# Influence of finite coherence length of incoming light on enhanced backscattering

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We have investigated the effects of the finite coherence length of the incoming light on multiple scattering in a random medium. Large optical fluctuations observed with coherent light gradually decrease their intensity as the coherence length of the incoming light is shortened. When the coherence length is shortened below the elastic mean free path, the enhanced backscattering peak is revealed without ensemble average of the fluctuations. This variation of the fluctuations as a function of the coherence length of the incoming light has been analyzed on the basis of the diffusion approximation.

Propagation of coherent waves in disordered media has been recently the focus of intense interest. An essential feature for this transport phenomenon is phase-coherent multiple scattering. Peculiar interference effects due to multiple scattering of waves appear in a variety of situations. For example, electronic transport through a structure smaller than the inelastic scattering length yields large fluctuations in conductance as a function of the applied magnetic field.[1] Propagation of coherent light in a rigid disordered medium manifests large-amplitude fluctuations in the scattering intensity as a function of angle.[2–6] An ensemble average of the fluctuations decreases their intensity and reveals a sharp peak in the backscattering direction which results from constructive interference of the time-reversed pair of scattering sequences.[7–11]

Optical fluctuations arise from interference effects due to multiple scattering of the light whose trajectory is shorter than the coherence length of the incoming light. Therefore, by changing the coherence length of the incoming light, we will be able to study the contributions of light trajectories of a specific path length to the backscattering interference pattern. Furthermore, if the coherence length is shorter than the elastic mean free path, the transport phenomena will be described using the classical diffusion theory in which any interference effects are neglected. As a result, the backscattering peak is expected to be directly observed without ensemble averaging because the constructive interference between the time-reversed scattering processes is still preserved. In this context, we have investigated the effect of the finite coherence length of the incoming light on multiple scattering in a random medium. We have performed a backscattering experiment using laser pulses and have observed the variation of the optical fluctuations as a function of the coherence length. The coherence length of the incoming laser pulses was varied from 9 to 2100 $\mu$m, which range corresponds to the typical lengths of the light trajectories in the sample. The observed experimental results are analyzed on the basis of the diffusion approximation.

Our experimental setup is shown in Fig. 1. An important component in our experiment is a nitrogen-laser-pumped Hansch-type dye laser which provides optical pulses of 5 ns duration at 550 nm with arbitrary coherence length. The cavity of the dye laser is about 20 cm long and has a 600 mm$^{-1}$ echelett-type grating as the end reflector. We control the coherence length of the dye laser from 9 to 2100 $\mu$m by changing the diffraction order of the end reflector. The longest coherence length in our experiment is obtained with a beam expander inside the dye laser cavity. By virtue of the Wiener-Khinchin's theorem, the coherence length is calculated from the observed spectral linewidth of the dye laser. An aqueous suspension of 10% solid-fraction polystyrene spheres of 0.46 $\mu$m diameter from Dow Chemical is used as a sample. The collimated beam from the dye laser is reflected onto the sample from a beam splitter. The backscattered light polarized parallel to the polarization of the incoming light is detected with a change-coupled-device (CCD) camera followed by a two-dimensional image processor. In Fig. 2(a) an example of the observed scattering patterns is shown, where the coherence length of the laser pulse is the longest one used (2100 $\mu$m). The Brownian

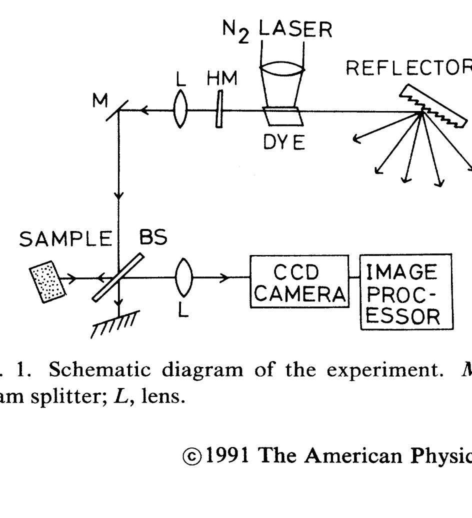


FIG. 1. Schematic diagram of the experiment. *M*, mirror; *BS*, beam splitter; *L*, lens.

 

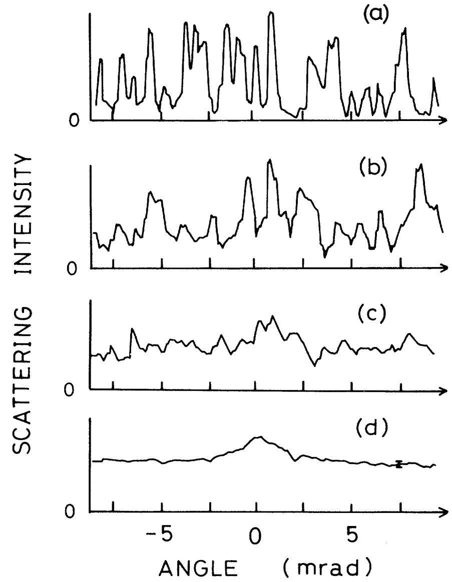


FIG. 2. Examples of the backscattering patterns from the aqueous suspension of the polystyrene spheres as a function of the coherence length of the incoming laser pulses. The coherence length is (a) 2100 $\mu$m, (b) 950 $\mu$m, (c) 320 $\mu$m, and (d) 18 $\mu$m, respectively. The error bar in (d) represents the noise level of the detection system.

motion of polystyrene spheres is not important in this case because the pulse duration (5 ns) of the incoming beam is much shorter than the time scale of the Brownian motion. Thus, the scattering pattern shows a fine speckle structure in our liquid sample, as in rigid disordered solid samples.[2,3] This speckle pattern, however, changes its profile for every laser shot, because the scattering particles are rearranged by the Brownian motion during the repetition period of the dye laser (0.25 s). In Figs. 2(b)–(d), examples of the scattering patterns for various coherence lengths are shown. When the coherence length is shortened below the typical length of light trajectories in the sample, the fluctuations are reduced, because the incoming light cannot coherently sample the entire volume. It should be noted that the coherent backscattering peak appears without any ensemble-averaging process when the coherence length is shortened below $\sim 18$ $\mu$m [Fig. 2(d)].

Let us consider the quantitative behavior of the variation of the speckle patterns. We observed backscattering

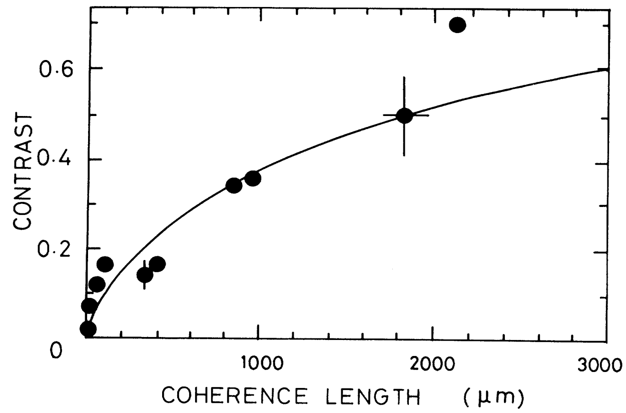


FIG. 3. Contrasts of the backscattering speckle patterns as a function of the coherence length of the incoming light pulses. The solid circles with error bars are experimental results and the solid line represents a result of calculation based on Eq. (4).

patterns like Fig. 2 slightly away from the backscattering peak ($\sim 4$ mrad) in order to avoid the effect of the peak. In Fig. 3 the solid circles represent the contrasts calculated from the observed patterns as a function of the coherence length of the laser pulse, where the contrast of the speckle pattern is defined by the square of the normalized standard deviation of the speckle intensity. As the coherence length of the laser pulse is shortened, the contrast decreases slowly in the coherence length $>200$ $\mu$m and rapidly below 200 $\mu$m. The value of the contrast for the observed patterns is smaller than unity, even in the longest coherence length of 2100 $\mu$m, which is about 100 times as long as the elastic mean free path.[12] This suggests that there exist much longer light trajectories in the sample.

The experimental results are analyzed on the basis of the diffusion approximation. The reflected (cycle-averaged) intensity of a speckle pattern is expressed as a sum of many phasor contributions from all trajectories $\xi$,

$$I(\theta)=\int_{-\infty}^{\infty} d\omega S(\omega)\sum_{\xi_1}\sum_{\xi_2}\varepsilon_{\xi_1\omega}(\theta)\varepsilon^*_{\xi_2\omega}(\theta)\ , \tag{1}$$

where $S(\omega)$ is the power spectrum of the incoming light and $\varepsilon$ is the scattered field at an observation angle $\theta$ that is expected from incident light of unit intensity. The second-order intensity correlation function $G_2$ of the speckle pattern is written as

$$\begin{aligned} G_2(\theta,\Delta\theta) &= \langle I(\theta)I(\theta+\Delta\theta)\rangle \\ &= \left\{\int_{-\infty}^{\infty} d\omega S(\omega)\left[\sum_{\xi}\left|\varepsilon_{\xi\omega}(\theta)\right|^2\right]\right\}^2 \\ &\quad + \int_{-\infty}^{\infty} d\omega_1 S(\omega_1)\int_{-\infty}^{\infty} d\omega_2 S(\omega_2)\left\langle\sum_{\xi_1}\varepsilon_{\xi_1\omega_1}(\theta)\varepsilon^*_{\xi_1\omega_2}(\theta+\Delta\theta)\right\rangle\left\langle\sum_{\xi_2}\varepsilon^*_{\xi_2\omega_1}(\theta)\varepsilon_{\xi_2\omega_2}(\theta+\Delta\theta)\right\rangle\ , \end{aligned} \tag{2}$$

where $\langle\cdots\rangle$ means an ensemble average over many scattering patterns. We assumed that the scattering process is a stationary Gaussian random process, and thus divided the fourth-order moment of the field into the sum of the two second-order moments. This assumption is valid, provided that the typical length of the trajectories is much longer than the wavelength. In our case, even the shortest path length is more than ten times longer than the wavelength. The first term of Eq. (2) is the square of the averaged intensity, and the second term of Eq. (2) is the square of the standard deviation.

The phase deviation among the scattering fields in the angular brackets in Eq. (2) is written as[13,14]

$$\phi = -i[c^{-1}(\omega_1-\omega_2)(l_1-l_2) + (\Delta k_{\text{in}}\,\Delta\xi_{\text{in}} - \Delta k_{\text{out}}\,\Delta\xi_{\text{out}})] , \tag{3}$$

where $l_1$ and $l_2$ are the lengths of light trajectory 1 and 2, respectively, and $c$ is the velocity of light. The vector $\Delta k_{\text{in}}$ is defined as the difference between two wave vectors of the incoming light of frequency $\omega_1$ and $\omega_2$, and $\Delta k_{\text{out}}$ is that of outgoing light. $\Delta\xi_{\text{in}}$ in the scattering plane represents the vector between the first scattering centers of the light trajectory 1 and 2, and $\Delta\xi_{\text{out}}$ is the vector between the last scattering centers. The first term in Eq. (3) represents the phase deviation caused by the difference in the path length of trajectories, and the second term is the linear phase shift in the scattering plane. In our experimental conditions, the incoming beam can be assumed to be normal to the scattering plane and the scattering pattern is observed in the backscattering direction. Thus the linear phase term can be neglected. We replace the ensemble average $\langle \cdots \rangle$ in Eq. (2) by integrals over distributed trajectory lengths; then the speckle contrast is

$$C = \frac{\int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 S(\omega_1)S(\omega_2) \int_{l^*}^{\infty} dl_1 \int_{l^*}^{\infty} dl_2\, g(l_1)g(l_2)\exp[-ic^{-1}\omega_1(l_1-l_2)+ic^{-1}\omega_2(l_1-l_2)]}{\left\{\int_{-\infty}^{\infty} d\omega S(\omega)\right\}^2}$$

$$= \int_{l^*}^{\infty} dl_1 \int_0^{\infty} dL\, |G_1(L/c)|^2 g(l_1)g(l_1+L) , \tag{4}$$

where $L = l_2 - l_1$, $g(l)$ is the normalized probability of the trajectory, $G_1$ is the first-order correlation function of the incoming light, and $l^*$ is the elastic mean free path. In Eq. (4) the Wiener-Khinchin's theorem has been used. We calculated $g(l)$ in Eq. (4) on the basis of the diffusion approximation.[15-17] A calculated result of Eq. (4) is shown in Fig. 3 with the solid curve. The only parameter used here is the elastic mean free path, which was estimated as 38 $\mu$m from the observed width of the coherent backscattering peak and from Eq. (4) in Ref. 16. As shown in Fig. 3, the theory agrees well with the observations. The diffusion approximation will not be valid for short trajectories.[15] The calculated curve, however, shows a good agreement with the experimental data. This possibly originates from using the effective elastic mean free path estimated from the observed width of the backscattering peak.

There is a wide range of the path lengths of the light trajectories in the sample. Inside the backscattering peak, the longer loops contribute to the smaller-angle components of the peak.[18] The optical fluctuations, therefore, will first decrease in the central region of the peak when the coherence length of the incoming light is shortened. Ultrafast time-resolved spectroscopy with femtosecond optical pulses can also be used to study the distribution of light trajectories. Vreeker *et al.*[19] have reported the time evolution of the shape of the enhanced backscattering peak with a temporal resolution of 30 fs. The shortest coherence time in our experiment is as short as 40 fs. Since temporally incoherent light interferes only within its coherence time, our results directly show that the observed backscattering peak is constructed by the interference between the two time-reversed scattering processes within 40 fs. In this sense, our method is one of ultrafast spectroscopy with temporally incoherent light.[20-22] Our method is based on a linear optical phenomenon, while spectroscopy with incoherent light, so far reported, is based on nonlinear optical phenomena, such as degenerate four-wave mixing.

We can classify light sources of short coherence length into two categories. One is an incoherent, or phase-modulated, light source, which has a long pulse duration compared with its coherence time. The other is a coherent short pulse, which has a Fourier-transform-limited pulse duration with respect to its spectral width. The light source used in our present experiment is classified in the former category, while our analysis of the experimental results is applicable to both of the experiments with the above two types of light sources. The time-resolved behavior of the scattered light is, however, very different in these two cases. If we use coherent short pulses whose duration is shorter than the time for light to pass through the typical path length, the scattered light is expected to show large temporal intensity fluctuations of the order of the incoming pulse duration because of the random interferences of many light trajectories in the time domain. These new types of temporal fluctuations,[23] which are essentially different from the ones originating from the Brownian motion of the scatterers,[15,17,24-26] reduce the contrast of the speckle pattern as is the case with the present experiment using an incoherent light source. This unique behavior of the propagation of coherent pulses through a random medium is discussed elsewhere.[23]

In summary, we have experimentally found that the coherent backscattering peak appears without any ensemble-averaging process when the coherence length of the incoming light becomes shorter than the elastic mean free path. We have also presented speckle contrasts calculated from the backscattering interference patterns as a function of the coherence length of the incoming light, and have provided a simple diffusion theory which is in good agreement with the data.

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