Diffusing wave spectroscopy with a small number of scattering events: An implication to microflow diagnostics

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(Received 19 June 2004; revised manuscript received 16 February 2005; published 3 November 2005)

Limits of applicability of classical diffusing wave spectroscopy (DWS) are essentially extended. DWS is adapted to the case of small number of scattering events. An explicit formula for correlation function of intensity fluctuations of scattered light is derived. Potentials of DWS for diagnostics of random microflow are demonstrated.

DOI: 10.1103/PhysRevE.72.052902 PACS number(s): 87.64.Cc, 82.70.−y

I. INTRODUCTION

One of the methods that can be effectively applied for the diagnostics of highly scattering dynamic media is diffusing wave spectroscopy (DWS) [1–3]. In particular, DWS is used for *in vivo* blood flow diagnostics [4], study of microrheology of complex fluids [5], analysis of particles suspensions [6–8], foams and sands [9,10], etc. In general, DWS approaches the problem of multiple scattering in the limit when light is scattered a very large number of times. The breakdown of DWS is analyzed in Refs. [11,12].

Sometimes it is necessary to study the objects, which are characterized by a small number of scatterers, but, nevertheless, exhibit a very high degree of multiple scattering. Usually this situation is typical for biomedical applications. For example, it is important to provide laser measurements of velocity of the isolated random microflow in the single blood vessel (such as capillaries, smallest arterioles, or venules). Speckle or Doppler methods are powerful tools for the diagnostics of blood flow in microvessels with diameters less than 50 μm [13]. For vessels with a larger diameter, Monte Carlo simulations may be successfully applied for the analysis of the processes of light scattering [14,15]. Regretably, the Monte Carlo technique does not possess the analytical power of DWS to interpret the measured decay time of the correlation function of the scattered light. On the other hand, traditional DWS cannot be used in the case when the light undergoes only one or even a small number of scattering events. The failure of the DWS theory for samples of small thicknesses has been predicted theoretically [11] and observed experimentally [12].

For blood microvessels with diameter d up to 100 μm, the ratio between d and mean-free path l^* lies within the range $1<d/l^*<4$, and the scattering anisotropy factor g is close to 1. The analytic expressions, those that are accurate for the mentioned case, are not obtained in DWS. This Rapid Communication partially fills this gap. The theory of DWS is adapted to the case of a small number of scatterers. The explicit formula for correlation function of scattered intensity fluctuations of light is derived.

II. STATEMENT OF THE PROBLEM

Only the most basic elements of the theory of classical DWS are presented here; more detailed accounts are given in Refs. [16–18].

A photon, which propagates through the microflow, scatters N times (see Fig 1). The single photon, after passing from a laser source to the detector acquires the total phase shift $\psi(t)$,

$$\Psi(t) = k_0 \cdot s(t) = \sum_{i=0}^{N} \vec{k}_i \cdot \left[\vec{r}_{i+1}(t) - \vec{r}_i(t)\right], \tag{1}$$

where i is the running number of the scatterers that are involved into the scattering processes, N is the total number of scatterers interacting with the photon, $\vec{k}_i$ is the wave vector of scattered light after the i scattering events, $|\vec{k}_i|=k_0$, $\vec{r}_i$ is the radius vector of ith scattering particle, and $s(t)$ is the total path length of the photon.

The whole field at the observation point is the superposition of the fields from all light path through the microflow to the detector,

$$E(t) = \sum_{p} E_p \exp\left(i\Psi_p(t)\right), \tag{2}$$

where Σ_p represents the sum over path, and E_p and Ψ_p are the amplitude and phase from path p at the detector. In the multiple-scattering case, there are many scatterers for each light path, so the last relation involves two sums: one over light paths, another over the scattering events in each path [17].

The field autocorrelation function is

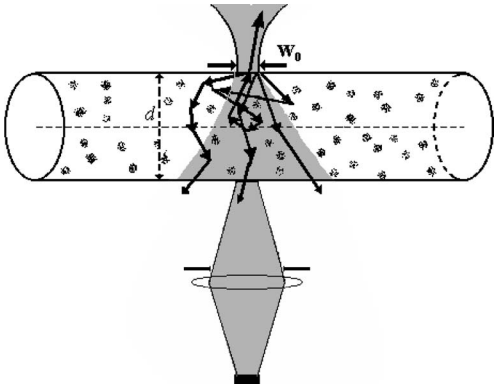

FIG. 1. Scheme of photon migration in microflow with a small number of scattering events.

$$g_1(t) = \frac{1}{\langle I \rangle} \left\langle \left[\sum_p E_p \exp\left(i\Psi_p(0)\right) \right] \sum_p E_p^* \exp\left(-i\Psi_p(t)\right) \right\rangle, \tag{3}$$

where $\langle I \rangle$ is the total average scattered intensity at the detector, * is the symbol of complex conjugation. For independent scatterers, the fields from the different paths are not correlated, so

$$g_1(t) = \sum_p \frac{\langle I_p \rangle}{\langle I \rangle} \cdot \left\langle \exp\left[i\left(\Psi_p(0) - \Psi_p(t)\right)\right] \right\rangle, \tag{4}$$

where $\langle I_p \rangle = \langle |E_p|^2 \rangle$ is the average intensity from path p. The phase Ψ_p and field amplitude E_p are assumed to be independent at the detector.

The last relation is treated in DWS as a sum over path length rather than a sum over individual path, provided that it replaces the fraction of scattered intensity in paths of length s. Also,

$$\Delta\Psi_p \equiv \Psi_p(0) - \Psi_p(t) = \sum_{i=1}^{N} 2 \cdot k_o \sin\left(\frac{\Theta_i}{2}\right) \Delta r_i(t), \tag{5}$$

where N is the number of scattering events in each path p, θ_i is the scattering angle corresponding to each scattering event, Δr_i is the displacement of scattering particles in microvessel.

Let us suppose that all the scatterers (for example, blood form elements, albumin of blood plasma, etc.) are uniformly distributed at random over a cross section of Poiseuille's flow. Then, in the following case:

$$\Delta \boldsymbol{r}_i(t) = \boldsymbol{v}_0 \left[1 - \left\{ \mathrm{rnd}\left(\frac{r_i}{r_0} \right) \right\}^2 \right] t, \tag{6}$$

where v_0 is centerline velocity of the flow, r_i is the distance from the particle to the center of the flow, r_0 is the radius of microvessel, and $\mathrm{rnd}(r_i/r_0)$ is the random value, uniformly distributed within the range $[0;1]$.

To make further progress we need to calculate $\langle I_p \rangle / \langle I \rangle$ and $\langle \exp(i\Delta\Psi_p) \rangle$ in Eq. (4). In traditional *DWS* theory two key approximations are accepted: total number of scatterers is large and tends to infinity; $\Delta\Psi_p$ is a random value, which obeys the Gaussian statistics.

Regretfully, these postulates cannot be used in the case of a small number of scattering events. Application of other theoretical approaches (such as the Feynman diagram technique [19]) also cannot provide the analytical expression in evident form. The general purpose of this paper is to rearrange Eq. (4) for the case of small number of scattering events.

III. CALCULATION OF $\langle I_p \rangle / \langle I \rangle$

In Ref. [17] it is demonstrated that $\langle I_p \rangle / \langle I \rangle$ may be interpreted as the distribution of a fraction of scattered intensities in diffraction paths of length s. Let us calculate path-length distribution function for photons, propagating in a nonabsorbing medium.

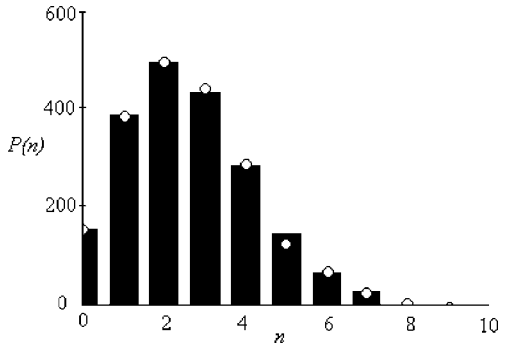

FIG. 2. Histogram of the number of scattering events. Bars: histogram of photons; circles: Poissonian distribution.

If each photon is scattered only one time, the distribution of path lengths between elementary scattering events is expressed by the formula [20]

$$p(s|1) = \mu_s \exp(-\mu_s s), \tag{7}$$

where μ_s is the scattering coefficient.

If a photon has been involved in two independent scattering events, the probability density function of the sum of two random paths is given by

$$p(s|2) = p(s|1) \otimes p(s|1) = \mu_s^2 s \exp(-\mu_s s), \tag{8}$$

where $\otimes$ is the convolution symbol.

For three scattering events, the probability density function of the sum of three paths equals to

$$p(s|3) = p(s|2) \otimes p(s|1) = \mu_s^3 \frac{s^2}{2} \exp(-\mu_s s). \tag{9}$$

Evidently, for n scattering events, the recurrent formula may be derived

$$p(s|n) = \mu_s^n \frac{s^{(n-1)}}{(n-1)!} \exp(-\mu_s s), \tag{10}$$

where $p(s|n)$ is the conditional probability density function that corresponds to the case that a photon passes length s and participates in n scattering acts. Clearly, $s > 0$ in Eqs. (7)–(10).

Finally, in accordance to Bayes's theorem, the total probability density equals

$$p(s) = p(s|n)P(n), \tag{11}$$

where $P(n)$ is discrete probability distribution of number of scattering events.

An example of the histogram, obtained with a small number of scattering events, is presented in Fig. 2. The number of scattering events obeys Poissonian distribution

$$P(n) = \frac{\langle n \rangle^n \exp\left(-\langle n \rangle\right)}{n!}, \tag{12}$$

where $\langle n \rangle$ is the average number of scattering events. Monte Carlo simulation [20] has been used to test this hypothesis. As an investigation of the statistical properties of the scattered light shows, the hypothesis about Poissonian distribution in a number of scattering events should be accepted [21]

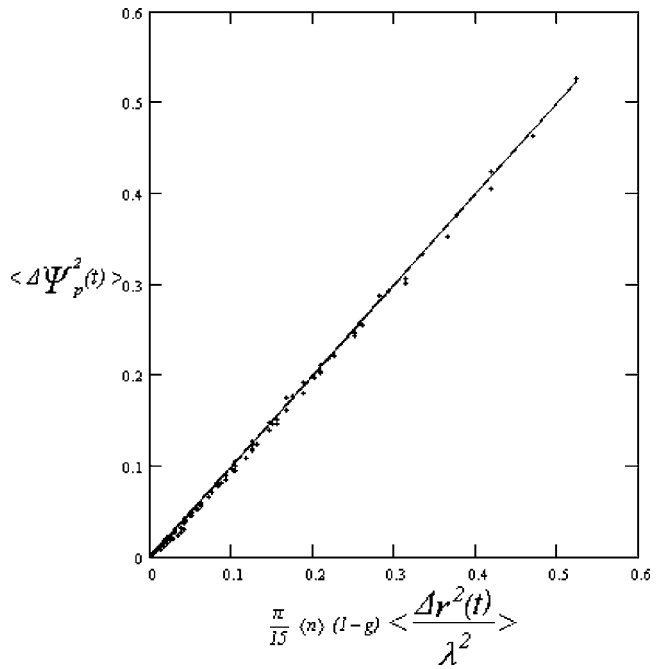

FIG. 3. Dependence of $\langle \exp[i\Delta\Psi_p] \rangle$ on the g factor and the average number $\langle n \rangle$ of scattering events.

at a significance level of $\alpha=0.05$, at least when the average number of scattering events lies in the interval $\langle n \rangle \in [1;4]$, with a waist beam radius within the interval $W_0 \in [0.5\lambda;10\lambda]$, and a factor of anisotropy of the medium $g \in [0.9;0.99]$).

So, finally the explicit formula for $\langle I_p \rangle / \langle I \rangle$ may be derived from Eqs. (10)–(12),

$$\frac{\langle I_p \rangle}{\langle I \rangle} \propto p(s) \equiv \mu_s^n \frac{s^{(n-1)}}{(n-1)!} \exp(-\mu_s s) \frac{\langle n \rangle^n \exp(-\langle n \rangle)}{n!}. \tag{13}$$

IV. CALCULATION OF $\langle \exp[i\Delta\Psi_p] \rangle$

The dependence of $\langle \exp[i\Delta\Psi_p] \rangle$ on the g factor and the average number $\langle n \rangle$ of scattering events cannot be obtained analytically. Regression analysis [22], however, based on the results of Monte Carlo simulations (for more details see [14,15]), allows us to find out the relation between the mentioned value and scattering characteristics of microflow in the explicit form

$$\langle \exp[i\Delta\Psi_p] \rangle = \exp\left[\frac{\pi}{15} \langle n \rangle (1-g) \left\langle \frac{\Delta r^2(t)}{\lambda^2} \right\rangle \right], \tag{14}$$

where Δr is described by Eq. (6), and λ is the wavelength of scattered light.

This dependence is presented in Fig. 3 (the points are the result of Monte Carlo simulation, the solid line is an approximation of data points, obtained on the base of the linear regression model [21]).

The obtained formula (14) is also valid within the range of $g \in [0.9;0.99]$ and $\langle n \rangle \in [1;4]$.

The expression for $\langle \Delta_{r^2}(t) \rangle$, after the averaging using Eq. (6), takes the following form:

$$\langle \Delta r^2(t) \rangle = \frac{v_0^2 t^2}{5}. \tag{15}$$

So, finally, for DWS with a small number of scattering events, $\langle \exp[i\Delta\Psi_p] \rangle$ equals to

$$\langle \exp[i\Delta\Psi_p] \rangle = \exp\left[\frac{\pi}{75} \langle n \rangle (1-g) \frac{v_0^2 t^2}{\lambda^2} \right]. \tag{16}$$

V. SECOND-ORDER STATISTICS OF SCATTERED LIGHT

Using Eqs. (4), (13), (16), the autocorrelation function of the fluctuations of complex amplitude of scattered light may be expressed as

$$g_1(t) = \int_0^\infty \sum_n \mu^n \frac{s^{(n-1)}}{(n-1)!} \exp(-\mu_s s) \cdot \frac{\langle n \rangle^n \exp(-\langle n \rangle)}{n!} \times \exp\left\{ -\frac{\pi}{75} \langle n \rangle (1-g) \left[\frac{v_0 t}{\lambda} \right]^2 \right\} ds. \tag{17}$$

It should be mentioned that [23]

$$\sum_n \frac{\langle n \rangle^n}{n!} = \exp(\langle n \rangle) \tag{18a}$$

and

$$\int_0^\infty s^{(n-1)} \exp(-\mu_s s) ds = \frac{(n-1)!}{\mu_s^n}. \tag{18b}$$

After some algebra we obtain the final expression for the correlation function of complex field fluctuations

$$g_1(t) \propto \exp\left\{ -\frac{\pi}{75} \langle n \rangle (1-g) \left[\frac{v_0 t}{\lambda} \right]^2 \right\}, \tag{19}$$

where v_0, again, is center line velocity of the microflow.

In Ref. [2] it has been indicated that the correlation function g_2 of scattered intensity fluctuation is given by the Siegert relation

$$g_2 \propto |g_1|^2. \tag{20}$$

Then, we can recast Eq. (19) and thus Eq. (20) becomes

$$g_2(t) \propto \exp\left\{ -\frac{2\pi}{75} \langle n \rangle (1-g) \left[\frac{v_0 t}{\lambda} \right]^2 \right\}. \tag{21}$$

Clearly, the correlation function of the intensity fluctuations of scattered light has a Gaussian shape.

The typical experimental arrangement for microflow measurements [24] in a transmitting geometry is shown in Fig. 4. A conventional optical microscope fitted with a charge-coupled device (CCD) camera (elements 8, 7, 4, and 10 in Fig. 4) enables visual observation of the blood flow in a microvessel (an isolated capillary of white rat mesentery). A laser beam ($\lambda=630$ nm) is focused on a spot of a small ra-

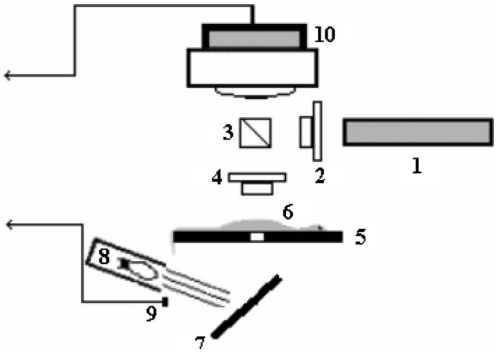

FIG. 4. Optical scheme. 1: laser; 2, 4: microobjectives with $10^{\times}$ magnification, 3: beam splitter; 5: stage; 6: microflow (blood capillary of mesentery of white rat); 7: mirror; 8: lamp; 9: photoreceiver with a pinhole; 10: CCD camera, supplied by focusing optics.

dius (W_0=1.5 μm) in the investigated microflow. As blood flows through the microvessel, the strongly focused laser beam is modulated in the waist plane. This leads to the formation of the dynamic speckles. The temporal fluctuations of the scattered intensity are detected by the photoreceiver.

Experimental correlation function of intensity fluctuations of light, scattered from 100-μm blood flow is presented in Fig. 5. Evidently, experimental data is in good agreement with the theoretical prediction (i.e., the forms of comparing correlation functions are practically the same).

VI. CONCLUSIONS

The decay time ΔT of the correlation function of the fluctuations of scattered intensity can be derived from Eq. (21)

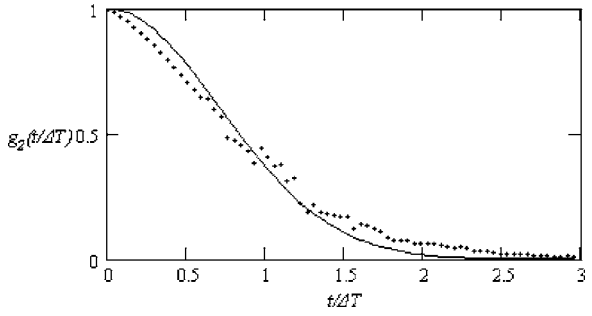

FIG. 5. Correlation function of intensity fluctuations of light, scattered from 100-μm blood flow. The solid line is a theoretical result (based on Eq. (21)); the dots are the experimental data.

$$\Delta T = \frac{\lambda}{v_0 \sqrt{\frac{2\pi}{75}\langle n\rangle (1-g)}}. \tag{22}$$

Formula (22) is the main theoretical result of this Rapid Communication. Evidently, ΔT depends not only on velocity v_0 of the flow, but on its scattering characteristics as well. Equation (22) allows us to measure the center line velocity of the highly scattering microflow in the case when the g factor and the average number of scattering events are estimated anticipatorily. Usually, these parameters are well known, at least for blood flows in microvessels.

ACKNOWLEDGMENTS

This research has been supported by Russia Foundation of Basic Researches through Grants No. N04-04-48279 and N03-04-39021, a grant of the President of the Russian Federation (Grant No. MD 358.2003.04), Grant No. E02-6.0-159 of Ministry of Education of Russia and by the US Civil Research and Development Foundation (Award No. REC~006).

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