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Temporal fluctuations in disordered static optical media

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We report an observation of large picosecond optical fluctuations of transmitted light when a coherent picosecond pulse propagates through a strongly disordered solid. These temporal fluctuations are a direct result of random interference among multitudinously scattered waves from different trajectories in the time domain. They appear without any dynamic motions of the medium, such as the Brownian motion of scatterers. A spatial average of the fluctuations yields a temporally smooth transmitted profile, while a time average of the fluctuations yields a spatially smooth scattering profile.

Propagation and scattering of a coherent wave in random media show inherently noiselike behavior due to random interference among multitudinously scattered waves. In static disordered systems, this interference forms a sample-specific random scattering pattern—"a fingerprint"—which reflects a sample-specific arrangement of the disordered structure. The optical speckle in disordered optical systems[1-3] and the universal conductance fluctuations as a function of applied magnetic field in mesoscopic electronic systems[4] are examples of this noiselike structure.

The purpose of this paper is to consider the above interference of an optical pulse in the time domain and to report large temporal fluctuations (on the order of an incident-picosecond-pulse duration) in the transmitted pusle profiles. These temporal fluctuations are essentially different from those originating in the thermal motion of scatterers, which are often discussed in quasielastic light scattering (QELS) or diffusing-wave spectroscopy (DWS).[5-12] They appear even in rigid solid samples where the Brownian motion of the scatterers is absent. The profile of the temporal fluctuations is sample specific and reproducible. Thus we can recognize it as a fingerprint in the time domain for the specific arrangement of the disordered structure.

The simplest way to understand the above type of fluctuations is to consider "transient speckle patterns" by an incident coherent optical pulse. Consider an experimental condition where the incident pulse propagates through a disordered medium, in which the lengths of the photon trajectories are widely distributed beyond the incident pulse length. Light pulses come out from all possible trajectories with the optical phases inherent to the trajectories. Thus a certain set of photons from shorter trajectories forms a certain spatial speckle pattern first on the observation plane, which stays for a moment of the order of the incident pulse duration. After this pattern goes out, another set of photons from longer trajectories forms a new pattern which has no correlation to the previous one. In this way transmitted light of a short pulse has large fluctuations in time with a correlation time of the order of the incident pulse duration.

A schematic diagram of the experiment for the observation of the above temporal fluctuations is shown in Fig. 1. We used the second harmonic of a cw mode-locked Nd^{3+}:YAG laser which provides controlled optical pulses of 65, 110, and 150-psec duration at 82-MHz repetition rate as the light source. Corpuscles of $BaSO_4$ (Kodak White Reflectance Standard) compacted to a 680-μm thickness were used as the disordered solid samples. A beam from the YAG laser was focused on the incident plane of the sample to a $\sim$100-μm spot, and the transmitted light from the sample was led to a synchroscanning streak camera. The main difference between our present experiment and previously reported time-resolved pulse transmission experiments[13-16] is that we used two apertures of 0.5 mm in diameter in order to detect a single coherent cell of the transient speckle. The time resolution of the detection system was about 20 psec. Examples of the temporal profile of the transmitted 65-psec pulse are shown in Fig. 2, where the polarization of the detected light is parallel [Fig. 2(a)] and perpendicular [Fig. 2(b)] to that of the incident light. We see that the transmitted pulse profiles through a strongly disor-

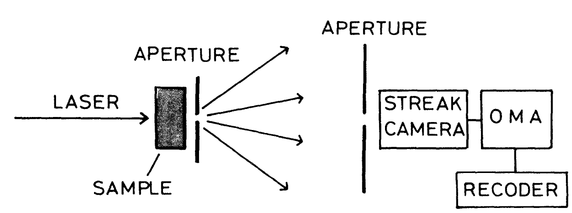

FIG. 1. Schematic diagram of the experiment. OMA is an optical multichannel analyzer.

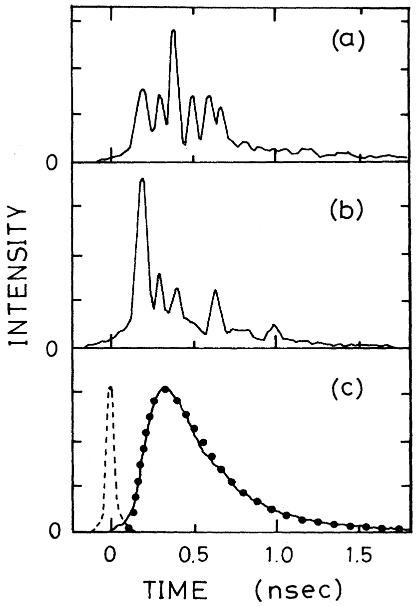

FIG. 2. Examples of the temporal profile of the transmitted 65-psec pulse, where the polarization of the detected light is (a) parallel and (b) perpendicular to the polarization of the incident pulse, respectively. (c) The solid line is the temporal profile observed with a corrective lens between the two apertures; the solid circles, a calculated curve with the photon diffusion approximation; the dashed line, the instrument response function to the 65-psec input pulse.

dered medium have inherently large temporal fluctuations of the order of the incident pulse duration. These profiles are reproducible for every laser shot as long as the geometrical configuration of the experiment is kept unchanged, while a slight translation of the sample by an order of the beam spot size, or detection of different coherent cells, changes completely the temporal profiles. Thus we can recognize it as a fingerprint in the time domain for a specific sample. The temporal profiles of Figs. 2(a) and 2(b) have no correlation with respect to each other, because the different polarizations yield different combinations of trajectories owing to the polarization-dependent cross sections of the Rayleigh scattering.

We calculated the second-order intensity correlation function

$$G(\tau)=\frac{\sum_{k=1}^{N}\int_{-\infty}^{\infty}dt[I_k(t)I_k(t+\tau)-\langle I(t)\rangle\langle I(t+\tau)\rangle]}{N\int_{-\infty}^{\infty}dt\langle I(t)\rangle\langle I(t+\tau)\rangle}\,, \tag{1}$$

where

$$\langle I(t)\rangle=N^{-1}\sum_{k=1}^{N}I_k(t)\,,$$

for a quantitative comparison between the pulse width and the correlation time of the fluctuations. In Fig. 3 the intensity correlation function of fluctuations calculated for ten sets of measured profiles [$N=10$ in Eq. (1)] including those of Figs. 2(a) and 2(b) is shown together with the intensity correlation function of the incident pulse (dashed line). The peak heights of the two curves are normalized with respect to each other. The width of the correlation peak, 92 psec is in good agreement with that of the autocorrelation function of the incident pulse. This agreement means that the spectral width of the transmitted light is same to that of the incident light, unlike the case of dynamic light-scattering experiments where the spectral width of the scattered light is broadened because of the dynamic motion of the scattering medium. The smaller value of $G(0)=0.43$ (Fig. 3) than 1.0, which is expected in stationary Gaussian randomness,[17] would probably originate in the imperfection of the spatial or temporal coherence of the incident beam.

The fact that the correlation time of the fluctuations is determined by the pulse width of the incident pulse is shown more clearly when the incident pulse width is controlled by the rf power to the mode locker of the YAG laser. In Fig. 4 examples of the temporal profile observed with optical pulses of 110-psec [Fig. 4(a)] and 150-psec [Fig. 4(b)] duration are shown. We see clearly the broadening of fluctuation peaks with the incident pulse width. If we use coherent cw light as the incident light source, the time scale of the present temporal fluctuations should become infinite, because the coherence time of the cw light is much longer than the trajectory-loop length in the random media. The use of this long coherence light corresponds to the condition of observing usual optical speckles.

Every small area of the output plane of the scattered light has a different temporal profile. Thus we can obtain an ensemble average of the fluctuations over many samples of fluctuations, if we observe sufficiently large scattering area and/or angle. For this purpose we inserted a 25-cm-focal-length, 50-mm-diam lens between the

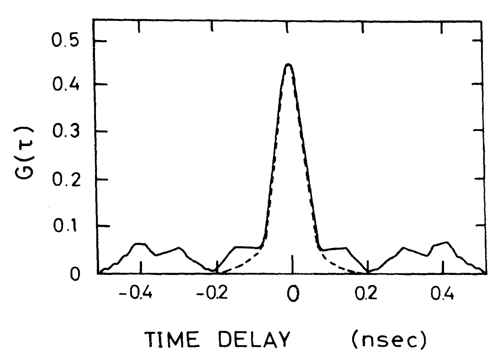

FIG. 3. The solid line is the intensity correlation function of Eq. (1). The dashed line is the autocorrelation function of the incident pulse.

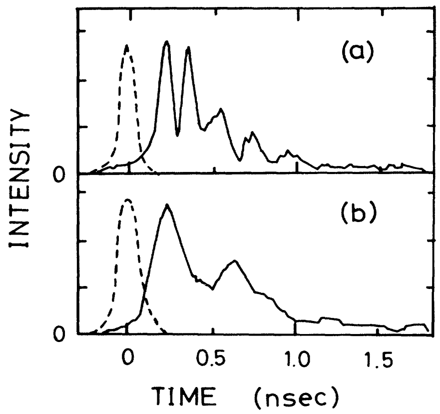

FIG. 4. Examples of the temporal profiles observed with (a) 110-psec and (b) 150-psec pulses, respectively. The dashed lines are the instrument response functions to the input pulses.

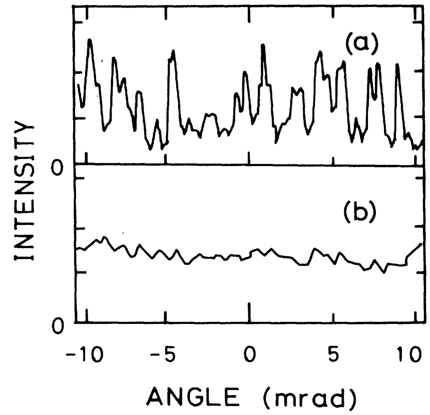

FIG. 5. (a) Time-resolved and (b) time-averaged spatial scattering patterns observed with 65-psec pulse as a function of the scattering angle, respectively.

two apertures. The result is shown in Fig. 2(c), where we obtained a smooth temporal profile of the transmitted pulse as a result of an ensemble average of more than 10^4 samples of fluctuations. The solid circles in Fig. 2(c) represent a fitted curve calculated from the diffusive transport[14,18] where the diffusion constant is $D=0.14$ mm^2/nsec and photon lifetime is $\tau_a=1.5$ nsec.

The search for photon localization in the time domain is one of the most promising means for the study of the Anderson localization, because it offers reliable values of the diffusion constant and the photon lifetime independently. Thus it has been intensively investigated in a wide range of time scales, in nanosecond,[13] picosecond,[14,15] and femtosecond regions,[16] depending on the elastic mean free path of the samples and the experimental geometric configurations. The reported temporal profiles in all previous papers are smooth profiles like Fig. 2(c). Theoretical treatments so far reported[19–21] on the fluctuations for wave propagation in random media also overlooked the present fluctuations. The fluctuations involve statistical information on multitudinously scattered waves.

A series of transient speckle patterns at various time points form a speckle in the time-space domain. For a comparison of fluctuations in time and space, we observed time-resolved transient speckle. An example of such is shown in Fig. 5(a) as a function of the scattering angle at a fixed delay time, about 0.5 nsec. Also shown is the time-integrated spatial profile in Fig. 5(b). The fine structure in the time-resolved transient speckle pattern [Fig. 5(a)] means that even though the coherence length of the incident light is much shorter than the typical system size, we can obtain a fine speckle pattern if it is time resolved. We see here a corresponding relationship between the temporal and spatial fluctuations. When the spatially resolved temporal fluctuations [Figs. 2(a) and 2(b)] are spatially averaged, the smooth temporal transmitted pulse profile [Fig. 2(c)] results. While the time-resolved spatial fluctuations [Fig. 5(a)] are time averaged, the smooth spatial scattering profile [Fig. 5(b)] results. The speckle contrast[1] of Fig. 5(b) is calculated as 0.02 when the reduction of the contrast due to the imperfection of the temporal or spatial coherence of the incident beam (Fig. 3) is compensated for. This value shows a good agreement with the theoretical value of 0.018 calculated from Eq. (4) of Ref. 22, which gives a contrast ratio for a coherent incident pulse. We, therefore, recognize that the fluctuations of the ordinary speckle with cw light make a transition to the classical diffusive transport when the coherence time of the incident light is shortened in the time-averaged measurement.[22]

We can derive another statement on the relation between the ordinary speckles and the present temporal fluctuations as follows. The spatial size of the speckle is inversely proportional to the illuminated spot size of the incident beam, while the time scale of the temporal fluctuations is inversely proportional to the illuminated spectral width of the incident pulse.

In summary, we have observed large optical fluctuations in the picosecond time domain when a coherent picosecond optical pulse propagates through a strongly disordered solid and the transmitted light is observed with spatial resolution. On the other hand, in the spatial domain, we have also shown that, even though the coherence length of the incident light is much shorter than the typical system size, we can obtain a fine-speckle pattern if the pattern is suitably time resolved. When these temporal and spatial fluctuations are spatially and temporally averaged, respectively, smooth temporal and spatial profiles are obtained. The temporal fluctuations discussed here are quite general so that they are expected to appear not only in the classical waves but also in quantum waves. For example, electronic quantum wave packet transmitted through a disordered medium would yield temporal fluctuations in the conductance, if suitable experimental conditions are satisfied. The temporal fluctuations in conductance would have an universal order of

e^2/h, and in addition, it would change its profile completely as a function of applied magnetic field by an amount of order e/h.[32]

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