PHYSICAL REVIEW B VOLUME 45, NUMBER 2 1 JANUARY 1992-II

Rapid Communications

Rapid Communications are intended for the accelerated publication of important new results and are therefore given priority treatment both in the editorial office and in production. A Rapid Communication in **Physical Review B** *should be no longer than 4 printed pages and must be accompanied by an abstract. Page proofs are sent to authors.*

Time-resolved fluctuations and the memory effect in disordered optical media

Makoto Tomita
Department of Physics, Faculty of Science, Shizuoka University, 836 Ohya, Shizuoka City 422, Japan
(Received 26 July 1991)

The memory effect that appears in disordered optical media is investigated by using time-resolved spatial optical fluctuations. It is theoretically predicted that the memory effect is lost with time after the incident laser pulse enters the medium. This prediction is verified by experiments in the picosecond time region.

Optical waves propagating through disordered media appear randomly scattered. However, they actually retain many interesting regularities. Weak localization of photons[1–3] by constructive interference of time-reversed pairs of the scattering sequence is one of such regularities. Various correlations that develop in optical fluctuations known as speckles are of recent interest in research concerning wave propagation through disordered systems.[4] The memory effect[5–8] appearing in short-range correlations shows that waves propagating through strongly disordered media still retain information on the wave front of the incoming light. Long-range and infinite-range correlations, which are closely related to the universal conductance fluctuations in disordered electric systems, are also predicted[4] and are experimentally confirmed in carefully prepared optical media.[9,10] These correlations have been used in recent research of the frequency window of photon localization in strongly disordered optical media.[11,12] From the viewpoint of applications, speckles have also been used in a wide variety of optical measurements, such as the measurement of small displacement or oscillation, surface conditions, and material fatigue on the order of optical wavelength.

Speckles are usually observed by using continuous-wave (cw) lasers of a suitable coherence length.[13] In contrast with the use of cw lasers, when coherent-light pulses propagate through such samples in which the typical length of the light trajectory is longer than the pulse length, conventional spatial speckles reduce in contrast because the light cannot coherently sample the entire volume.[14] Under this condition, however, large fluctuations appear in the time domain due to the random interference of multitudinously scattered waves. These temporal fluctuations have recently been observed in the picosecond time region.[15] They are not fluctuations that reflect the dynamic properties of scatterers as discussed in the field of quasielastic light scattering[16] and diffusing-wave spectroscopy,[17–20] but are fluctuations that reflect the geometrical configuration of the scatterers.

We can recognize these temporal fluctuations as speckles in the time domain. The fluctuations have the following similar characteristics of conventional spatial speckles: (a) They are observed when coherent light is illuminated onto disordered media; (b) they are produced by the random interference of multitudinously scattered waves; (c) they reflect the random sample configuration; and (d) they are reproducible, noiselike, irregular scattering patterns following a stationary Gaussian process (ignoring higher-order correlations). The temporal speckles form "spatial-temporal speckles" combining with the conventional spatial speckles.[15]

The purpose of the present paper is to show that a wide variety of scientific and application-oriented speckle experiments, which previously have been performed only in the steady state by using cw lasers, can be studied by the time-resolved approach, utilizing the spatial-temporal speckles. We consider, as an example, the memory effect, which has been investigated most widely among the speckle correlations in disordered media,[5–8] and report a joint theoretical and experimental study of the time-resolved memory effect. It is found that the memory effect is lost with time after the incident light pulse enters the medium. A similar technique has been applied to examine the line shape of the coherent backscattering peak.[21] It should be noted, however, that the time-resolved speckle correlations cannot be experimentally studied but for the concept of the spatial-temporal speckles treated here.

We apply the real-space theory of the time-resolved memory effect.[7] Ignoring higher-order terms, the correlation function representing the memory effect is given by

$$C_{aba'b'}(t)=\langle \delta T_{ab}(t)\delta T_{a'b'}(t)\rangle$$
$$=\sum_{\xi_1}\sum_{\xi_2}\{W_{\xi_1}(t-s_1/c+q_aR_{\mathrm{in}1}-q_bR_{\mathrm{out}1})W_{\xi_2}(t-s_2/c+(q_a+\Delta q_a)R_{\mathrm{in}2}-(q_b+\Delta q_b)R_{\mathrm{out}2})\}^2$$
$$\times\exp\{-i[\Delta q_a(R_{\mathrm{in}1}-R_{\mathrm{in}2})-\Delta q_b(R_{\mathrm{out}1}-R_{\mathrm{out}2})]\}\,, \qquad (1)$$

where $\langle \cdots \rangle$ is an ensemble average, q_a and $q_a' = q_a + \Delta q_a$ are the transverse incident wave vectors, q_b and $q_b' = q_b + \Delta q_b$ correspond to the transmitted light, T_{ab} is the scattering intensity in the q_b direction produced by light with the incident wave vector q_a, $\delta T_{ab}(t) = T_{ab}(t) - \langle T_{ab}(t) \rangle$, c is the effective velocity of light in the medium, s and W are the optical path length and the time-dependent transmittance of trajectory ξ, and R_{in} and R_{out} are vectors parallel to the sample surfaces representing positions of the first and last scatterers of the trajectory, respectively. As for the sum of the trajectories in Eq. (1), it is necessary to add up trajectories with optical path length $t - t_p < s < t + t_p$, where t_p is the incident pulse duration. From this summation, the time scale of the temporal fluctuations is on the order of the incident pulse duration.[15] This sum of trajectories can be replaced with the following integration,

$$\sum_{\xi} \rightarrow \int ds \int d|R_{\text{in}} - R_{\text{out}}| P(s, |R_{\text{in}} - R_{\text{out}}|) , \qquad (2)$$

where $P(s, |R_{\text{in}} - R_{\text{out}}|)$ is the distribution function of the trajectory, which is calculated based on the diffusion approximation. Integration with respect to $|R_{\text{in}} - R_{\text{out}}|$ leads to the time-resolved correlation function,

$$C_{aba'b'}(t) = I(t)^2 \delta_{\Delta q_a, \Delta q_b} G(t, \Delta q_a) , \qquad (3)$$

where

$$G(t, \Delta q_a) = \exp[-(2Dt\Delta q_a^2)],$$
$$I(t) \propto (4\pi Dt)^{-1/2} \sum_{n=-\infty}^{\infty} \left[\exp\left(\frac{-[(2n-1)L + 2a]^2}{4Dt} \right) - \exp\left(\frac{-[(2n-1)L]^2}{4Dt} \right) \right] . \qquad (4)$$

δ is a function that has a width comparable to that of the spatial speckle size, L is the sample thickness, D is the diffusion constant, a is a parameter on the order of the transport mean free path l, representing the boundary condition of the light injection. We assumed that the linear optical path differences appearing at the incoming and outgoing surfaces of the sample are sufficiently short compared to the incoming light pulse length, and set $I(t)$ equal to the transmitted pulse profile. The correlation function $G(\Delta q_a)$ for $\Delta q_a = \Delta q_b$ is Gaussian shaped. The full width at half maximum (FWHM) decreases with time and the memory effect is lost with time after incident pulse enters the medium. The correlation function of the time-resolved memory effect in the presence of absorption is given by replacing $I(t)$ in Eq. (3) with $I_a(t) = I(t) \times \exp(-t/\tau_a)$, where τ_a is the photon lifetime in the medium. Unlike the steady-state memory effect,[7] the shape and width of the correlation function of time-resolved memory effect with respect to Δq_a is not subject to the effect of absorption.

We have experimentally examined the time-resolved behavior of the memory effect and have confirmed the possibility of the time-resolved speckle experiments. The experimental setup is similar to that used in the previous experiments.[15] The light source is the second harmonic of a mode-locked Nd^{3+} yttrium aluminum garnet laser, the pulse duration is 70 ps, and the repetition rate is 82 MHz. The sample is corpuscles of $BaSO_4$ compacted to a thickness of 820 μm between two optically flat glass plates. The laser light is expanded into a collimated beam of about 6 mm in diameter and illuminated onto the incoming surface of the sample. The light transmitted to the other surface of the sample forms far-field speckles. The incident vector of the incoming light beam and the outgoing vector of the observed speckles are both nearly normal to the sample surfaces. The speckles are detected as a function of scattering angle by a two-stage microchannel plate photomultiplier tube mounted on a translational stage and are time resolved by a time-correlated single-photon counting system. The angular resolution of the detection system is 0.03 mrad and is high enough to resolve one coherent cell of the time-resolved spatial speckles.

First, the experimental result of ordinary transmitted pulse is shown in Fig. 1. This spatially ensemble-averaged smooth profile was obtained by observing a large scattering area with a condenser lens placed between the sample and photomultiplier tube. The solid circles indicate a theoretically calculated curve based on the diffusion approximation.[22] The parameters used are $D = 0.13$ mm^2/ns and $\tau_a = 4$ ns, which give a good fit to the experimental curve. The arrows A and B indicate the points of time when the time-resolved memory effect is examined. The points of time A and B are 380 and 820 ps, respectively. Figure 2 shows examples of time-resolved speckle patterns obtained when the sample was rotated by five different angles. Since the incident wave vector of the incoming laser beam and the outgoing vector of the observed speckles are both nearly normal to the sample surfaces in our experi-

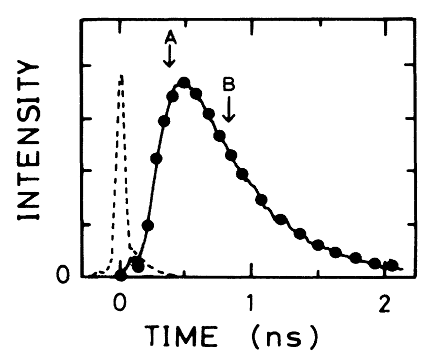

FIG. 1. The solid line is a spatially ensemble-averaged temporal profile of a transmitted 70-ps pulse. Solid circles represent a calculated curve based on the diffusion approximation. The dashed line is the instrument response function to the input pulse. Arrows A and B indicate points of time when the time-resolved memory effect was examined.

mental arrangement, the characteristic structure of the speckles does not change in position but gradually changes in shape. To compare quantitatively the loss of the speckle correlations with the theory, the correlation functions were calculated for 20 sets of experimentally observed time-resolved speckle patterns including those shown in Fig. 2. The value of the correlation functions at $\delta\theta=0$ is about 0.6 in our experiments and is smaller than the value of unity expected from theory. This is probably because the time resolution of the experimental system is not high enough to observe the temporal speckles completely. The correlation functions normalized by the value of $C(0)$ are shown in Figs. 3 and 4 on linear and semilog scales, respectively.

The behavior of the correlation functions show that the memory effect is more enhanced at 380 ps than at 820 ps, as predicted from the theory. However, contradictory to the theoretical prediction, the correlation curves as a function of the angle of the sample rotation are not Gaussian shaped. This discrepancy is marked in the region where the angle is small. The correlation functions theoretically calculated based on the diffusion constant ($D=0.13$ mm^2/ns) determined by the pulse transmission experiment shown in Fig. 1 are indicated by the dashed lines in Fig. 3. The solid lines in Fig. 3 indicate the fits of experimental data to a Gaussian line shape by the least-squares method, ignoring the small angle-of-rotation region where the experimental data differ significantly from the Gaussian shape. FWHM of the correlation function at 380 ps is 1.4 times larger than that at 820 ps. This ratio is almost the same as the theoretically predicted ratio of 1.47 by Eq. (3).

The origin of the discrepancy between the theory and the time-resolved speckle experiments is not certain, but it may originate in the internal surface reflection of the sample, which is the case with the steady-state memory effect.[6,23] When the light wave reaches the outgoing surface, it would suddenly increase in the transverse diffusion area due to the internal surface reflection in the sample. The memory effect is expected to be lost to a considerable degree in place of this effect. This effect, on the other hand, does not appreciably affect the diffusion constant determined by the pulse transmission experiment,[24] when the sample thickness is much greater than the elastic mean free path. Consequently, differences would appear between the theoretical curves that are calculated based

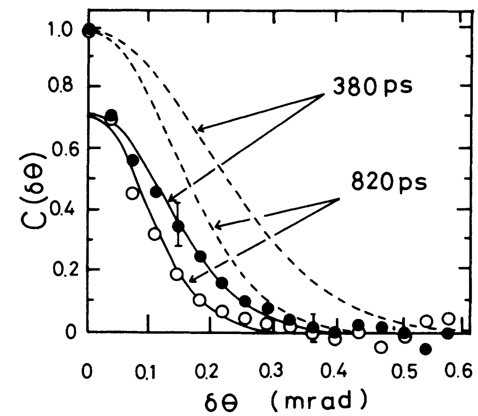

FIG. 3. Solid and open circles represent the normalized correlation function $C(\delta\theta)$ for sampling times of 380 and 820 ps, respectively. The solid lines are fitted curves to the experimental data. The dashed lines are theoretically calculated curves with the diffusion constant determined from the pulse transmission experiment shown in Fig. 1.

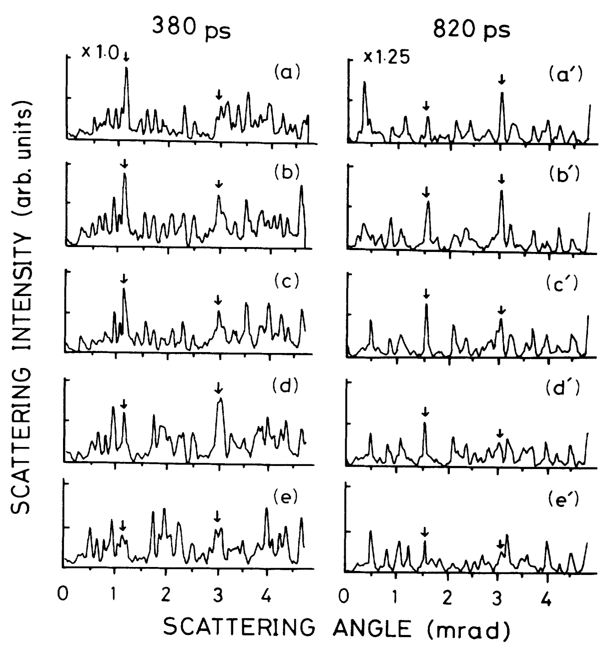

FIG. 2. Examples of the time-resolved spatial speckle patterns. The left-hand side and the right-hand side (multiplied by 1.25 with respect to the left-hand side) show time-resolved speckles at sampling times of 380 and 820 ps, respectively. (a) and (a′) are initial reference patterns, while in (b) and (b′), the sample was rotated by 36 μrad, in (c) and (c′) by 72 μrad, in (d) and (d′) by 108 μrad, and in (e) and (e′) by 142 μrad. Small arrows are indicated to call attention to characteristic structures in patterns.

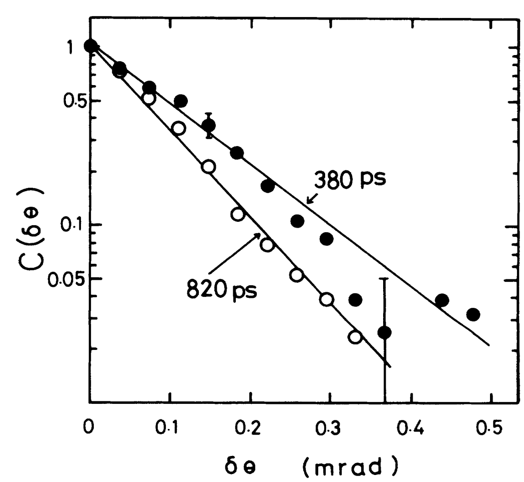

FIG. 4. Same plots of Fig. 3 in a semilog scale.

on the diffusion constant determined in the pulse transmission experiment and the experimentally determined correlation curves from time-resolved speckles (Fig. 3). The ratio of the FWHM of experimental curves at two different points in time closely agree with that predicted by theory. This suggests that transverse diffusion area of light at different points of time at the outgoing surface in the actual sample is proportional to that expected from a simple diffusion picture.

To summarize, we have shown that the speckle problems that have been experimentally studied only in the steady state in the past can be dealt with by the time-resolved approach. It has been made clear, theoretically and experimentally, that the memory effect is lost with time after the incident pulse enters the medium. Utilizing the concept of spatial-temporal speckles, other speckle correlations of interest[4] as well as a wide variety of application-oriented speckle measurements, such as the measurement of small displacement or oscillation, surface conditions, material fatigue on the order of optical wavelength, etc., are now under investigation by the time-resolved approach in our laboratory.[25]

The author is pleased to acknowledge useful discussion with T. Matsumoto and Professor H. Ikari. This work was supported by Matsuo Foundation and by Research Foundation for Opto-Science and Technology.

[1]M. P. van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985).
[2]P. E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).
[3]S. John, Comments Condens. Matter Phys. **14**, 193 (1988).
[4]S. Feng, C. Kane, P. A. Lee, and A. D. Stone, Phys. Rev. Lett. **61**, 834 (1988).
[5]I. Freund, M. Rosenbluh, and S. Feng, Phys. Rev. Lett. **61**, 2328 (1988).
[6]I. Freund, M. Rosenbluh, and R. Berkovits, Phys. Rev. B **39**, 12403 (1989).
[7]R. Berkovits, M. Kaveh, and S. Feng, Phys. Rev. B **40**, 737 (1989).
[8]R. Berkovits and M. Kaveh, Phys. Rev. B **41**, 2635 (1990).
[9]M. P. van Albada, J. F. de Boer, and A. Lagendijk, Phys. Rev. Lett. **64**, 2787 (1990).
[10]A. Z. Genack, N. Garica, and W. Polkosnik, Phys. Rev. Lett. **65**, 2129 (1990).
[11]N. Garica and A. Z. Genack, Phys. Rev. Lett. **66**, 1850 (1991).
[12]A. Z. Genack and N. Garica, Phys. Rev. Lett. **66**, 2064 (1991).
[13]*Laser Speckles and Related Phenomena*, edited by J. C. Dainty (Springer-Verlag, Berlin, 1984).
[14]M. Tomita and H. Ikari, Phys. Rev. B **43**, 3716 (1991).
[15]M. Tomita and M. Matsuoka, Phys. Rev. B **43**, 13579 (1991); *Ultrafast Phenomena VII* (Springer-Verlag, Berlin, 1990), p. 151.
[16]B. J. Berne and R. Pecora, *Dynamic Light Scattering* (Wiley, New York, 1967).
[17]G. Maret and P. E. Wolf, Z. Phys. B **65**, 409 (1987).
[18]M. Rosenbluh, M. Hoshen, I. Freund, and M. Kaveh, Phys. Rev. Lett. **58**, 2754 (1987).
[19]D. J. Pine, D. A. Weitz, P. M. Chaikin, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).
[20]F. C. Mackintosh and S. John, Phys. Rev. B **40**, 2383 (1989).
[21]R. Vreeker, M. P. van Albada, R. Sprik, and A. Lagendijk, Phys. Lett. **132**, 51 (1988).
[22]J. M. Drake and A. Z. Genack, Phys. Rev. Lett. **63**, 259 (1989).
[23]I. Freund and R. Berkovits, Phys. Rev. B **41**, 496 (1990).
[24]A. Lagendijk, R. Vreeker, and P. Devries, Phys. Lett. **136**, 81 (1989).
[25]M. Tomita (unpublished).