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(Received 28 February 1996)

We confirmed that the uncertainty relation between time and energy is established in fluctuations in the space and time domains that appear when coherent optical pulses propagate through a disordered static medium. The widths of temporal and spectral correlation functions of these fluctuations satisfy the relation $\delta\tau\delta\omega\sim1$, that is, $\delta\tau\delta E\sim\hbar$. The number of fluctuations is discussed on the basis of the uncertainty relation. [S0163-1829(96)52330-1]

The Heisenberg uncertainty principle states that conjugate physical quantities cannot both be precisely specified at the same time. The time-energy uncertainty relation is one example of this principle. With a relation $E=\hbar\omega$, where E is energy, $\hbar$ is Planck's constant, and ω is angular frequency, this principle represents the mathematical fact that the extension of a wave packet and that of its Fourier transform cannot simultaneously be made arbitrary small. The uncertainty relation is a fundamental feature of wave phenomena. We can find many examples in interactions between light and matter. In order that physical properties of the system be substantially modified over a time interval t, the product of t and energy uncertainty δE, must satisfy $t\delta E=\hbar$.[1] When an atom, excited to a state of mean lifetime of t, emits light, the uncertainty of energy of the emitted light is on the order of t^{-1}.[2] It is well known that in the generation of ultrashort laser pulses, a broad spectrum light source of the order of the inverse of the pulse duration is necessary.

In this paper, we deal with fluctuations in static disordered media. Various correlations that develop in these fluctuations are of recent interest.[3,4] Memory effects have been investigated in various conditions.[3–6] The existence of long-range correlations in fluctuations has been theoretically predicted and confirmed by careful experiments.[7,8] It was also shown that two-dimensional fluctuations appear in the space and time domains when coherent optical pulses propagate through random medium and the transmitted light is time resolved.[9,10] Here, we examine the temporal and spectral correlations of these two-dimensional fluctuations in the space and time domains. As a result, we demonstrate that the uncertainty relation $\delta\tau\delta\omega\sim1$ or $\delta\tau\delta E\sim\hbar$ is established in these fluctuations. The number of fluctuations in the frequency and time domains are discussed on the basis of the uncertainty relation.

We have experimentally examined the uncertainty relation between time and frequency in the fluctuations in the space and time domains. The sample was microcrystallite of $BaSO_4$ compacted to a thickness of 1000 μm between two optically flat glass plates. The incoming light source was the second harmonic of a mode locked Nd^{3+} yttrium aluminum garnet (YAG) laser. The pulse duration was 80 psec. A quartz plate of 2 mm in thickness was inserted in the laser cavity as a wavelength tuning element. The plate was rotated around an axis vertical to the laser cavity. Controlling the angle of this quartz plate, we tuned the wavelength of the laser emission in the range of 400 GHz in the vicinity of 532 nm. The wavelength was monitored using 2 m-length single monochrometer. The diameter of the laser beam was about 2 mm. The incident laser light illuminated the surface of the sample. The light transmitted to the back surface of the sample forms a far-field speckle pattern. When the transmitted light is time resolved, fluctuations appear in the space and time domains. The incident wave vector of the incoming light and the wave vector of the outgoing light were both nearly normal to the sample surface. The scattered light was detected using a synchronously scanning streak camera. The angular resolution of the detection system was 0.1 mrad. The time resolution was 5 psec.

Figure 1 shows examples of the time-resolved fluctuations obtained when the incoming light was scanned at seven different frequencies. It is seen that the characteristic structures of the fluctuations gradually change in shape. To analyze quantitatively the loss of the correlation, the correlation was calculated for 15 sets of experimentally obtained time-

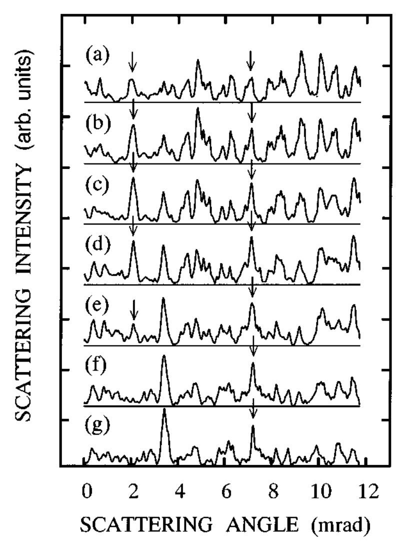

FIG. 1. Examples of the time-resolved fluctuations as a function of scattering angles at an observation time of 1230 psec. (a) is an initial reference pattern, while in (b) the spectrum was changed by 12.3 GHz, in (c), 24.6 GHz, in (d), 36.9 GHz, in (e), 49.2 GHz, in (f), 61.5 GHz, and in (g), 73.8 GHz, respectively. Small arrows are shown to call attention to characteristic structures in patterns.

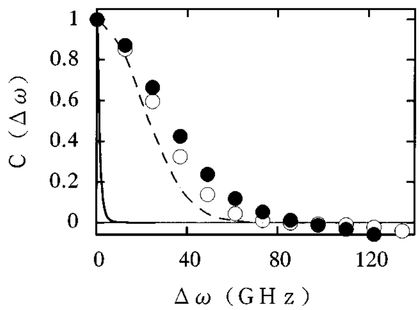

FIG. 2. Normalized spectral correlation function $C(\Delta\omega)$. Solid circles and open circles are for the observation time of 916 and 1230 psec, respectively. The solid and the broken lines are theoretically calculated curves for the steady state measurement and the time-resolved measurement, respectively.

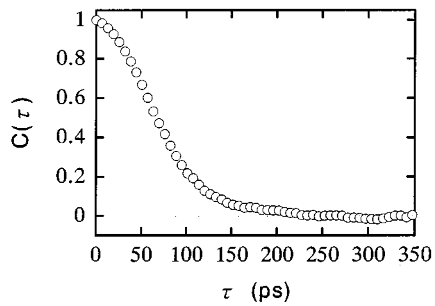

FIG. 3. Normalized temporal correlation function $C(\tau)$.

resolved speckle patterns including those shown in Fig. 1. The correlation curve $C(\Delta\omega)$ as a function of the spectral shift normalized at the value of $C(0)$ is shown in Fig. 2. The full width at half maximum (FWHM) of the correlation function is $\delta\omega=5.6\times10^{10}$ Hz for the observation time of 1230 psec. The correlation curve as a function of delay time is shown in Fig. 3. The FWHM of the correlation function is $\delta\tau=1.2\times10^{-10}$ sec. The product of $\delta\tau\delta\omega=6.7\sim1$, or with the relation $E=\hbar\omega$, $\delta\tau\delta E=\hbar$, satisfies the uncertainty relation.

To understand the uncertainty relation in the time-resolved fluctuations, we apply a real-space theory of the fluctuations. We assume that the scattered light has a linear polization. Other cases can be dealt with in an identical fashion. The electric field of the incoming light at a point (ξ,ζ) on the medium at a time t is represented as,

$$E_{\mathrm{in}}(t,\xi,\zeta)=\widetilde{E}_{\mathrm{in}}(t,\xi,\zeta)\exp[i\omega t], \tag{1}$$

where ω is the central frequency and $\widetilde{E}_{\mathrm{in}}(t,\xi,\zeta)$ is the slowly varying envelope function. The electric field of the out going light is represented as

$$E_{\mathrm{out}}(t,\xi,\zeta)=\int_0^t dt'\int_{-\infty}^{\infty}d\xi'\int_{-\infty}^{\infty}d\zeta' \times L(t\text{-}t',\xi,\xi',\zeta,\zeta')E_{\mathrm{in}}(t',\xi',\zeta'), \tag{2}$$

where $L(t\text{-}t',\xi,\xi',\zeta,\zeta')$ is the field propagator from (ξ',ζ') to (ξ,ζ) in the static sample, which is made up all possible trajectories and given as,[10]

$$L(t\text{-}t',\xi,\xi',\zeta,\zeta')\rightarrow\Sigma W\delta[(t\text{-}t')-s/c], \tag{3}$$

where $s=ct$ is the length of the trajectory, c is the velocity of light in the medium, and W is the amplitude probability of the trajectory. The 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 $\omega+\Delta\omega$, is represented,

$$\begin{aligned}C(t,\tau,\omega,\Delta\omega)&=\langle I_{\mathrm{out}}(t,\omega,\xi,\zeta)I_{\mathrm{out}}(t+\tau,\omega+\Delta\omega,\xi,\zeta)\rangle\\&=\Big\langle\int_0^{\infty}dt'\int_0^{\infty}dt''\int_0^{\infty}dt'''\int_0^{\infty}dt''''\Sigma\Sigma\Sigma\Sigma W_1^*W_2W_3^*W_4\delta[(t-t')-s_1/c]\delta[(t-t'')-s_2/c]\\&\quad\times\delta[(t+\tau-t''')-s_3/c]\delta[(t+\tau-t'''')-s_4/c]\widetilde{E}_{\mathrm{in}}^*(t')\widetilde{E}_{\mathrm{in}}(t'')\widetilde{E}_{\mathrm{in}}^*(t''')\widetilde{E}_{\mathrm{in}}(t'''')\exp[-i\omega t']\exp[i\omega t'']\\&\quad\times\exp[-i(\omega+\Delta\omega)t''']\exp[i(\omega+\Delta\omega)t'''']\Big\rangle,\end{aligned} \tag{4}$$

where angular brackets $\langle\ \rangle$ denote ensemble average. The four complex phasers in Eq. (4) are independent random variables having zero mean value. There are two types of terms in the summation for which the ensemble average do not vanish. The nonzero contributions arise when $\xi_1=\xi_2$ and $\xi_3=\xi_4$, or $\xi_1=\xi_3$ and $\xi_2=\xi_4$, where ξ_i represents trajectory i. The sum of trajectories can be replaced with the following integration of the trajectory distribution function, $T(s/c)$,

$$\Sigma|W|^2\rightarrow c^{-1}\int dsT(s/c), \tag{5}$$

with the normalization condition,

$$c^{-1}\int_0^{\infty}dsT(s/c)=1. \tag{6}$$

Here we assume that the distribution of the length of trajectories is much broader than the pulse duration. In our experiment, the typical flight time of the transmitted pulses is of the order of 1 nsec, while the pulse duration of the mode-locked laser pulse is 80 psec. With this approximation, the correlation function is,

$$C(t,\tau,\omega,\Delta\omega)$$

$$=\{T(t)\}^2\int_0^\infty ds_1\int_0^\infty ds_2 c^{-2}\{I(t-s_1/c)I(t+\tau-s_2/c)$$
$$+\widetilde{E}^*(t-s_1/c)\widetilde{E}(t+\tau-s_1/c)$$
$$\times\widetilde{E}(t-s_2/c)\widetilde{E}^*(t+\tau-s_2/c)$$
$$\times\exp[-i\Delta\omega s_1/c]\exp[i\Delta\omega s_2/c]\}, \qquad (7)$$

where $I(t)$ is the intensity function of the incoming pulse. It is convenient to define a spectral correlation function with a condition that $\tau=0$. This function is given as,

$$C(t,0,\omega,\Delta\omega)=\{T(t)\}^2\{1+|P(\Delta\omega)|^2\}, \qquad (8)$$

where

$$P(\Delta\omega)=c^{-1}\int_0^\infty ds I(s/c)\exp[i\Delta\omega s/c] \qquad (9)$$

is the Fourier transform of the intensity function of the incoming light. It is predicted that the width of spectral correlation function is independent of the observation time. We also define a temporal correlation function with a condition that $\Delta\omega=0$. This function is given as,

$$C(t,\tau,\omega,0)=\{T(t)\}^2\{1+|G(\tau)|^2\}, \qquad (10)$$

where

$$G(\tau)=c^{-1}\int_0^\infty ds\widetilde{E}^*(s/c)\widetilde{E}(s/c+\tau) \qquad (11)$$

is the field correlation function of the incoming light. We can now quantitatively compare the theory with experimental results. Figure 2 are theoretically calculated curves for the steady state[11,12] and the time-resolved measurements. The diffusion constant used is 0.13 mm^2/nsec.[6] It is seen that the theoretically calculated curve for the time-resolved measurement shows good agreement with the experiments. For simplicity, we assume that the intensity function of the incoming pulse is Gaussian shaped. From Eqs. (9) and (11) the product of $\delta\tau\delta\omega$ is theoretically calculated to be 5.5, which shows good agreement with the experimental value of 6.0.

Let's consider a relation between the degrees of freedom in the medium and the fluctuations. In the steady state measurement, each Airy area has a fixed complex phase. Therefore, the number of fluctuations in the time domain in a single Airy area is, $N_T^{\text{steady}}=1$ for the steady state measurement. When the fluctuations are time resolved, the number of fluctuations in a single coherence area increases. The typical flight time for the transmitted pulses is $\delta t_{\text{trans}}\sim L^2/\ell^*c$, where ℓ^* is the transport mean-free path and L is the sample thickness. Therefore, each Airy area has fluctuations of $N_T^{\text{pulse}}=\delta t_{\text{trans}}/t_p$ in number in the time domain. Thus,

$$N_T^{\text{pulse}}/N_T^{\text{steady}}\sim L^2/c\ell^*t_p. \qquad (12)$$

From Eq. (9), the spectral width of the fluctuations is $\delta\omega^{\text{pulse}}\sim t_p^{-1}$ in the time-resolved measurement. This value is $\delta\omega^{\text{steady}}\sim c\ell^*/L^2$ in the steady state measurements with continuous wave lasers.[11,12] We can understand these situations as follows. The propagating modes in the medium have a lifetime of the order of $\delta t_{\text{life}}\sim L^2/\ell^*c$. When wave is not localized,$(\delta t_{\text{life}})^{-1}/\Delta\omega_M$ modes in number are excited simultaneously in the medium, where $\Delta\omega_M$ is the mode spacing in the medium. The fluctuations will change its profile when the spectrum of the incoming light is changed of the order of $(\delta t_{\text{life}})^{-1}$. In the time-resolved measurement, the number of modes to be excited is $t_p^{-1}/\Delta\omega_M$. The profile of fluctuations, thus, changes when the spectrum is changed of the order of t_p^{-1}. The rate of number of fluctuations included within a certain frequency window observed in the steady state and in the time-resolved measurements are

$$N_S^{\text{pulse}}/N_S^{\text{steady}}\sim\ell^*t_pc/L^2. \qquad (13)$$

From Eqs. (12) and (13) the total ratio of the number of fluctuations in frequency and time domains is $[N_T^{\text{pulse}}/N_T^{\text{steady}}][N_S^{\text{pulse}}/N_S^{\text{steady}}]\sim 1$, and is constant. It is concluded that the difference between the steady state and the time-resolved measurements is the distribution of fluctuations between the frequency and time domains. The total number of fluctuations is constant.

In summary, we have experimentally confirmed that the uncertainty relation $\delta\tau\delta\omega\sim 1$ or $\delta\tau\delta E\sim\hbar$ is established in the time-resolved fluctuations in the space and time domains. Seemingly, time resolving increases the number of fluctuations. However, time resolving only changes the distribution of fluctuations between the frequency and time domains.

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