Dynamic light scattering in localized coherence volumes

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We introduce a novel light-scattering technique for investigating the dynamics of random media with a broad range of optical densities. By use of the spatial coherence properties of a single-mode optical fiber and the temporal coherence of a broadband source, the measurement volume is isolated at the end of the optical waveguide. Optical mixing between the fluctuating scattered light and the Fresnel-reflected field at the fiber–medium interface is analyzed directly in the frequency domain. The unique characteristics of this new technique are discussed in the context of simultaneous measurement of average scatterer size and concentration in dense colloidal suspensions.

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Dynamic light scattering (DLS) has been established as a powerful technique for investigating dynamic processes.[1] Originally, DLS applications were limited to weakly scattering media, in which light propagation can be described by single-scattering models. An important breakthrough in the field of DLS is represented by the extension of DLS to strongly scattering media. This technique is referred to as diffusing-wave spectroscopy and has been used to study particle motion in concentrated colloidal suspensions, microemulsions, and other systems that are characterized by significant multiple scattering.[2,3] In analyzing a temporal autocorrelation, diffusing-wave spectroscopy relies on accurate knowledge of the optical path-length distribution $P(s)$ of multiply scattered waves. This most important descriptor of the scattering process has been commonly inferred on the basis of various photon-diffusion models, and it is only recently that its experimental accessibility has been proved.[4,5]

Because of their flexibility and their extended applicability, modern experimental geometries of DLS involve single-mode optical fibers.[6] Detecting light with a single-mode optical fiber allows the collection of the fluctuating signal from a spatially coherent area, determined by the properties of the optical waveguide. However, the temporal coherence of the incident light is known to influence the properties of the dynamic signals in the context of multiple light scattering.[7] By use of partially coherent light in an interferometric geometry, Brownian motion of colloidal particles in highly scattering media has been investigated.[8] It is to be expected that tuning the coherence properties of incident radiation could add a new dimension to the scattering process and, therefore, lead to novel investigation techniques for situations that cannot be tackled with conventional approaches.

One of the yet to be solved problems is scattering from inhomogeneous media that are neither single scattering nor highly diffusive. In dense colloidal systems, for instance, the quantity that defines the scattering regime is the optical density, defined as the ratio between the geometrical dimension of the sample and the characteristic scattering length, i.e., the mean free path of the medium. For a range of practical applications, the scattering regime falls in the transition domain between a low optical density regime, where DLS has been successfully applied for many years, and the regime of high optical density, where diffusing-wave spectroscopy approach can give an accurate description. So far, there have only been a few attempts to fill this gap. DLS of coherent light in this regime has been described by use of a combination of cumulants weighted by empirical constants.[9] It is of considerable interest to be able to optically probe the scattering systems that lie in this intermediate regime of optical densities in a consistent manner.

In this Letter we explore the potential of dynamic scattering with low-coherence light for investigation of the local dynamic properties of media over a broad range of optical densities. The new technique is fiber-optics based and permits collection of backscattered light from a small volume localized at the end of the optical waveguide. A pigtailed superluminescent diode with a central wavelength of 824 nm and a coherence length of 30 μm is coupled into a single-mode optical fiber, which represents one arm of a 1×2 fiber coupler. The output of the coupler is immersed in the colloidal suspension under investigation, and the backscattered light is collected through the same fiber. Without additional optical components, the signal is detected and further analyzed in the frequency domain by a spectrum analyzer. Owing to the refractive-index contrast between the fiber core and the suspension solvent, the signal that is detected has two components: the light that is backscattered from the dynamic system and the component that is due to the Fresnel reflection at the fiber–medium interface, as described in Fig. 1. The coherence length and the transversal dimension of the fiber core define a coherence volume in which the optical fields preserve relative phase correlations. Considering the terms that survive the time averaging, we were able to derive the expression for the intensity autocorrelation function as

$$G^{(2)}(\tau) = {I_0}^2 + 2I_0I_s + {I_s}^2 + I_0\sum_j I_j[g_j(\tau) + g_j(\tau)^*] \times \exp[-2(s_j - s_0)^2/{l_c}^2] + \sum_{k\neq l} I_k I_l g_k(\tau) g_l(\tau)^* \times \exp[-2(s_k - s_l)^2/{l_c}^2]. \quad (1)$$

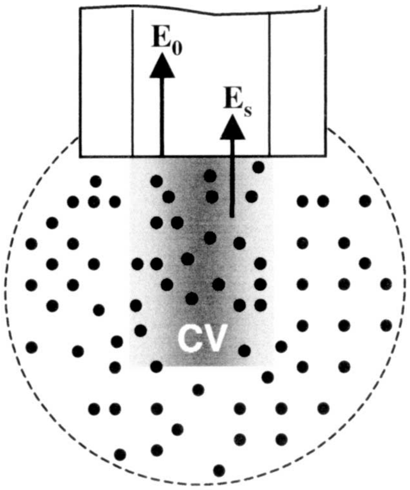

Fig. 1. Coherence volume (CV) determined by the single-mode fiber and the properties of the broadband radiation.

In Eq. (1), I_0 and $I_s = \sum_j I_j$ are the average intensities of the specular and the scattered components, respectively, and I_j is the intensity associated with the backscattered component of trajectory j; g_j represents the normalized first-order correlation function corresponding to this component, defined as $g_j(\tau) = \langle E_j(\tau)E_j(t + \tau)^* \rangle / I_j$, with the angle brackets denoting time averaging and the symbol $*$ representing complex conjugation. The optical path lengths associated with the scattered component j and the specular field are denoted s_j and s_0, respectively, and l_c is the coherence length. It is worth mentioning that, for the limit of $I_0 = 0$, Eq. (1) is consistent with the result derived in Ref. 7 for a self-beating geometry. An important consequence of Eq. (1) is that for scattering media with mean free paths longer than $l_c/2$ the backscattered light undergoes on average only one scattering event in the coherence volume, which is defined approximately by the coherence length and the area of the fiber core. Consequently, the autocorrelation functions g_j are independent of the length of the scattering trajectory and are given by the well-established formula for quasi-elastic light scattering. If $I_s \ll I_0$, which is the case in all our experiments, the last (self-beating) term of Eq. (1) becomes negligible. With these assumptions, Eq. (1) can be arranged to give the normalized autocorrelation function

$$g^{(2)}(\tau) = 1 + 2\,\frac{I_0 {I_s}^{\mathrm{CV}}}{(I_0 + {I_s}^{\mathrm{CV}})^2}\, g^{(1)}_{(\tau)}\,. \tag{2}$$

In Eq. (2), $g^{(1)}(\tau) = \exp(-q^2 D\tau)$, where D is the particle diffusion coefficient and q is the scattering vector, which, for our backscattering geometry, equals twice the wave number $(4\pi/\lambda)$. For Brownian particles of diameter d, the diffusion coefficient relates to the temperature T and the viscosity η of the medium through the well-known Stokes–Einstein expression $D = k_B T / 3\pi\eta d$, where k_B is Boltzmann's constant. The quantity ${I_s}^{\mathrm{CV}}$ in Eq. (2) represents the average intensity of the light scattered from the coherence volume, ${I_s}^{\mathrm{CV}} = \sum_j I_j \exp[-2(s_j - s_0)^2/{l_c}^2]$. For all the real light sources, the coherence length has a finite value, and therefore the inequality ${I_s}^{\mathrm{CV}} < I_s$ always applies.

In our experiments the coherence length is $\sim$30 μm, which means that the investigated media can be optically dense but can still be analyzed by a single-scattering model, as described in Eq. (2). The fluctuations of the scattered light have been analyzed in the frequency domain, based on the Fourier-transform relationship between the intensity autocorrelation function $G^{(2)}(\tau)$ and the power spectrum $P(\omega)$. The associated power spectrum has a Lorentzian shape,

$$P(\omega) = \frac{A_0}{\Omega}\,\frac{1}{1 + (\omega/\Omega)^2}\,, \tag{3}$$

where $\Omega = Dq^2$ and A_0 is the spectrum amplitude proportional to the product $I_0 {I_s}^{\mathrm{CV}}$. Thus the amplitude A_0 of the power spectrum can be expressed in simple form as $A_0 = \alpha\rho Q_b/d$, where d is the diameter of the particle, Q_b is the backscattering efficiency, ρ is the density of particles by volume, and α is an experimental constant. Since the dimension of the particle can be determined from the width of the power spectrum, as described above, the backscattering efficiency Q_b can be calculated for particles with known optical properties. Consequently, the particle concentration is obtained by measurement of the amplitude A_0 and calibration for the experimental constant α.

We accurately determined both the linewidth Ω and the amplitude A_0 of the measured power spectra by fitting the data with Eq. (3). Figure 2(a) shows the linewidth Ω of the power spectrum for polystyrene microsphere aqueous suspensions with various volume fractions (ρ) and three particle dimensions, as indicated. Remarkably, for the whole concentration interval, the results are within 4% of error with respect to the values calculated with Mie theory, which are represented by the solid lines. The independence of the power-spectrum linewidth from the volume fraction undoubtedly proves that the single-scattering description given by Eq. (2) is correct and that our technique can be used for dynamic systems over a broad range of concentrations. Therefore, performing DLS in localized coherence volumes should have important applications in subdiffusive or inhomogeneous scattering systems, which cannot be characterized by DLS with coherent light. Owing to the inherent high dynamic range of heterodyne detection operated efficiently in single-mode optical fibers, and with effective isolation of the measurement volume, our technique allows for simultaneous determination of particle concentration.

Figure 2(b) shows the measured values of the power-spectrum amplitude A_0 for all the samples, and it can be seen that A_0 depends linearly on the volume fraction ρ for all the particle dimensions. The slope of the linear behavior is dependent on the particle diameter, as shown in the logarithmic plot of Fig. 2(b). The curves have been further normalized by Q_b/d for each particle dimension and, as a result, all the experimental dependences have been collapsed into a single curve, as indicated by the inset of Fig. 2(b). This remarkable result permits us to find the experimental constant α, which, in turn, gives access

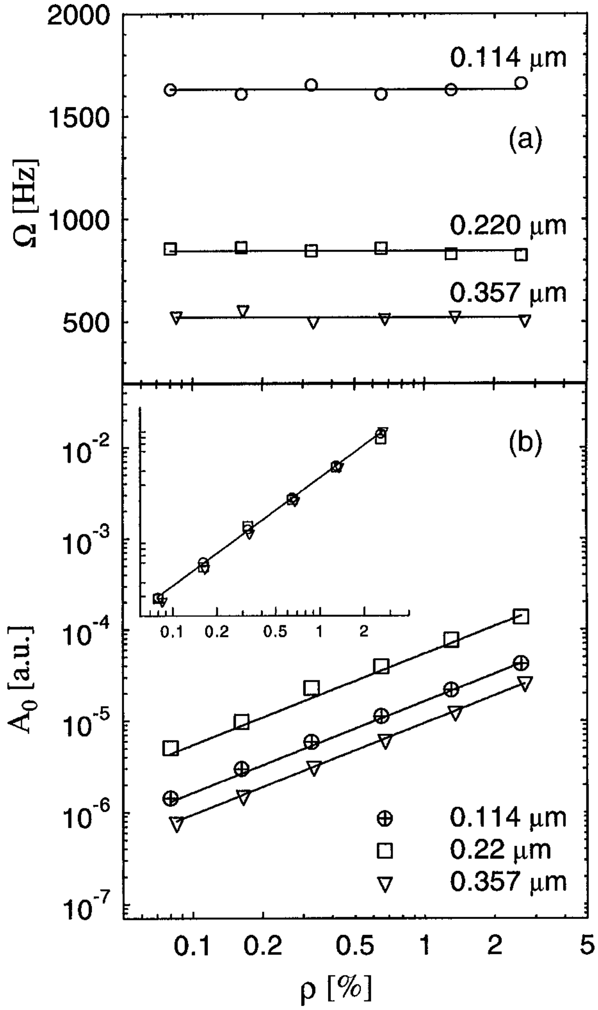

Fig. 2. Power-spectrum (a) linewidth and (b) amplitude as a function of volume fraction for colloidal suspensions of different particle sizes, as indicated. The inset of (b) shows curves $A_0(\rho)$ collapsing into a unique dependence after normalization with Q_b/d.

to information regarding the particle concentration. Alternatively, for a medium with known density, the additional information contained in the spectrum amplitude offers backscattering efficiency Q_b, which can be further related to the optical properties of the particles.

In conclusion, we have introduced a novel low-coherence dynamic light-scattering technique with which to investigate localized dynamics. Using the same single-mode optical fiber to launch and detect light with a finite coherence length, we effectively generate a measurement volume at the end of the optical waveguide. Taking advantage of this localized coherence volume, we find that the technique isolates the single-scattering contributions from the medium. The high dynamic range of the heterodyne detection permits accurate measurement of both the shape and the amplitude of the power spectrum and, therefore, offers the possibility of determining simultaneously the size and the concentration of the scattering centers. The flexibility of this fiber-optic-based technique, together with the possibility of adjusting the coherence properties of the probing radiation, should be appealing for a range of applications involving the detection of localized dynamics in highly scattering media.

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