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Christiansen effect in disperse systems with resonant absorption

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Abstract. **We discuss the results of experimental studies of competition of absorption and scattering of laser radiation propagating in dispersive media with resonant absorption. As media under study, use is made of a suspension of polystyrene particles in solutions of rhodamine 6G in ethylene glycol probed by laser light with a wavelength of 532 nm. It is found that an increase in the dye concentration leads to an increase in optical transmittance of suspensions and an increase in speckle modulation of the forward-scattered radiation. We interpret these features as a manifestation of Christiansen effect in disperse systems with resonance absorption.**

Keywords: *speckles, scattering, absorption, laser light, dispersive media, Christiansen effect.*

1. Introduction

Propagation of light in randomly inhomogeneous media is accompanied by fundamental phenomena arising from the interference of partial components of scattered light field in the medium [1]. Among these phenomena are coherent backscattering, sometimes interpreted as a phenomenon of weak localisation of the electromagnetic field [2, 3], the occurrence of temporal, spatial and frequency correlation of light fields, the generation and amplification of laser radiation in randomly inhomogeneous media (random lasing). The effect of laser generation takes place in the case of multiply scattering disperse systems containing components with a high quantum yield of fluorescence under their pumping by external radiation sources. This effect considered by Letokhov in 1967 [4] has become the subject of intense research in the last two decades [5–8].

Spatial scales of interaction of radiation with a medium l^*, l_s and l_a are frequency-dependent (here, l^* is the transport mean free path of radiation propagation in a medium, l_s is the scattering mean free path, l_a is the absorption length [9,10]; active randomly inhomogeneous media in which the effect of laser generation is possible are also characterised by the gain length l_{gain} [11]). This is due to the influence of the wave parameter ka of scattering centres (k is the wave number of radiation propagating in the medium, a is the characteristic size of scattering centres) on the scattering efficiency Q_{sca} [12] and the scattering anisotropy g (and, hence, on l^* and l_s), and also due to the influenced of the selective absorption of the medium (on l_a). For laser media with a random structure, the controlled changes in the frequency dependences of the absorption coefficient μ_a (and, hence, $l_a = \mu_a^{-1}$) and fluorescence quantum yield open up new avenues for controlling the generation process. This can be done by changing the volume fractions of the components in a mixture of two dyes with different quantum yields – in a gain medium in a disperse system [13]. The maximum sensitivity of the transport characteristics of the medium to changes in wavelength and structural parameters is observed in the spectral regions where resonance effects manifest themselves upon scattering and absorption.

In some cases, changes in the absorption coefficient of a component of the disperse system can significantly affect the radiation scattering in it because of strong interaction of real and imaginary parts of the refractive index of the absorbing component near the resonance absorption. This can lead to significant changes in the transport characteristics of the system and, consequently, influence the generation of laser radiation in it. Similar phenomena should also be taken into account when developing methods for laser probing of biological tissues in the absorption bands of natural chromophores (e.g., haemoglobin) and of those artificially administered into the tissue (photosensitizers) [14, 15].

The aim of this work is to study the effect of partial bleaching of multiply scattering disperse systems based on suspensions of polystyrene particles in solutions of rhodamine 6G in ethylene glycol with increasing dye concentration in the case of probing the systems by 532-nm laser radiation (near the absorption maximum of the dye solution in ethylene glycol).

2. Experimental

The test samples represented layers of suspensions of polystyrene particles of diameter 1 μm (the rms deviation of particle diameters was no more than 0.05 μm, the host medium was a solution of rhodamine 6G in ethylene glycol) in flat glass cells of thickness 70 μm. The volume fraction of particles in the suspensions was 0.06, the concentration of the dye in the host medium varied from zero to 2.15×10^{-3} mol L^{-1}. During the preparation, the suspensions were homogenised by ultrasound, then the required volume of the suspension was placed between two slides, which were separated by spacers of thickness 70 μm and acted as the cell walls. During the experiment, the cells were fixed vertically.

Figure 1 shows the experimental setup used for the analysis of small-angle forward scattering of laser radiation by the specimens under study. The samples placed on a high-preci-

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sion translation stage were irradiated by a slightly divergent laser beam formed by a telescopic system with a spatial filter (aperture of diameter 10 μm). Forward-scattered radiation was detected with a micro-objective consisting of an M-42 microscope ($8^{\times}$, numerical aperture of 0.2) and a receiving aperture, whose role was fulfilled by the entrance end of a fibreoptic patch cord (diameter 50 μm) connected to the photodetector (Hamamatsu H7468). The microscope was adjusted so that during transverse scanning of the sample, the medium–glass interface facing the micro-objective was optically conjugate to the image plane, where the entrance end of the patch cord was located. The distance between the micro-objective and image plane was 194 mm. The sample was scanned with a step of 1 μm; at each step of scanning, the signal was recorded. The samples were probed by 532-nm laser radiation (linearly polarised frequency-doubled diode-pumped Nd : YAG laser with an output power of 2 mW). For comparison, some samples were also probed using 633-nm radiation of a He–Ne laser. To increase the signal-to-noise ratio, an interference filter for $\lambda_0 = 532$ or 633 nm was placed before the entrance end of the patch cord (depending on the laser used).

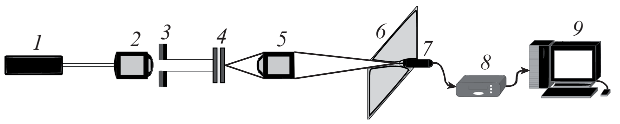

Figure 1. Scheme of the experimental setup: (*1*) laser; (*2*) telescopic system – beam expander with a spatial filter; (*3*) aperture limiting the probe beam; (*4*) cell with the sample under study; (*5*) micro-objective; (*6*) removable interference filter; (*7*) fibreoptic patch cord; (*8*) photodetector; (*9*) computer.

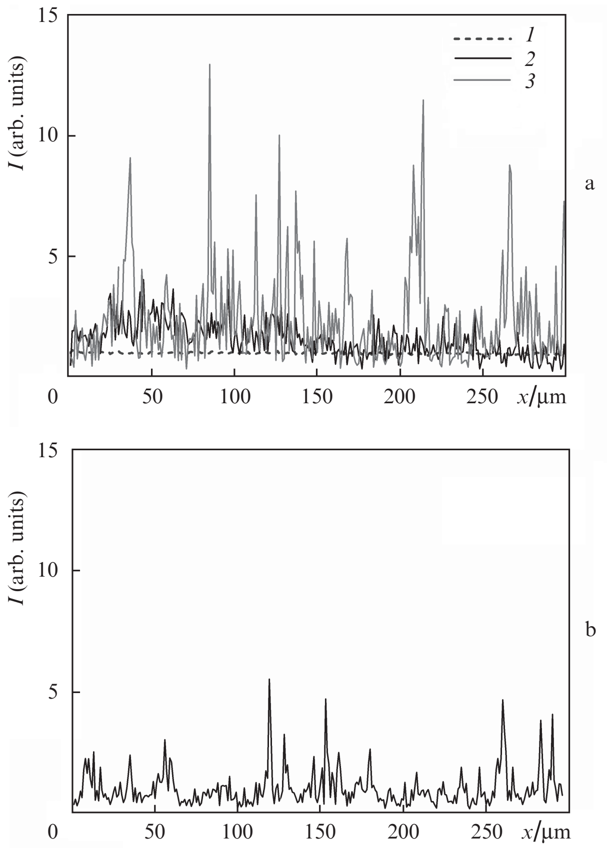

Figure 2. Dependences of the optical signal intensity I on the transverse displacement of the sample, x, upon scanning for $\lambda_0 = 532$ nm and concentration of rhodamine 6G in the host medium $c = 0$ (*1*), 7.05×10^{-4} (*2*) and 2.15×10^{-3} (*3*) (a), and for $\lambda_0 = 633$ nm and $c = 2.15\times10^{-3}$ mol L^{-1} (b).

Figure 2a shows the dependences of the intensity I of the detected optical signal at $\lambda_0 = 532$ nm on the transverse displacement of the sample at various concentrations of rhodamine 6G in a dispersive medium, whereas Fig. 2b presents a similar dependence at $\lambda_0 = 633$ nm, obtained at a maximum concentration of the dye ($c = 2.15\times10^{-3}$ mol L^{-1}). It should be noted that for the dependences demonstrated in Fig. 2a, we observe an increase both in the average intensity of the detected signal $\langle I \rangle$ and in the rms value of σ_I of the intensity fluctuations $I - \langle I \rangle$ with increasing c; in this case, σ_I increases essentially greater than $\langle I \rangle$.This leads to a significant speckle modulation of the optical signal at the maximum dye concentrations in the experiment. Thus, for the data presented in Fig. 2a, the normalised intensity $\langle I \rangle / \langle I \rangle_{c=0}$ and contrast $\sigma_I / \langle I \rangle$ of the speckles modulating the detected optical signal are equal to 1 and 0.04 at the zero dye concentration, to 1.55 and 0.75 at $c = 7.05\times10^{-4}$ mol L^{-1} and to 2.4 and 0.82 at $c = 2.15\times10^{-3}$ mol L^{-1} (the value of $\langle I \rangle_{c=0}$, used to normalise the average signal intensity at $\lambda_0 = 532$ nm, corresponds to the zero concentration of the dye). The contrast of the speckles, shown in Fig. 2b and obtained at a maximum concentration of the dye and $\lambda_0 = 633$ nm, is equal to 0.73 (the average intensity in this case was not normalised).

We should note the absence of 'fast' dynamics of the speckle inherent in suspensions and caused by the Brownian motion of scattering centres. We observed slow decorrelation of the speckle patterns in the detection region, which, in these experiments, is typical for samples with a fairly stable structure (*in vitro* biological tissues and polymer materials) and results from fluctuations of the laser radiation frequency, temperature instabilities, etc. These low-frequency fluctuations of the speckles had no significant effect on the process of their registration, because the characteristic decorrelation time markedly exceeded the scan time of a path segment having the length of the order of an average speckle size. The absence of pronounced 'Brownian' dynamics of the speckles is due, in our opinion, to the high viscosity of the host medium, to large enough volume fraction of scattering centres and to the space confinement of disperse systems under study.

3. Discussion of experimental results

At first glance, the increase in the average intensity of the forward-scattered laser light with increasing dye concentration in the probed disperse system is unusual. This effect can be interpreted on the basis of an analysis of the combined effect of the host medium absorption and scattering by particles on the transport properties of disperse systems. At a fixed concentration f of scattering centres of radius r in the system, in the weak scattering limit we have $\mu_s = 3Q_{sca} f/(4r)$, where Q_{sca} depends on the wave parameters kr and the real parts of the refractive indices of the host medium n'_b and the scattering centres n'_p (for the dielectric scattering particles we suggest that the imaginary part of the refractive index is $n''_p = 0$). In addition, Q_{sca}, under certain conditions, can be significantly influenced by the imaginary part of the refractive index of the host medium n''_b, determined by its absorption coefficient μ_a.

A theoretical analysis of the influence of the absorption coefficient μ_a of a medium containing spherical scatterers on the scattering efficiency is given in a number of papers [16–18], where a modified Mie theory is used to calculate the cross sections of a dielectric sphere immersed in an absorbing medium. Effect of absorption on Q_{sca} increases with kr, resulting in a substantial decrease in the scattering efficiency for large particles.

Another reason that leads to a decrease in Q_{sca} with increasing dye concentration in the host medium is the increase in n'_b, manifested most significantly for wavelengths near the maximum absorption of the medium (in the range corresponding to the transition from the segment of the dispersion characteristics of the medium with anomalous behaviour to the region of normal dispersion). For disperse systems with $n'_p > n'_b$ it will lead to suppression of scattering with increasing μ_a. To some extent, this effect is similar to the classical Christiansen effect (optical bleaching of two-phase disperse systems in the spectral intervals corresponding to the matching of the real parts of the refractive indices of different phases), which is the basis of the theory and practice of synthesis of dispersion spectral filters [19, 20].

In order to analyse the influence of these reasons on the observed features of small-angle laser radiation scattering (Fig. 2), we additionally measured n'_b and n''_b as functions of the concentration c of rhodamine 6G in the host medium at $\lambda_0 = 532$ nm. The dependence $n''_b(c)$ was obtained with the help of spectrophotometric measurements by the standard method for sample of the host medium without scatterers. It should be noted that the absorption maximum of the dye solution in ethylene glycol, caused by the electronic transition $S_0 \to S_1$ of the molecules of rhodamine 6G [21], corresponds to $\lambda_0 \approx 530$ nm, i.e., when using a 532-nm laser radiation as a probe, we can talk about its resonance absorption in the probed medium. The dependence $n'_b(c)$ was found from measurements of the reflection coefficient at the host medium – free space interface at normal incidence of the laser beam in accordance with the method of [21]. The obtained dependences $n''_b(c)$ and $n'_b(c)$ allow linear approximation with a high degree of accuracy. In this case, the coefficients $\partial n'_b/\partial c$ and $\partial n''_b/\partial c$ at $\lambda_0 = 532$ nm are equal to 16.7 and 0.364 L mol^{-1}, respectively. The value of $n'_b \approx 1.448$ measured in the absence of the dye agrees well with data for ethylene glycol (1.45) from paper [22]. For polystyrene, the value of $n'_p \approx 1.599$ was reported in [22].

Using the values of $n'_b(c = 0)$, $\partial n'_b/\partial c$, $n''_b(c = 0)$, $\partial n''_b/\partial c$ and n'_p (at $\lambda_0 = 532$ nm) obtained in the experiment and taken from [22], we estimated the effect of absorption of the host medium and the dependence $n'_b(c)$ on the efficiency of scattering of polystyrene spheres in a ethylene glycol – rhodamine 6G mixture. It was found that, in accordance with a modified Mie theory for dielectric spheres in an absorbing medium [18], in the case of the disperse system under study, a decrease in Q_{sca} even at the highest concentration of rhodamine in the range of the used values of the dye concentration does not exceed 4% of Q_{sca} when $c = 0$. Much more significant is the effect of suppression of scattering in a disperse system at $n'_b \to n'_p$ (Christiansen effect). The analysis was carried out by studying the dependence of Q_{sca} and of the scattering anisotropy parameter g on n'_b at $n'_p = 1.599$ in the framework of the Mie theory, the effect of n''_b on Q_{sca} and g being neglected. This approach is associated with the concept of independence of the radiation diffusion coefficient D of a randomly inhomogeneous medium, which is introduced within the framework of the diffusion approximation of radiative transfer theory [9], on the absorption coefficient of the medium [23–25]. Based on a comparison of experimental data with theoretical results obtained in the framework of this concept, it was shown that such ideas can interpret the experimental data with a higher accuracy compared with the classical diffusion model of radiative transfer, considering D as a function of μ_a [9, 26]. In this case, the absorption effect is taken into account by representing the field scattered in the medium as a superposition of partial components propagating along statistically independent paths of length s, and by introducing the Bouguer factor $\exp(-\mu_a s)$ for each component [27].

Figure 3 shows the theoretical dependences of the transport characteristics of the investigated disperse system [the scattering coefficient, $\mu_s = l_s^{-1}$; the transport scattering coefficient, $\mu'_s = (l^*)^{-1}$; the scattering anisotropy parameter, g; and the extinction coefficient, $\mu_t = \mu_s + \mu_a = (l_s + l_a)/(l_s l_a)$] on n'_b. It should be noted that in our case there is a rather specific regime of multiple small-angle forward scattering, characteristic of disperse systems with a high scattering anisotropy: the ratio of the cell thickness L to the scattering mean free path of probe radiation in the medium is $L/l_s = L\mu_s \gg 1$, while the ratio of the cell thickness to the transport mean free path is $L/l^* = (1 - g)L\mu_s \leqslant 1$. In our experiment, the value of L is such that when the volume fraction of the scattering centres in probing disperse systems is equal to 0.06 at $\lambda_0 = 532$ nm, there occurs a transition between the diffusion regime of radiation propagation and the regime of multiple small-angle forward scattering. In the latter case, the detected optical signal exhibits mainly near-axis components, propagating due to scattering at small angles to the axis of the probe beam (in the English literature such components called snake-like photons or zigzag photons [28]). Despite the increase in n''_b (and, consequently, μ_a), the extinction length $l_t = \mu_t^{-1} = (\mu_a + \mu_s)^{-1}$ and transport mean free path l^* decrease with increasing c, which leads to the experimentally observed increase in the average intensity of forward-scattered radiation.

Statistic simulation of radiation transfer in these samples made it possible to obtain the ratio of the average intensities of the detected optical signal at the maximum and zero concentrations of rhodamine 6G, equal to $\langle I \rangle_{c=c_{max}} / \langle I \rangle_{c=0} \approx 1.8$.

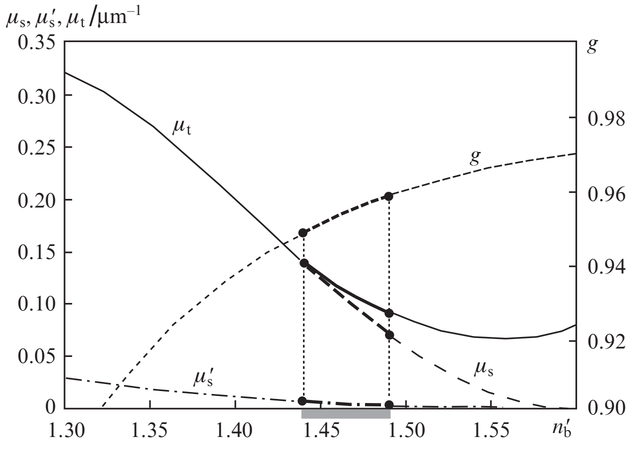

Figure 3. Theoretical dependences of the transport characteristics of the studied disperse systems on the real part of the refractive index of the host medium. The shading shows the range of n'_b values used in our experiment.

This value is somewhat smaller (by about 25%) than that obtained in the experiment (~2.4), which can be explained by differences in the behaviour of the Henyey–Greenstein phase function used in simulations [29] and the real phase function of scatterers with $g \geqslant 0.9$, as well as by deviations of the geometry of the experiment from the ideal geometry (in particular, small divergence of the probe beam). Nevertheless, statistical simulation using data from Fig. 3 adequately describes the experimentally observed behaviour of the investigated disperse system with increasing c.

The increasing role of the field components that are unscattered and scattered a few times with increasing n_b' also leads to the experimentally observed pronounced speckle modulation of the detected optical signal (Fig. 2). To analyse the effect of n_b' on speckle modulation of the optical signal, we introduced two parameters, i.e., Ξ and Θ. The parameter Ξ is the ratio of the number of unscattered ('ballistic') and scattered (a few times) partial components of the radiation propagating in a medium to the total number of partial components:

$$\Xi = \int_L^{L+\lambda_0} \rho(s)\,\mathrm{d}s \Big/ \int_L^{\infty} \rho(s)\,\mathrm{d}s,$$

where $\rho(s)$ is the probability density of the optical paths of partial components of the scattered field; L is the thickness of the probed layer. As a criterion for selecting the components scattered a few times, we selected the condition $L \leq s \leq L + \lambda_0$, i.e., we assumed that the main contribution to the speckle modulation is made by the interfering partial components propagating in a medium at a distance of the order of L with an optical path difference no greater than λ_0.

The parameter Θ is numerically equal to the product of the scattering spot diameter at the exit boundary of the layer upon its probing by a point source of collimated coherent radiation by the doubled average value of the deviation angle of the scattered field components coming out of the layer: $\Theta = 2\langle\theta\rangle\langle d\rangle$ (see inset in Fig. 4). The physical meaning of this parameter should be considered in more detail. As a result of multiple scattering of the light field produced by a coherent source with a δ distribution of the amplitude at the input boundary, the intensity distribution of scattered radiation with a characteristic size $\langle d\rangle$ is formed at the exit boundary; at each point of this region, the field amplitude is determined by the superposition of partial components with random phase values.

Thus, the scattering spot at the exit surface can be considered as an ensemble of phase-uncorrelated elementary sources of the light field with a characteristic size $\langle d\rangle$. By analogy with [30], where the criterion of interference pattern visibility from an extended quasi-monochromatic source with a finite interference aperture is studied, we impose some restriction on the parameter Θ: $\Theta \leq \lambda_0$. In the case $\Theta \gg \lambda_0$ and $\Xi \ll 1$, when the detected optical signal mainly exhibits diffusely scattered components with large path differences and large angles θ of deviation from the axis of the probe beam, a small-scale speckle structure with the correlation radius of the order the wavelength must be formed in the plane of the detector aperture. At the finite size of the detector aperture, the detected signal fluctuations are suppressed. An additional contribution to the suppression of the fluctuation component will be made by depolarisation of multiply scattered laser light. In contrast, in the case of $\Theta \ll \lambda_0$ and $\Xi \approx 1$, the interference of unscattered components and components with small values of θ and $s \approx L$ will mainly contribute to the stochastic modulation of the optical signal. Accordingly, in the detector plane there will be formed relatively large-scale speckles with high contrast [curves (*2*) and (*3*) in Fig. 2].

Figure 4 shows the theoretical dependences of Θ and Ξ on n_b' for the studied disperse system, obtained by statistical simulation of the probe radiation transfer. Restriction imposed on the volume fraction of particles in the synthesis of disperse systems in order to obtain the transport mean free path $l^* \approx (2 \div 3)L$ made it possible to obtain the criterion $\Theta = \lambda_0$ and to pass from $\Xi \ll 1$ to $\Xi \approx 1$ in the range of c values used in our the experiment. Qualitative agreement between experimental results and simulation is evident (Figs 2 and 4).

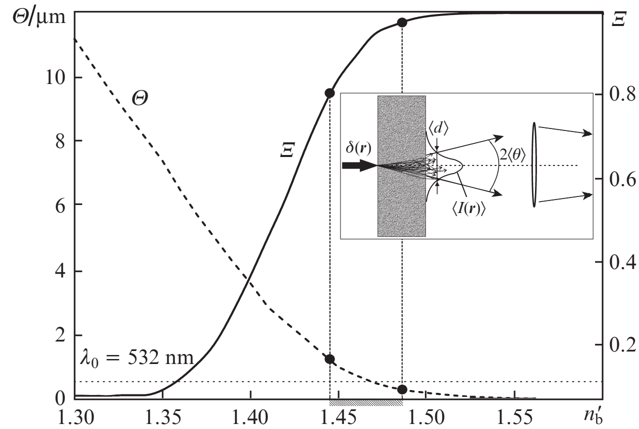

Figure 4. Dependences of the parameters Θ and Ξ for the studied disperse system on the real part of the refractive index of the host medium (the results of statistical simulation). The shading shows the range of n_b' values used in our experiment. The inset illustrates the principle of introducing the parameter Θ for the scattering geometry used.

Analysis of the relationships between the parameter L/l^* and speckle contrasts measured in the experiment (Fig. 5) suggests the existence of a correlation of these variables in the region of transition between the two propagation regimes. Note that in the case of probing by 633-nm radiation at a maximum concentration of the dye, the value of l^* was evalu-

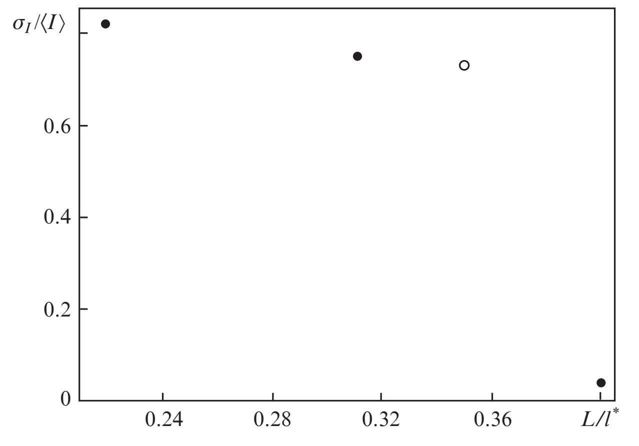

Figure 5. Relationship of speckle contrast $\sigma_I/\langle I\rangle$, observed in the image plane of the medium–cell wall interface and the parameter L/l^* for the studied disperse system at λ_0 = 532 (●) and 633 nm (○).

ated using the Mie theory, based on the assumption that n'_b for a solution of rhodamine 6G in ethylene glycol away from the absorption maximum behaves similarly to n'_b for this dye in a different solvent (methanol [21]). Based on this assumption, we can assume that the difference between n'_b $(c = c_{max})$ $- n'_b$ $(c = 0)$ at $\lambda_0 = 633$ nm is about 30%–35% from the corresponding value at $\lambda_0 = 532$ nm. The parameter $L/l^* \approx 0.35$ obtained using such approximate estimates for $c = 2.15 \times 10^{-3}$ mol L^{-1} and $\lambda_0 = 633$ nm is in good agreement with the $L/l^* = 0.311$ for $c = 7.05 \times 10^{-4}$ mol L^{-1} and $\lambda_0 = 532$ nm; in this case, the experimentally measured speckle contrasts are also similar (Figs 2 and 5).

4. Conclusions

Experimental studies and statistical simulation of the laser radiation transport in the dye-saturated disperse systems have shown that in the region of maximum absorption of the dye different tendencies in the behaviour of absorption and scattering coefficients of the disperse system with increasing dye concentration are caused by the partial optical bleaching of the system. This behaviour of the coefficients can be interpreted as a manifestation of Christiansen effect in dispersive media with absorption. Competition between scattering and absorption in these scattering systems can play an important role in the case of the synthesis of laser media with a randomly inhomogeneous structure, because the increase in the volume fraction of the dye with a high quantum yield of fluorescence will reduce the characteristic length of the amplification of radiation in the medium [11]. On the other hand, the effect discussed in this paper may lead to an increase in the critical thickness of the medium, necessary to ensure the conditions for generation, and to increase in the lasing threshold.

It should be noted that in case of saturation of the dye with the resonant absorption by scatterers rather than by a host medium, the increase in the dye concentration in the material of the particles should exhibit the opposite effect, corresponding to the trivial behaviour of the dispersive medium (increase in extinction with increasing concentration of the absorber).

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