Temporal Response of a Random Medium from Third-Order Laser Speckle Frequency Correlations

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(Received 25 June 2001; published 2 January 2002)

We demonstrate for the first time that the temporal response of a random medium can be obtained from optical intensity fluctuations. Our method uses third-order intensity correlations of measured speckle patterns from a multiple scattering random medium as a function of optical frequency. In particular, our experimental results for the temporal response extracted from third-order intensity correlations are in good agreement with the predictions of a diffusion model. Our results are valid for waves in random media where the scattered field is described by circular complex Gaussian statistics.

Coherent waves propagating in a random medium lead to fluctuations in the measured intensity due to interference of the multiply scattered waves. The coherent waves can be classical waves such as electromagnetic or acoustic waves, or quantum waves such as those associated with electrons in a mesoscopic system where inelastic scattering does not dominate. The former gives rise to observed speckle patterns in the intensity [1], whereas the latter leads to the universal conductance fluctuations seen in mesoscopic samples of disordered metals [2]. For optical speckle patterns, it is possible to directly measure the correlation between intensities at two different illumination frequencies, or between speckle patterns with different incident and scattered wave vectors. These correlations provide insight into the scattering properties of the random medium [3].

We demonstrate for the first time that third-order intensity correlations provide the temporal response of a random medium. A key point is that the extra degree of freedom involved in third-order intensity correlations provides important new information not present in second-order intensity correlations. The correlations are performed over speckle pattern intensities measured at different frequencies, which not only gives the Fourier magnitude of the temporal response (which can be obtained from second-order correlations) but also the Fourier phase (which cannot be obtained by second-order correlations). Then, through the application of an inverse Fourier transform, the temporal response can be obtained. In particular, we investigate an optical multiple scattering random medium that is in the weak scattering limit where the transport mean free path is much greater than the wavelength, and in the diffusive regime where the sample size is much greater than the transport mean free path. This is an important class of scattering media, with biological tissue in the near-IR being a prime example.

Previous investigations into intensity correlations (see Refs. [3–7] for a review) have focused on second-order intensity correlations, highlighting the contribution of short-range, long-range, and infinite-range terms. The long- and infinite-range correlations were found to be inversely proportional to powers of the dimensionless conductance, g [8]. For optical experiments involving a slab geometry, $g \gg 1$ is typical [9], thus making the contribution of the long- and infinite-range correlations negligible. This is the assumption we make for our experiments, which is supported by the data.

We show experimentally the remarkable result that the temporal response of a random medium can be obtained using third-order correlations of speckle patterns in the frequency domain, for the speckle field described by circular complex Gaussian statistics. Third-order correlations have been investigated in the past for several different applications. Gamo [10,11] proposed using third-order correlations to extend the information obtainable from the intensity interferometer of Hanbury Brown and Twiss [12] for the analysis of optical spectrum profiles. Lohmann, Weigelt, and Wirnitzer [13] applied a third-order correlation in the speckle masking technique for astronomical applications, which provided for stellar imaging. In a deterministic application, Blount and Klauder [14] proposed a third-order correlation technique for determining the intensity pulse shape from a short-pulse laser. They established that third-order correlations are sufficient to fully characterize the intensity pulse shape.

The temporal response we investigate is the ensemble averaged normalized Green's function (impulse response function) for intensity of the scattering medium at some specified source-to-detector separation, which we denote by the real function $p(t)$. It can also be physically described as the ensemble averaged photon time of flight distribution through the random medium for an ultrashort optical intensity pulse as the input. This distribution provides an important measure of scatter, and as a function of position provides data which can be used in, for example, biological tissue imaging applications [15–17]. Direct measurement of $p(t)$ typically requires a substantial investment in instrumentation, due to the simultaneous

requirements of high speed and high sensitivity for detecting multiply scattered short optical pulses.

Genack [18] and Genack and Drake [19] have shown that the correlation of two speckle *fields* at different frequencies from a scattering medium is

$$\langle E(\nu_0)E^*(\nu_0 + \Delta\nu)\rangle = \langle I\rangle P(\Delta\nu), \qquad (1)$$

where $P(\Delta\nu)$ is the Fourier transform of $p(t)$, $E(\nu_0)$ is the speckle field complex amplitude, and the brackets $\langle\cdots\rangle$ represent the ensemble average over all possible scatterer configurations. The optical center frequency is given by ν_0, and $\Delta\nu$ represents a small shift frequency, typically in the gigahertz range. The mean intensity, $\langle I\rangle$, is assumed independent of $\Delta\nu$. This result is valid under the assumption of the field $E(\nu_0)$ being considered a random phasor sum of many scattered fields, with the phase of the scattered fields uniformly distributed over modulo 2π [18]. The field correlation expression of Eq. (1) is valid regardless of any long-range intensity correlation effects present in the random medium [18,20].

Neglecting the long-range correlation effects implies that all the scattered fields in the random phasor sum are assumed statistically independent, and thus the resultant speckle fields obey circular complex Gaussian statistics, as described by Goodman [21]. Therefore, the Gaussian moment theorem of Reed [22] can be used to evaluate intensity correlation expressions. Defining a normalized intensity $\tilde{I} = (I - \langle I\rangle)/\langle I\rangle$, and using the expression of Eq. (1), the second-order intensity correlation becomes

$$\langle \tilde{I}(\nu_0)\tilde{I}(\nu_0 + \Delta\nu)\rangle = |P(\Delta\nu)|^2. \qquad (2)$$

The Fourier phase information is lost, and it is not possible to reconstruct the temporal response from second-order intensity correlations without *a priori* information about the form of the temporal response. The measured data are normally used to fit the temporal response derived from a diffusion model [19,23], which we also investigated in a previous study [24].

Third-order intensity correlations, unlike second-order intensity correlations, contain information about the Fourier phase. Consider the intensities at three different frequencies given by $I_1 = I(\nu_0)$, $I_2 = I(\nu_0 + \Delta\nu_1)$, and $I_3 = I(\nu_0 + \Delta\nu_1 + \Delta\nu_2)$, with $\Delta\nu_1, \Delta\nu_2 \ll \nu_0$. The third-order correlation for the normalized intensities is then [22]

$$\langle \tilde{I}_1\tilde{I}_2\tilde{I}_3\rangle = 2\,\text{Re}\{P(\Delta\nu_1)P(\Delta\nu_2)P^*(\Delta\nu_1 + \Delta\nu_2)\}, \quad (3)$$

which is the important result from which information on the Fourier phase can be obtained. Consider the third-order correlation of $p(t)$, given by

$$g^{(3)}(\tau_1, \tau_2) = \int_{-\infty}^{\infty} dt\, p(t)p(t + \tau_1)p(t + \tau_2). \qquad (4)$$

The bispectrum [25] of $p(t)$ is defined as the Fourier transform of the third-order temporal correlation (with respect

to τ_1 and τ_2) and is given by

$$G^{(3)}(\Delta\nu_1, \Delta\nu_2) = P(\Delta\nu_1)P(\Delta\nu_2)P^*(\Delta\nu_1 + \Delta\nu_2). \qquad (5)$$

Hence, the third-order speckle intensity correlation of Eq. (3) is twice the real part of the bispectrum of $p(t)$.

Denoting the phase of Eq. (5) as $\psi(\Delta\nu_1, \Delta\nu_2)$ (the bispectral phase) and the phase of $P(\Delta\nu)$ as $\phi(\Delta\nu)$ (the Fourier phase), the bispectral phase and Fourier phase are then related by

$$\psi(\Delta\nu_1, \Delta\nu_2) = \phi(\Delta\nu_1) + \phi(\Delta\nu_2) - \phi(\Delta\nu_1 + \Delta\nu_2). \qquad (6)$$

The Fourier phase can be reconstructed from this expression when the bispectral phase is known over a range of frequencies by discretizing Eq. (6) and solving the resultant recursion expressions explicitly [26]. The bispectrum is invariant to the linear Fourier phase [25], thus allowing an arbitrary linear Fourier phase, which results in an arbitrary time delay in the reconstructed temporal response $p(t)$.

Note that, while the Fourier phase can be obtained by knowing the full bispectral phase, the third-order intensity correlation in Eq. (3) yields only the cosine of the bispectral phase. Thus, there is a sign ambiguity in the experimentally determined bispectral phase, resulting in a time reversal indeterminacy.

We used the experimental setup shown in Fig. 1 to obtain data for the third-order speckle correlation measurements. The output of a single mode, narrow linewidth (nominally 5 MHz) external-cavity tunable laser diode with a center wavelength of $\lambda = 850$ nm was focused onto the front face of the sample. The center frequency of the laser could be scanned over a range of 60 GHz, thus allowing $\Delta\nu$ of Eq. (1) to vary over this range. A small 1 mm $\times$ 0.8 mm area on the back face of the sample was imaged onto a CCD camera using a lens and aperture. A linear polarizer ensured that we imaged only the scattered light which had the same linear polarization as the input light. The scattering samples used were commercial white acrylics (Cyro Industries, Acrylite FF) with the scattering due to small TiO_2 (of average diameter approximately 50 nm) particles within the acrylic background. We investigated two sample thicknesses of 6 and 12 mm. A typical speckle image is shown in Fig. 2, along with insets showing the speckle pattern decorrelate as $\Delta\nu$ is increased.
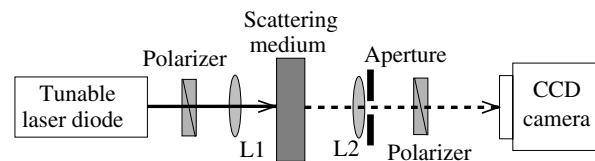

FIG. 1. The experimental setup used for measuring the speckle intensity patterns from a scattering medium. A series of intensity speckle patterns were obtained as the laser diode center frequency was scanned.
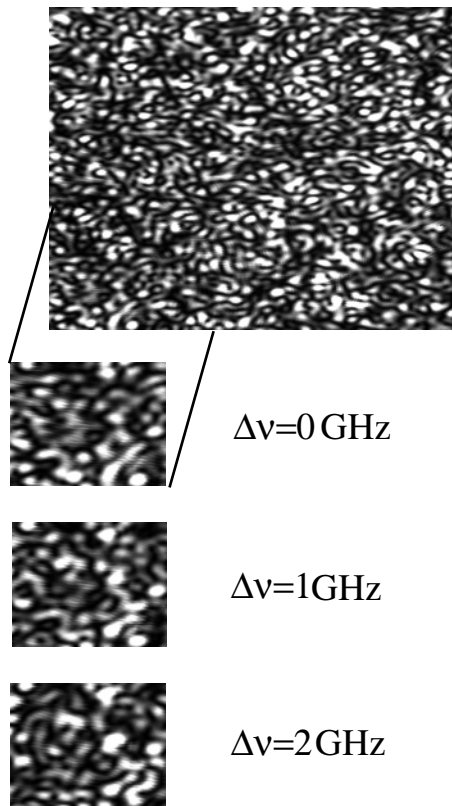
FIG. 2.   A typical measured speckle pattern. The insets show an expanded image of the lower left sixteenth of the speckle pattern, $I(\nu_0 + \Delta\nu)$, as $\Delta\nu$ is varied. This shows the speckle pattern slowly decorrelate as $\Delta\nu$ is increased for the $d = 6$ mm sample thickness.

We measured speckle patterns at 25 discrete center frequencies of the laser diode in increments of 1 GHz. Based on fixed frequency correlation measurements, we verified that the scattering sample and laser diode were stable over the duration of the experiment, and thus did not contribute anomalous decorrelation due to drift.

We first calculated the probability density function for intensity from the measured speckle data (first-order statistics). The negative exponential behavior expected from a speckle field with circular complex Gaussian statistics was obtained. The second-order intensity correlation was also measured and is shown in Fig. 3(a), and the rapid decay towards zero indicates that it is free of any long-range correlation effects. This gives evidence for a large g value [27], which in turn supports our assumption that we have circular complex Gaussian statistics for the measured speckle field [28], required for the validity of Eq. (3). Figure 3(b) shows a plot of the third-order intensity correlation of Eq. (3) from the measured speckle data. The third-order correlation of Eq. (3) is symmetric with respect to interchanging $\Delta\nu_1$ and $\Delta\nu_2$, thus only half of the data presented in Fig. 3(b) is unique. In fact, the general bispectrum expression for $G^{(3)}(\Delta\nu_1, \Delta\nu_2)$ in Eq. (5) contains only one octant of unique data [25]. The remaining data may be averaged with the unique octant to reduce noise.
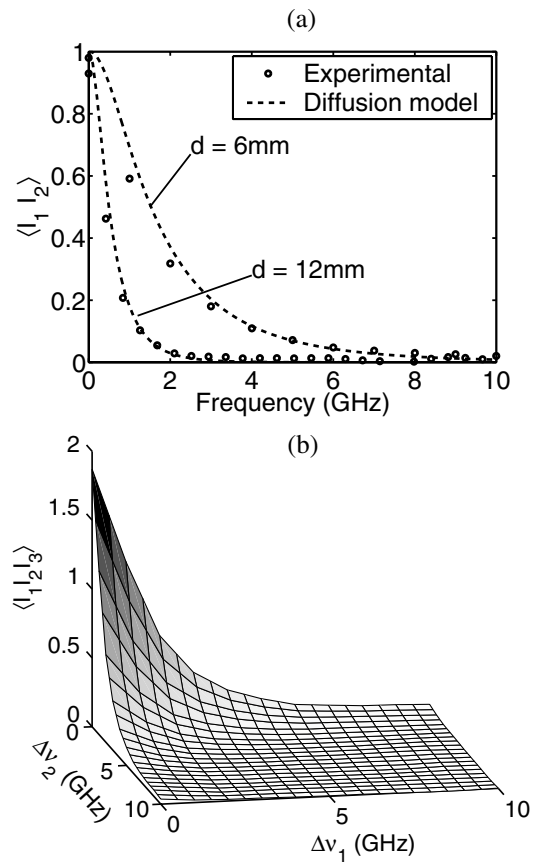

FIG. 3.   (a) The measured second-order intensity correlation described by Eq. (2) for two slab thicknesses. The symbols are the measured data, and the broken lines are the results from a diffusion model for $P(\Delta\nu)$. These results show the absence of any long-range correlation effects. (b) The measured third-order intensity correlation described by Eq. (3) for the $d = 6$ mm slab thickness.

In order to demonstrate reconstruction of the temporal response, we computed the Fourier magnitude from Eq. (2) and the Fourier phase by solving Eq. (6), then we performed an inverse fast Fourier transform to obtain the temporal response. The results are shown in Fig. 4 and compared with the temporal response of a diffusive medium calculated using $\mu_s' = 15$ cm^{-1} and $\mu_a = 0$ cm^{-1} for a uniform slab using image theory [29]. As can be seen, the measured results are in good agreement with the analytic diffusion model for a slab and, in particular, reproduce very well the large change in $p(t)$ as a function of sample thickness. The arbitrary linear phase in the solution of Eq. (6) was chosen to overlap the reconstructed and analytic temporal responses. The oscillations are due to errors in the reconstructed Fourier magnitude and phase at high frequencies, where the intensity correlations are small and greatly affected by measurement noise. Nevertheless, the results clearly show the significant accomplishment of determining the temporal response of a random scattering medium using third-order frequency domain intensity correlations.
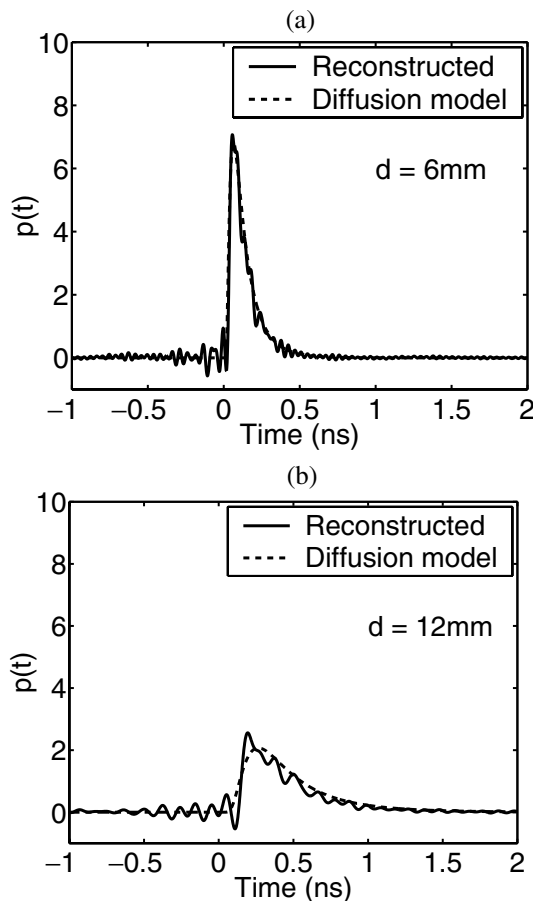
FIG. 4. Reconstructed temporal responses from the third-order intensity correlation measurements. The solid line is the reconstruction from measured data and the broken line is an analytic result from a diffusion model. The temporal response for a single slab of thickness 6 mm is given in (a), and for two identical slabs of 6 mm is given in (b), both showing good agreement with a diffusion model.

In conclusion, we have demonstrated a new approach for measuring the temporal response of a random scattering medium up to an arbitrary time offset using third-order intensity correlations in frequency. Our technique does not require any *a priori* information regarding the form of the temporal response and requires only that the speckle field be described by circular complex Gaussian statistics.

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