous disordered medium (in our case, paper). Preliminary results were reported in Ref. [8].

The optical heterodyne measuring instrument is based on a Michelson interferometer. We used in our experiments a 100-mW femtosecond Cr : forsterite laser emitting 50-fs pulses [9]. The signal-to-noise ratio was increased by using the noise compensation method with two photodetectors [10, 11]. We employed such an optical heterodyning scheme earlier to study laser-induced thermocapillary convection [12].

We measured the power of a heterodyne signal by detecting the intensity of a femtosecond laser pulse backscattered from paper as a function of the optical path z in the reference arm. This dependence can be conventionally divided into three parts (Fig. 1): a part with the intense leading edge produced due to scattering from the medium boundary (the part length is approximately equal to the duration of a probe femtosecond pulse), a short, rapidly decaying part corresponding to the passage from single to multiple scattering, and a long 'tail' with an exponential decay caused by multiple scattering of light in the medium.

The dependence in the second part was approximated by the exponential, the exponent being 11.8 ± 1.1 ps^{-1} for any thickness of a sample. We paid a special attention to the behaviour of the last part of the dynamic scattering curve.

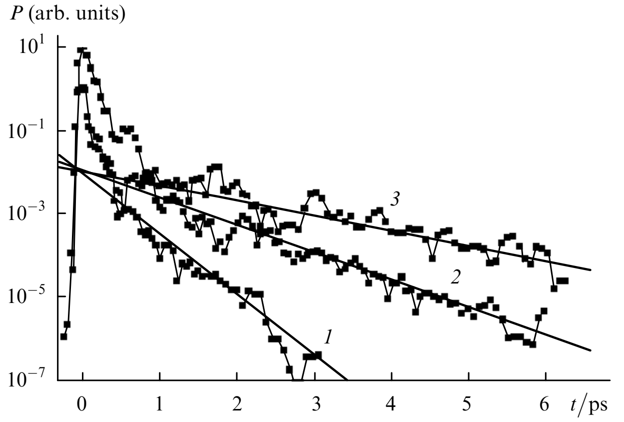

Figure 1. Dependences of the heterodyne signal power on the delay time for scattering in one (*1*), two (*2*), and three (3) paper layers. The thickness of a paper layer is ~ 80 μm.

Figure 1 shows the dependences of the heterodyne signal power on the time delay for one type of paper (high-porous paper with the average density of 0.4 g cm^{-3}, which is close in structure to filter paper), but for different thicknesses of a scattering layer. The sample thickness was varied by changing the number of paper layers, the air gap between the layers being minimised by slightly compressing the sample. One can see that, as the number of layers is increased, the tail of the pulse response becomes longer, which is manifested in the decrease in the slope of the dependence plotted at the logarithmic scale.

Figure 2a shows the dependences of the heterodyne signal power on the delay time t for a denser paper (the average density was 0.837 g cm^{-3}) with defects. The dependences were measured in the region of papers without defects and in the region of defects caused by a higher density. One can see that the signal lifetime decreases when the porosity is greater, i.e., when the density of scattering centres is lower. The behaviour of the heterodyne signal caused by the interference of scattered and reference radiation is determined by multiple scattering, which causes a noticeable delay of photons in the paper. The dependences presented in Fig. 2 are not smooth due to the presence of a stationary speckle pattern. To reduce the influence of speckles, it is necessary to perform spatial averaging. The results of spatial averaging obtained by rotating a paper disc are shown in Fig. 2b.

The slope of the exponential decay of the heterodyne signal is determined by the photon lifetime inside a given volume. In the diffusion approximation, taking absorption into account, the exponential decay is described by the expression [4]

$$I_{\rm m}(t) \sim \exp\left(-\frac{t}{\tau_0}\right) = \exp\left[-\frac{1}{\tau_\alpha} - \left(\frac{\pi}{L}\right)^2 D\right] t, \quad (1)$$

where τ_0 is the characteristic photon lifetime in a scattering medium; $D = c/3n_{\rm p}\mu_{\rm s}'$ is the diffusion coefficient of photons; $t = z/c$ is the delay time; c is the speed of light; $\mu_{\rm s}' = (1-g)\mu_{\rm s}$ is the reduced scattering coefficient; $n_{\rm p}$ is the average refractive index of paper; g is the anisotropy parameter; $\tau_\alpha = n/(c\alpha)$; and α is the absorption coefficient of the medium.

The average refractive index $n_{\rm p}$ of paper can be roughly represented in terms of the average phase shift after the

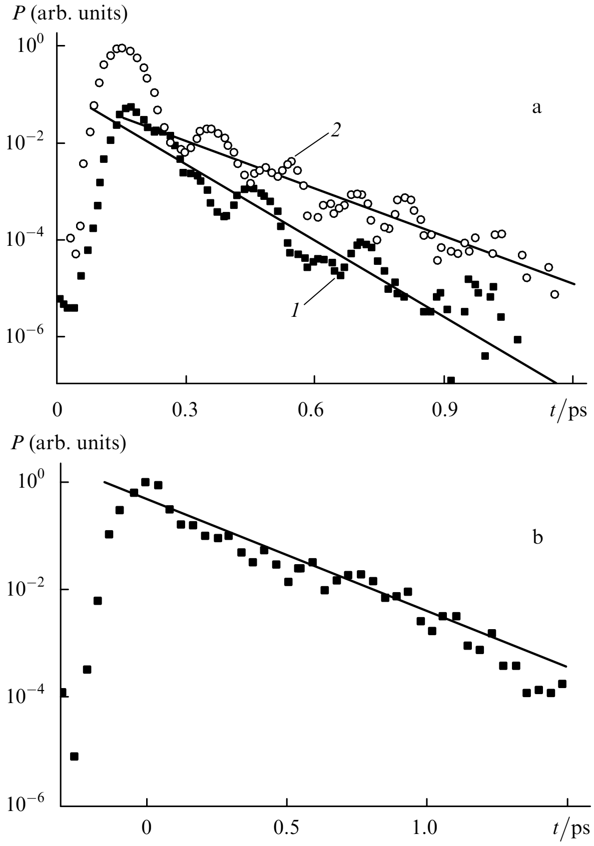

Figure 2. Dependences of the heterodyne signal power on the delay time obtained by probing paper in the defect region (with a greater porosity) (*1*) and in the region without defects (*2*) (a), and also by probing paper fixed on a rotating disc (b).

propagation of a photon through a layered structure consisting of cellulose (with the refractive index $n_{\rm c}$) and pores filled with air (with the refractive index $n_{\rm a}$):

$$n_{\rm p} = \frac{V_{\rm c}^{1/3} n_{\rm c} + V_{\rm a}^{1/3} n_{\rm a}}{V_{\rm c}^{1/3} + V_{\rm a}^{1/3}}, \quad (2)$$

where $V_{\rm c}$ and $V_{\rm a}$ are the volumes occupied by cellulose fibres and pores. Taking into account that the paper porosity $p = V_{\rm a}/V_{\rm p} = 1 - (\rho_{\rm p}/\rho_{\rm c})$, where $V_{\rm p}$ is the paper volume, $\rho_{\rm c}$ and $\rho_{\rm p}$ are the cellulose and paper density, respectively, and $1 - p = V_{\rm c}/V_{\rm p}$, expression (2) can be written in the form

$$n_{\rm p} = \frac{n_{\rm c} + [p n_{\rm a}/(1-p)]^{1/3}}{1 + [p/(1-p)]^{1/3}}. \quad (3)$$

The refractive index of cellulose is $n_{\rm c} \approx 1.5$ [13] and its density is $\rho_{\rm c} = 1.5$ g cm^{-3}, while the refractive index of air is $n_{\rm a} \approx 1$. The calculated values of the refractive index for both types of paper are presented in Table 1, where the paper porosity is also given.

By approximating the dependence of $1/\tau_0$ on the paper thickness by expression (1) (Fig. 3) and using the characteristic absorption time $\tau_{\rm a}$ as a fitting parameter, we can calculate the diffusion coefficient of photons in the medium and the reduced scattering coefficient of the medium (Table 1). As a result, we obtain the absorption time $\tau_\alpha = 1.49 \pm 0.22$ ps, which corresponds to the absorption

Table 1.

Paper type	Paper density/g cm^{-3}	Thickness/μm	Refractive index	Absorption coefficient/mm^{-1}	Reduced scattering coefficient/mm^{-1}	Mean free path/μm	Porosity
Writing	0.837	80	1.26	2.81 ± 0.37	62.3 ± 3.2	0.8	0.44
Porous	0.4	80	1.2	2.67 ± 0.36	46.8 ± 2.6	1.01	0.73

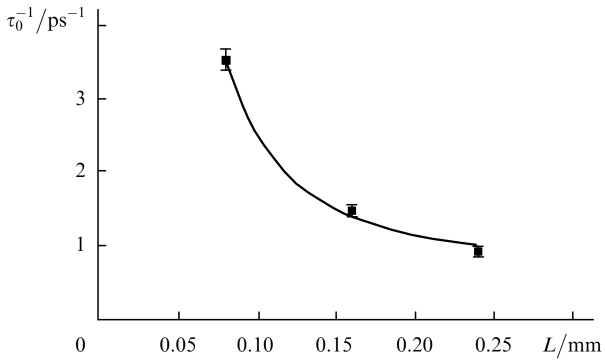

Figure 3. Dependence of the inverse lifetime of photons in the medium on the medium thickness (approximation by the expression $1/\tau_0 = 1/\tau_\alpha + (\pi/L)^2 D$ with $\tau_\alpha = 1.49$ ps and $D = 0.00188$ mm^2 ps^{-1}.

coefficient $\alpha = 2.67 \pm 0.36$ mm^{-1}, the diffusion coefficient $D = (1.88 \pm 0.11) \times 10^{-3}$ mm^2 ps^{-1}, and the reduced scattering coefficient $\mu_s' = 46.8 \pm 2.6$ mm^{-1}. By using the anisotropy parameter $g = 0.95$ [7], we can calculate the unreduced scattering coefficient of the medium and the mean free path of a photon in the medium (Table 1).

By probing a defect of a writing paper of density 0.837 g cm^{-3} (Fig. 2a) and size $\sim$ 1 mm, which had a great porosity, we determined the diffusion coefficient of photons equal to $(3.4 \pm 0.12) \times 10^{-3}$ mm^2 ps^{-1}. This coefficient outside the defect was $(1.2 \pm 0.11) \times 10^{-3}$ mm^2 ps^{-1}. This result suggests that the method proposed here can be used to control porosity and detect defects in statistically inhomogeneous media.

Therefore, the prolonged trailing edge of a backscattered femtosecond laser pulse contains information of the diffusion dynamics of photons. The special feature of the object studied is a great difference between the refractive indices of cellulose fibres forming a densely packed fibrillar chain and of the ambient air. Therefore, a long lifetime of photons in the porous medium or the 'capture' of light in the target volume is caused by efficient volume reflection of photons from scattering 'centres'.

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References

1. Dougherty A., Natan C. *Phys. Rev. E*, **58** (3), 2889 (1998).
2. Brezinski M.E., Fujimoto J.G. *IEEE J. Sel. Top. Quantum Electron.*, **5** (4), 1185 (1999).
3. Watson J., Fleury P., McCall S. *Phys. Rev. Lett.*, **58**, 945 (1987).
4. Gómez Rivas J., Sprik R., Lagendijk A., et al. *Phys. Rev. E*, **63**, 046613 (2001).
5. Johnson P., Imhof A., Bret B., Rivas J., Lagendijk A. *Phys. Rev. E*, **68**, 016604 (2003).
6. Zimnyakov D.A., Tuchin V.V. *Kvantovaya Elektron.*, **32**, 849 (2002) [*Quantum Electron.*, **32**, 849 (2002)].
7. Carlsson J., Hellentin P., Malmqvist L., et al. *Appl. Opt.*, **34** (9), 1528 (1995).
8. Bestemyanov K.P., Gordienko V.M., Konovalov A.N., Podshivalov A.A. *Program and Abstracts XI Conf. Laser Optics* (St.-Petersburg, Russia, 2003) p. 79; Bestemyanov K.P., Gordienko V.M., Konovalov A.N., Podshivalov A.A. *Program and Abstracts I Russian-Finnish Seminar: Photonics and Laser Symp.* (Saratov, Russia, 2003) p. 10.
9. Gordienko V.M., Ivanov A.A., Konovalov A.N., Podshivalov A.A., Pryalkin V.I., Savel'ev A.B. *Kvantovaya Elektron.*, **32**, 511 (2002) [*Quantum Electron.*, **32**, 511 (2002)].
10. Beaud P., Schultz J., Hodel W., et al. *IEEE J. Quantum Electron.*, **25**, 755 (1989).
11. Bestemyanov K.P., Gordienko V.M., Ivanov A.A., Konovalov A.N., Magnitskii S.A., Podshivalov A.A., Tursynov Zh.S. *Book Abstracts XI Intern. Laser Phys. Workshop* (Bratislava, Slovak Republic, 2002) p. 240.
12. Gordienko V.M., Konovalov A.N., Magnitskii S.A., Tursynov Zh.S. *Kvantovaya Elektron.*, **31**, 83 (2001) [*Quantum Electron.*, **31**, 83 (2001)].
13. Grigor'ev I.S., Meilikhov E.Z. (Eds) *Fizicheskie velichiny. Spravochnik* (Handbook of Physical Quantities) (Moscow: Energoatomizdat, 1991) p. 786.