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The precise shape and width of the temporal and spectral correlation functions in fluctuations that appear when coherent optical pulses propagate through static disordered media depend on the observation time and the pulse duration of the incident light. When the pulse duration of the incident light is much longer than the distribution of trajectory length, a previously shown minimum uncertainty relation in the temporal and spectral fluctuations breaks down. Detailed experimental results obtained using picosecond pulses show a good agreement with the theoretical calculation based on the diffusion approximation. [S0163-1829(98)06133-5]

When coherent laser beams irradiate disordered media, dark and bright irregular fluctuation patterns appear.[1] These fluctuation patterns are called speckles and are a direct result of random interference of light that traveled through a number of different scattering paths in disordered media. It is an analog of conductance fluctuations in microelectronics, where coherent quantum waves interfere with each other.[2] In the conventional speckle measurements, continuous wave lasers of sufficient coherence length are used as the incident light source.[1] It has also been shown that when coherent optical pulses propagate through a thick disordered medium and the transmitted light is time resolved, speckle appears not only in the space domain but also in the time domain.[3,4] Various correlations that appear in the fluctuations are of recent interest in fundamental physics.[5–10] In a previous paper, we examined the temporal and spectral correlation functions of the two-dimensional speckles in the space and time domains. As a result, we demonstrate that the time-energy uncertainty relations. $\delta\tau\delta\omega\sim 1$ or $\delta\tau\delta E\sim\hbar$ are established in these fluctuations.[11] In this paper, we examine the precise shape and width of spectral and temporal correlation functions in static disordered media and show that these correlation functions depend on the observation time and the pulse duration of the incident light. We also show that the previously shown minimum uncertainty relation between the temporal and spectral fluctuations breaks down when the condition $t_p \ll \delta t_{\mathrm{trans}}$, where t_p is the pulse duration and $\delta t_{\mathrm{trans}}$ is the typical flight time for transmitted pulse, is not valid. The dependence of the correlation functions on the observation time and the pulse duration is well explained when we consider distribution of the trajectory length for the photon migration in the medium.

The experimental setup was the same as the previous one in Ref. 11. We used thick and thin samples. The thick sample was microparticles of $BaSO_4$ of 1 μm in diameter compacted to a thickness of 1 mm between two optical flat glass plates. For the thin sample, we used a frosted glass of 1 mm thickness. Scattering of the incident light occurs at one grained surface of the frosted glass. The incoming light source was the second harmonics of a mode-locked Nd^{3+}:YAG (yttrium aluminum garnet) laser. The pulse duration was 93 psec, the wavelength was 532 nm, and the average power was 100 mW. A quartz plate of 2 mm in thickness was inserted in the laser cavity as a wavelength tuning element. The wavelength of the YAG laser was tunable within a range of 400 GHz. The transmitted light was time resolved using a streak camera in front of which a polarizer was placed. The time resolution of the experimental system was 5 psec. The scattered light forms two-dimensional speckle in space and time domains. We calculated mutual correlation between fluctuations observed at the time t with incoming light of frequency ω and those observed at the time $t+\tau$ with incoming light of frequency $\omega+\Delta\omega$.

Figure 1 shows the experimental result for normal transmitted pulse profile through the $BaSO_4$ sample. Open circles are calculated curves based on the diffusion approximation. Parameters used are for the diffusion constant $D=0.17$ m^2/nsec and for the absorption lifetime $\tau_a=0.9$ nsec. Arrows at 272 and 746 psec represent the time at which the temporal and spectral correlation functions are examined. Figures 2 and 3 show the experimental results of temporal and spectral correlation functions as a function of the delay time and the spectral shift, respectively, for the thick sample. The observation times are 272 and 746 psec. These curves are normalized at the value of $C(0)$. It is seen that the peak position and the width of the correlation functions depend on the observation time. In Fig. 3, the experimental results of

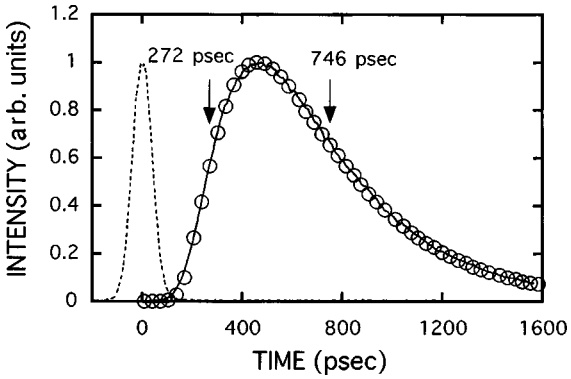

FIG. 1. The solid line is the transmitted pulse profile through the $BaSO_4$ sample. The dotted line is the incident pulse. Open circles are calculated curves based on the diffusion approximation. Arrows represent time when the correlation functions are examined.

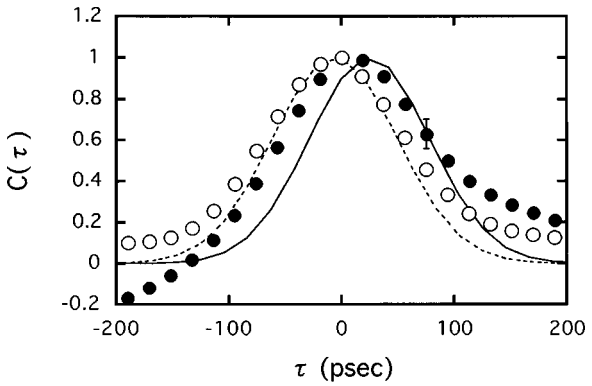

FIG. 2. Normalized temporal correlation functions $C(\tau)$ on the condition for the $BaSO_4$ sample. Solid and open circles are for the observation time of 272 and 746 psec, respectively. The solid and the broken lines are theoretical curves for the observation time of 272 and 746 psec, respectively.

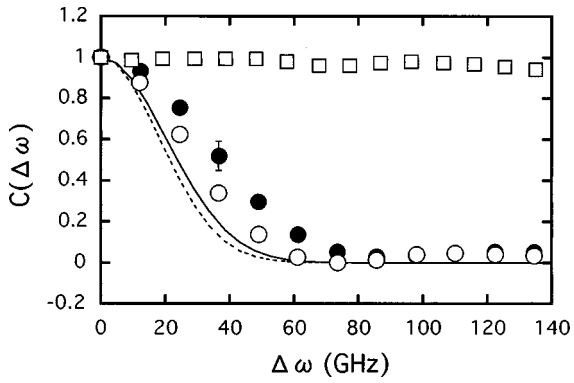

FIG. 3. Normalized spectral correlation functions $C(\Delta\omega)$. Solid and open circles are for the observation time of 272 and 746 psec, respectively, for the $BaSO_4$ sample. The solid and the broken lines are theoretical curves for the observation time of 272 and 746 psec, respectively. Squares are experimental correlation curves for the frosted glass.

spectral correlation function for the thin sample are also shown with open squares together with the correlation function for the thick sample. For the thin sample, the spectral correlation function is almost constant within the spectral range examined. In Fig. 4, solid circles represent the experimental result of the temporal correlation function for the thin sample. In the thin sample, the width of the temporal correlation function is 1.41 times as narrow as that of the thick sample. From Figs. 2 and 3, the products of the full width at the half maximum of the temporal and spectral correlation functions are 12.1 at 272 psec and 9.1 at 746 psec, respectively, for the thick sample. This value is more than 350 for the thin sample from Figs. 3 and 4. We see that the minimum uncertainty relation holds good in the thick sample, while it breaks down in the thin sample.

Now we compare experiments with the theory. Employing a diffusion approximation, the spectral and temporal correlation functions in static disordered optical media are given as[11]

$$\begin{aligned} C(t,\tau,\omega,\Delta\omega) &= \langle I(t,\omega) I(t+\tau,\omega+\Delta\omega)\rangle \\ &= c^{-2}\int_0^\infty ds_1 \int_0^\infty ds_2 T(s_1/c) T(s_2/c) \\ &\quad \times \{\tilde{E}^*_{\rm in}(t-s_1/c)\tilde{E}_{\rm in}(t+\tau-s_1/c) \\ &\quad \times \tilde{E}_{\rm in}(t-s_2/c)\tilde{E}^*_{\rm in}(t+\tau-s_2/c) \\ &\quad \times \exp(-i\Delta\omega s_1/c)\exp(+i\Delta\omega s_2/c)\}, \end{aligned} \tag{1}$$

where angular brackets denote an ensemble average, $T(s/c)$ is the trajectory-distribution function, and

$$E_{\rm in}(t,\omega) = \tilde{E}_{\rm in}(t)\exp(i\omega t) \tag{2}$$

is the electric field of the incoming light at a time t. ω is the central frequency and $\tilde{E}_{\rm in}(t)$ is the slowly varying envelope function, $s=c/t$ is the length of the trajectory, c is the velocity of light in the medium. We define temporal and spectral correlation functions with conditions that $\Delta\omega=0$ and $\tau=0$, respectively.

We consider two limiting cases in Eq. (1). The first one is the case $t_p \ll \delta t_{\rm trans}$, where the pulse duration t_p is much shorter than the typical flight time for the transmitted pulses $\delta t_{\rm trans}$, where $\delta t_{\rm trans} \sim L^2/l^* c$, L is the sample thickness, and l^* is the transport mean free path. The second one is the case $t_p \gg \delta t_{\rm trans}$, where the pulse duration is much longer than the flight time. The $BaSO_4$ sample corresponds to the first case, while the frosted glass corresponds to the second case. In the first case, the trajectory-distribution function does not change significantly during a period of pulse duration. The temporal correlation function is simplified as[11]

$$C(t,\tau,\omega,0) = c^{-2}|T(t)|^2 \left| \int_0^\infty ds\ E^*(s/c)E(s/c+\tau) \right|^2. \tag{3}$$

In this case, the temporal correlation function is the field correlation of the incoming pulse. The spectral correlation function is also written as

$$C(t,0,\omega,\Delta\omega) = c^{-2}|T(t)|^2 \left| \int_0^\infty ds\ I(s/c)\exp(i\Delta\omega s/c) \right|^2, \tag{4}$$

where $I(s/c)$ is the intensity function of the incoming light. The spectral correlation function is the Fourier transform of

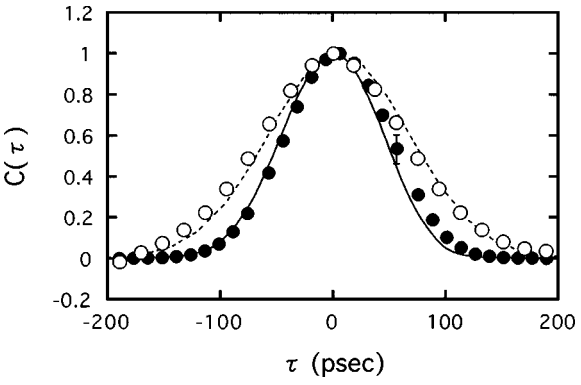

FIG. 4. Normalized temporal correlation functions $C(\tau)$. Solid and open circles are for the frosted glass and the $BaSO_4$ sample.

the intensity function of the incoming pulse. In the second limiting case, where $t_p \gg \delta t_{\rm trans}$, we can assume that the trajectory-distribution function is a δ function compared with the pulse duration

$$T(s/c) \rightarrow c\,\delta(s). \tag{5}$$

Then the temporal correlation function of Eq. (1) is represented as

$$C(t,\tau,\omega,0) = |\tilde{E}_{\rm in}(t)|^2 |\tilde{E}_{\rm in}(t+\tau)|^2. \tag{6}$$

The width of the correlation function is that of the envelope function of the pulse intensity. Assuming a Gaussian-shaped incoming pulse, the full width at the half maximum of Eq. (3) is $\sqrt{2}$ times as narrow as that in Eq. (6). This value shows a good agreement with experiments shown in Fig. 4. For the long pulse limit, $E(t-s/c)$ is constant during the integration with s, $E(t-s/c) \rightarrow E(t)$; then the spectral correlation function is represented as

$$C(t,0,\omega,\Delta\omega) = |I(t)|^2 \left| \int_0^\infty ds\ T(s/c) \exp(-i\Delta\omega s/c) \right|^2. \tag{7}$$

When $t_p \sim \delta t_{\rm trans}$, Eqs. (3), (4), (6), and (7) are not good. We must numerically calculate Eq. (1) to obtain the shape of the correlation functions.

We now compare experiments with the numerical calculation and examine the influence of the trajectory-distribution function on the correlation functions. Let us consider the thick sample. For the thick sample, where $t_p \ll \delta t_{\rm trans}$, Figs. 2 and 3 show the numerically calculated temporal and spectral correlation curves on the basis of Eq. (1). In Fig. 2, it is seen that the numerical curves show a good agreement with the experimental curves. At the observation time 272 psec, where the slope of $T(s/c)$ steeply rises in Fig. 1, the peak of the temporal correlation functions shifts to a later time region. For the spectral correlation functions, the relative width of the experimental and calculated curves show a similar tendency, that is, at the observation time 272 psec, the width is broader than that at 746 psec. This arises because the slope of the distribution function is steeper at 272 psec than that at 746 psec; therefore the effective distribution of the trajectory length contributing to fluctuations observed at 272 psec is narrow compared with that at 746 psec. The difference in absolute widths between experimental and calculated curves may be attributed to the frequency stability of the incident light source in our experiment. Considering the frequency jitter 20 GHz, we could explain the difference between experiments and the theory. Next we see the thin sample. In the thin sample, where $t_p \gg \delta t_{\rm trans}$, the spectral correlation function is the Fourier transform of the trajectory-distribution function. The width is of the order of $(\delta t_{\rm trans})^{-1} = (L^2/l^* c)^{-1}$.[12,13] On the other hand, the width of the temporal correlation function is only $\sqrt{2}$ times as narrow as that of the thin sample. The minimum uncertainty relation between the temporal and spectral fluctuations breaks down.

We may see an analog between the spectral correlation measurement in fluctuations in the disordered medium and homogeneous and inhomogeneous broadening in a resonance quantum two-level system. In a disordered medium, modes are spreading in frequency domain with a spectral density $(2\pi/L)^3$, where L is the size of the sample. This spreading of modes would correspond to the inhomogeneous broadening. On the other hand, each mode in the random medium has a spectral width determined by the lifetime of the mode $\delta t_{\rm trans}$, which would correspond to the homogeneous broadening. When $t_p \gg \delta t_{\rm trans}$, the spectral width of the correlation function is $(\delta t_{\rm trans})^{-1}$ and is independent of the pulse duration. The product of $\delta\tau\delta\omega$ is not that of the minimum value.

In summary, we have shown that the temporal and spectral correlation functions depend on the observation time and pulse duration of the incident pulse. This dependence is originated in the trajectory distribution function in the medium. The minimum uncertainty relation between the temporal and spectral fluctuation breaks down when $t_p \gg \delta t_{\rm trans}$.

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