CHARACTERISTIC SCALES OF OPTICAL FIELD DEPOLARIZATION AND DECORRELATION FOR MULTIPLE SCATTERING MEDIA

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ABSTRACT

Decorrelation and depolarization properties of multiply scattering media and tissues in the case of propagation of coherent probe beams are analyzed in terms of photon path distribution. A specific correlation time determining the relationship between correlation and polarization states of scattered optical fields is introduced. Results of correlation and polarization experiments with phantom scatterers (such as water suspensions of polystyrene spheres) and tissues with controlled optical properties (such as the human sclera) are presented. © 1999 Society of Photo-Optical Instrumentation Engineers. [S1083-3668(99)00901-6]

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1 INTRODUCTION

Significant interest to the investigations of polarization phenomena accompanying light propagation through optically dense media is caused by the possible applicability of these effects for tissue characterization and imaging in biology and medicine.1–4 One of the general properties of a multiple scattering medium which is observed in the experiments with a coherent probe beam is the formation of local polarization structure of the scattered optical field.5,6 Similar structures are correlated with small-scale intensity patterns of this field (usually named speckle patterns). The characteristic size of a coherence area, or an average size of speckles, is comparable with the wavelength used, if multiple scattering takes place. For a given polarization state of the probe beam we have random local polarization state of the scattered field in an arbitrarily selected observation point which stochastically changes from one point to another. If a probe beam with a linear polarization state is used, one of the convenient approaches for description of the scattered field is its interpretation as a superposition of linearly polarized uncorrelated speckle patterns with orthogonal directions of electric fields (copolarized and cross-polarized components; azimuth direction of the copolarized component is the same as that of the probe beam). In this case the resulting scattered field randomly changes its local polarization state (linear, elliptical, or circular) in the dependence on spatial distributions of amplitude and phase of copolarized and cross-polarized components. Such a generalized parameter of the total scattered field as its polarization degree can be introduced as

\[ P = \frac{\langle I_\parallel \rangle - \langle I_\perp \rangle}{\langle I_\parallel \rangle + \langle I_\perp \rangle}, \tag{1} \]

where \( \langle I_\parallel \rangle \) and \( \langle I_\perp \rangle \) are the mean intensities of the copolarized and cross-polarized components obtained by an ensemble averaging. It is necessary to note that for the given approach, when both uncorrelated components of the scattered field can be interpreted as totally developed speckle patterns with contrast introduced as (see, e.g., Ref. 7)

\[ V_{\parallel\perp} = \frac{\langle I_{\parallel\perp}^2 \rangle}{\langle I_{\parallel\perp} \rangle^2}, \tag{2} \]

which is equal to 2, the total scattered field (as sum of both components) in the case of an optically dense multiply scattering medium should be characterized by the value of contrast \( V_{\parallel\perp} = 1.5 \) (see the Appendix). For nonstationary scattering media such as ensembles of Brownian particles, averaging can be carried out over the time series of the intensity fluctuations of both components detected in the fixed observation point. For “steady-structure” scatterers and fixed point observation conditions, detected intensity fluctuations can be induced by scatterer transverse movement with respect to the probe beam and detector.

Normalized autocorrelation function of the scattered field,

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and autocorrelation function of intensity fluctuations,
\[ g_2(\tau) = \langle I(0)I(\tau) \rangle / \langle I(0) \rangle^2, \]
are traditionally used for characterization of coherent light interaction with multiple scattering media and quantification of the scattering ensembles of moving particles (see, e.g., Refs. 8–10). It is well known that for scattered fields with Gaussian statistics of amplitude induced by statistically homogeneous and ergodic scattering systems \( g_1(\tau) \) and \( g_2(\tau) \) are related with each other through the Siegert relation\(^\text{11}\)
\[ g_2(\tau) = 1 + \beta \left| g_1(\tau) \right|^2, \quad (3) \]
where parameter \( \beta \) depends on field formation and detection conditions. In the case of single scattering by the ensemble with a great number of particles and ideal detection conditions (when detector aperture is much less than the average size of speckles) we have \( \beta = 1 \). For multiple scattering mode with \( P \approx 0 \), when correlation properties of copolarized and cross-polarized components are similar, we should use the modified Siegert relation in the form
\[ g_2(\tau) = 1 + \beta \left| g_1(\tau) \right|^2 / 2 \]
(see the Appendix).

On the other hand, when turbidity of scattering media decreases, the polarization degree of scattered light becomes significantly different from zero and the copolarized component is dominant in comparison with the cross-polarized component. Differences between conditions of the formation of these components of the scattered field are also manifested in the decay parameters of their temporal autocorrelation functions. Recently, some manifestations of polarization effects in the correlation properties of multiply scattered dynamic speckles have become an object of significant research interest. In particular, unexpected behavior of intensity autocorrelation of “polarized” and “depolarized” components of scattered light has been observed for dense colloidal polyball crystals.\(^\text{12}\) The autocorrelation of intensity fluctuations of depolarized light decays for such systems in a short time as in a liquid but in the case of polarized light it exhibits a saturation at large times as expected for a frozen phase. Also, Dogariu et al.\(^\text{13}\) have been reported on depolarization measurements of short pulses of light backscattered from dense suspensions of silica microspheres. The relation between characteristic scale of depolarization and transport mean free path for considered scattering conditions has been found.

The goal of this work is to study the relations between correlation and polarization characteristics of scattered fields for nonsingle scattering media in the region of the transition from multiple-scattering mode of light transport to single-scattering mode. Water suspensions of polystyrene spheres and human sclera with artificially controlled optical parameters have been used as studied objects in correlation and polarization experiments.

## 2 RELATIONSHIP BETWEEN SCATTERED FIELD AUTOCORRELATION FUNCTION AND POLARIZATION DEGREE

One of the most convenient and universally adopted approaches in the characterization of scattering properties of turbid media is the usage of probability density function \( \rho(s) \), which describes the distribution of propagation paths of partially scattered waves forming the total scattered field and can be introduced in terms of the random walk model of light–media interaction (see, e.g., Ref. 9). For optically dense scattering media, when the diffusion mode\(^\text{14}\) of light transport through the scatterer takes place, \( \rho(s) \) can be interpreted as the probability density of the diffusing photon path distribution. In terms of propagation paths distribution all the statistical and correlation moments of scattered field, which describe in the general form its polarization and correlation properties, can be obtained as the integral transforms of \( \rho(s) \) with corresponding weighting functions. For example, it is well known that the field autocorrelation function for multiple scattering ensembles of Brownian spherical particles can be expressed as the Laplace transform of \( \rho(s) \),\(^\text{15}\)
\[ g_1(\tau) \sim \int_0^\infty \exp \left( -\frac{2\tau s}{\tau_0 I^*} \right) \rho(s) ds, \quad (4) \]
where \( \tau_0 \) is the characteristic time, defined by the expression \( \tau_0 = k_0^2 D_B / I^* \); \( D_B \) is the diffusion coefficient for Brownian particles, \( k_0 \) is the scattering vector module, and \( I^* \) is mean transport length for light propagation through a scattering medium. \( \rho(s) \) depends on boundary conditions which describe the scattering object geometry as well as on the illumination and detection conditions.

More generally, use of the above mentioned random walk approach in the description of coherent light transport through multiply scattering nonstationary media leads us to the following expression for the field autocorrelation function:\(^\text{9}\)
\[ g_1(\tau) \sim \sum_n \rho(n) \exp \left[ -n \left( 1 - \frac{B(q, \tau)}{B(q, 0)} \right) \right], \quad (5) \]
where \( \rho(n) \) is the probability density function of scattering events number \( n \) for each propagation path; \( B(q, \tau) \) is determined by the product of the so-called dynamic structure factor \( S(q, \tau) \) and the form factor \( |b(q)|^2 \) (see Ref. 9): \( B(q, \tau) = k_0^4 N |b(q)|^2 S(q, \tau) \), where \( N \) is the number of scattering particles. If the interparticle spacing is much larger than the wavelength and particle size, we can use the following approximation:
where \( W(\tau) \) is the mean-square particle displacement as a function of time delay and average \( \langle \hat{q}^2 \rangle \) is weighted by form factor.\(^9\) Thus, in the continuous limit we will obtain

\[
g_1(\tau) \sim \int_0^\infty \exp[-s\langle \hat{q}^2 \rangle W(\tau)/6l] \rho(s)ds, \quad (7)
\]

where \( l \) is the photon mean free path length in a scattering medium. Using the following expression for the ratio of \( l \) and \( l^* \): \( l^* / l = 2k_0^2 / \langle \hat{q}^2 \rangle \), we go to the final form of this expression:

\[
g_1(\tau) \sim \int_0^\infty \exp[-sk_0^2W(\tau)/3l^*] \rho(s)ds. \quad (8)
\]

Thus, we should expect the quadratic dependence of the weighting function \( r_0(s) \) on \( s \) in the case of regular movement of the multiple scattering "steady-structure" medium.

On the other hand, polarization degree of light scattered by multiple scattering media can be expressed as

\[
P = \int_0^\infty \varphi_p(s) \rho(s)ds, \quad (9)
\]

where weighting function \( \varphi_p(s) \) monotonically decreases as \( s \) increases. It had been shown in Ref. 13 by using the estimations from the Bethe–Salpeter equation that the polarization degree of the optical wave, which undergoes the sequence of scattering events, can be expressed in the dependence of the number of events as

\[
P = \frac{\sum_n \rho(n)P(n)}{\sum_n \rho(n)},
\]

Thus, in terms of photon path distribution, the weighting function \( \varphi_p(s) \) can be written as

\[
\varphi_p(s) = \exp(-s/kl).
\]

Term \( \eta = kl \) can be interpreted as the characteristic spatial scale of depolarization, where \( k \) depends only on optical properties of the single scatterers.

Thus, we can obtain finally

\[
P = \int_0^\infty \exp(-s/\eta) \rho(s)ds, \quad (10)
\]

where \( \eta \) determines propagation path required to destroy the initial polarization state of the probe beam. Comparing expressions (4), (8), and (10), we can see that for the exponential form of the weighting function \( \varphi_p(s) \) the equality of the polarization degree and field autocorrelation function for a certain fixed value of time delay \( \tau_{dp} \) should take place. In particular, for scattering systems consisting of noninteracting spherical Brownian particles this specific correlation time is equal to \( \tau_{dp} = \tau_0 l^* / 2\xi \). Taking into account that ratio \( l^* / \xi \) for the given scattering spheres does not depend on their concentration and is determined only by scattering properties of the single particle \( l^* \sim (\sigma d)^{-1} \), as well as \( \eta = 1 \) and \( l^* / l = 2k_0^2 / \langle \hat{q}^2 \rangle \), where \( \sigma \) is the scattering cross-section of the single particle and \( \phi \) is volume concentration of scattering particles, we should expect a constant value of \( \tau_{dp} \) for the wide range of concentration of scattering particles. Figure 1 illustrates the principle of evaluation of specific correlation time \( \tau_{dp} \), for which field autocorrelation functions for different volume concentrations of scattering particles \( \phi \) cross the levels determined by the corresponding polarization degrees.

### 3 EXPERIMENTAL RESULTS AND DISCUSSION

To verify such simple relations between characteristic scales of polarization and correlation decay in multiple scattering nonstationary media correlation experiments with phantom objects such as water suspensions of polystyrene spheres were performed. Experiments have been carried out with forward scattering geometry; a single-mode Ar laser operating at 514 nm wavelength was used as an illumination source; scattered light was collected by a single-mode fiber with a 5 \( \mu \)m core. To measure the polarization degree of scattered light in the detection point a manually rotating polarizer was placed between the cuvette with scattering suspension and fiber input aperture. Slab geometry of the scattering media with detection of the forward scattered light at zero scattering angle has been used. A cylindrical glass cuvette with 10 mm thickness and 60 mm diameter was filled with a water suspension of polystyrene particles with a given concentration.
Two kinds of scatterers have been used: with diameter $2a_1 = 0.460 \mu m$ and $2a_2 = 1.07 \mu m$; for the illumination conditions used (scattered light wavelength in water is equal to 385 nm; $k a_1$ is equal to 3.75 and $k a_2$ is equal to 8.73), this corresponds to a Mie scattering regime. For small volume concentrations of suspensions used in our experiments (less than 0.01) scattering particles can be considered as the uncorrelated spheres. Estimations of $L/I^*$ ratios (where $L$ is the cuvette thickness) made by using values of $I^*$ for water suspensions of polystyrene spheres previously obtained by other investigators (see, e.g., Ref. 17), have shown this value to be significantly larger than 1 for all used volume concentrations of particle suspensions. A photon-counting photomultiplier tube (Hamamatsu HC-120) and digital autocorrelator BIC-9000 (Brookhaven Instruments Corp., USA) were used for the detection of intensity fluctuations and processing of the detected signal.

Figure 2 displays the dependencies of specific correlation time $\tau_{dp}$ on volume concentration of polystyrene spheres $\phi$ in comparison with similar dependencies of the halfwidth of the scattered field autocorrelation peak (obtained by using the Siegert relation) via $\phi$. Despite the dramatic decrease of "conventional" correlation time $\Delta \tau_{0.5}$ (estimated as the half-width of the autocorrelation peak) with an increase of $\phi$, variations of $\tau_{dp}$ parameter in the analyzed region of scattering concentration are small enough to confirm the validity of the discussed relation between characteristic scales of polarization and correlation decay. Systematic diminishing of specific correlation time when $\phi > 1 \times 10^{-4}$ in the case of 1.07 $\mu m$ particles can be explained by the discrepancies in behavior of "long-tailed" parts of polarization and correlation weighting functions [$q_p(s)$ and $q_n(s)$, respectively]. Nevertheless, $\tau_{dp}$ changes very little (maximal variation is less than 15%) in comparison with $\Delta \tau_{0.5}$ [see Figure 2(b)]. Average values of specific correlation time in the used ranges of particle concentrations are equal to $\tau_{dp,0.46} = 1.04 \pm 0.03$ ms and $\tau_{dp,1.07} = 0.77 \pm 0.11$ ms, respectively.

Estimations of ratios $\xi/L$ made by using the experimental and simulation values of characteristic lengths of depolarization presented in Ref. 18 [this parameter had been introduced for characterization of the depolarization properties of scattering media in the case of slab geometry and is equal to $(\xi/L)_{1.07} \approx 1.0 \times 10^2$ and $(\xi/L)_{0.46} \approx 6.0 \times 10^2$. Taking into consideration that for 0.46 $\mu m$ particles calculated from Mie theory ratio $L/I^*$ is approximately equal to 26,15 we obtain $(\xi/L)_{0.46,0.46} = 3.84$.

On the other hand, using Brownian diffusion coefficients for particles equal to $D_{B,1.07,0.46,0.46} \approx 2.06 \times 10^{-9} \text{cm}^2/\text{s}, 5.35 \times 10^{-9} \text{cm}^2/\text{s}, 2.06 \times 9, 3.4$, respectively.

Coherent light scattering experiments with optically cleared human sclera6-22 also demonstrate simultaneous changes of correlation and polarization characteristics of scattered light caused by evolu-
tion of $\rho(s)$ with a decrease of the tissue scattering coefficient due to application of a special matching agent. In these experiments such a decrease is stimulated by administration of a special immersion agent—60% water solution of Trazograph (a derivative of the 2-4-6 triiodobenzene acid used as x-ray contrasting solution in medical applications) with a refractive index equal to 1.43 at 24°C. Interpretation of the optical clearing due to “matching” agent diffusion into the tissue volume has been discussed in detail in Refs. 20 and 21. Matching of the refractive indices of two basic components of sclera (collagen fibrils and “ground” substance—water solution of polysaccharides) leads to the dramatic changes of path distribution for photons traveling through the sclera layer even in the case of partially cleared sclera; thus, ballistic (unscattered) and “snake” (low-step scattered) photons significantly dominate in comparison with long-path photons (for more detail, see, e.g., results of Monte Carlo simulation cited in Ref. 21). Experiments have been carried out with samples of human sclera using the technique described in Ref. 20. An optical scheme of the experimental setup is presented in Figure 3. A single-mode He–Ne laser 1 is used as an illumination source. The probe beam is focused into the tissue volume by the microscope objective with a focal length equal to 33.1 mm and numerical aperture equal to 0.11. The sample under study (piece of sclera) is placed in the glass cuvette filled by Trazograph solution. Intensity fluctuations, which are caused by transverse movement (in the direction perpendicular to the axis of the illuminating beam) of the sample, are detected by a photomultiplier tube (PMT) with a pinhole. In order to select the linearly polarized component of the scattered field with a given azimuth orientation, a manually rotating polarization filter is placed before the pinhole. Movement of the studied sample is provided by the scanning device with a step motor; a minimal scanning step is equal to 5 μm; if driving frequencies larger than 500 Hz are used to control the scanning device, nonuniformity of the scanning velocity is less than 2.5% (except short initial and final parts of the scanning trace). The output photoelectric signal is converted to 12-bit digital form by using a DASH-16F analog-digital converter (ADC). To improve the spatial resolution of the intensity sampling procedure the following technique has been used: the ADC sampling frequency has been chosen much larger than the driving frequency of the scanning device. This allows us to obtain quasistationary movement of the studied sample value of the sampling step which is much less than the minimal value of the scanning step (5 μm). For example, if the ratio of frequencies is equal to 20, we have a sampling step equal to 0.25 μm. A typical length of scanning trace used in our experiments was equal to 10 mm. Time series of scattered light intensity fluctuations induced in the paraxial region by the sample scanning were sampled for two orthogonal positions of the polarization filter (cross-polarized and copolarized components of the scattered field) for different time intervals elapsed after Trazograph administration. After this normalized autocorrelation functions of intensity fluctuations for cross- and copolarized components as well as polarization degree of the scattered light were obtained for each set of intensity time series corresponding to different time delays after the beginning of the optical clearing.

We should note the qualitative difference between the above discussed case of dilution of the water suspensions of polystyrene spheres and optical clearing of sclera. In the later case with an increase of $l^s$ we will have the simultaneous changes of $\xi/l^s$ ratio due to the matching of refractive indices of collagen fibrils and ground substance. Thus, the specific correlation time will depend on the time elapsed after matching agent administration and this dependence can be used for analysis of the “slow” dynamics of the scattering structure during optical clearing.

Figure 4 shows the typical forms of the normalized autocorrelation functions of the intensity of
cross-polarized and copolarized components of the scattered field, which corresponds to the middle stage of sclera clearing. We can see the fast decaying main correlation peak associated with intensity dynamics of single speckles in the detection zone, and a slow decaying “tail,” caused by spatial inhomogeneities of the sclera optical properties along the scanning trace due to inhomogeneities of the matching agent concentration. Dependencies of correlation time $\Delta\tau_{0.5}$ for both linearly polarized components on polarization degree are presented in Figure 5 for an arbitrarily selected sclera sample. Peculiarity in the behavior of $\Delta\tau_{0.5}$ for the copolarized component (less values in comparison with the values for the cross-polarized component as well as a slow increase with an increase of polarization degree) in the case of cleared sclera can be explained by the significant contribution of the specular (unscattered) component for highly cleared tissue fragments appearing along the scanning trace. Such a contribution should strongly distort the form of the autocorrelation peak. Thus, in the presence of specular component separate analysis of cross-polarized and copolarized components can be used in polarization-correlation “forward-scattering” measurements for selection of respectively, the long-path and short-path parts of $s$ distributions.

4 CONCLUSIONS

Thus, specific correlation time $\tau_{dp}$ can be considered as one of the multiple scattering media that describes the connection between decorrelation and depolarization of coherent light propagating through media. Being evaluated from simultaneous polarization and correlation measurements this parameter equal to $\tau_{dp} = W^{-1}(3l^* / k_0^2 \xi)$ seems an attractive tool for analysis of averaged optical characteristics of individual scatterers in the nonstationary scattering ensemble as well as for analysis of peculiarities of ensemble dynamics. This can be made by the study of the behavior of $W(t)$ in the case of scattering media with controlled optical properties (particularly, with controlled ratio $l^*/\xi$); in this turn use of the controlling technique on the basis of solute-induced changes of tissue optical properties (see, e.g., Refs. 21 and 23) in combination with polarization and correlation measurements can be recommended for tissue structure diagnostics. The object of further investigations could be the connection between correlation and polarization properties of scattered light in the intermediate region between single- and multiple-scattering modes of the light propagation through tissue [when a significant contribution of specular (unscattered) and low-step scattered components in the optical paths statistics takes place].

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APPENDIX

Considering the dynamic scattered field in the observation point as sum of two uncorrelated linearly polarized cross-oriented and cooriented components (in the case of linearly polarized illuminating beam), we can express momentary value of detected intensity as

$$I_{\text{total}}(t) = I_\parallel(t) + I_\perp(t).$$

Calculating normalized autocorrelation function of $I_{\text{total}}(t)$, we obtain

$$g_2^{\text{total}}(\tau) = \frac{\langle I_{\text{total}}(t)I_{\text{total}}(t + \tau) \rangle}{\langle I_{\text{total}}(t) \rangle^2} = \frac{G_2^{\parallel}(\tau) + G_2^{\perp}(\tau) + 2\langle I_\parallel \rangle \langle I_\perp \rangle}{\langle I_\parallel \rangle^2 + \langle I_\perp \rangle^2 + 2\langle I_\parallel \rangle \langle I_\perp \rangle},$$

where $G_2^{\parallel}(\tau), G_2^{\perp}(\tau)$—unnormalized autocorrelation functions of the intensity fluctuations of the copolarized and cross-polarized components. In the case of uncorrelated and statistically identical fluctuations of co- and cross-components we can assume that $\langle I_\parallel \rangle = \langle I_\perp \rangle = \langle I_\parallel \rangle$. Thus,

$$g_2^{\text{total}}(\tau) = \frac{1}{2} \frac{1 + \beta |g_1^{\parallel}(\tau)|^2 + 1 + \beta |g_1^{\perp}(\tau)|^2}{2} + \frac{1}{2}.$$

For the same behavior of $|g_1^{\parallel}(\tau)|^2$ and $|g_1^{\perp}(\tau)|^2$ we have $g_2^{\text{total}}(\tau) = 1 + (\beta/2) |g_1^{\parallel}(\tau)|^2$ and for ideal detection conditions ($\beta$ is equal to 1) contrast is equal to 1.5.

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