Nonstationary time-domain statistics of multiply scattered broadband terahertz pulses

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We examine the time-domain statistics of randomly varying electric fields generated by multiple scattering of single-cycle electromagnetic pulses in a random medium. This analysis emphasizes the fact that these measured random fields are not stationary, as is commonly assumed for diffusing photons generated with a narrowband light source. We demonstrate that the nonstationarity is a consequence of the time dependence of the configurationally averaged intensity, and that the statistical properties of the random field can be predicted if this quantity is known. We also discuss an approach for describing the transition from nonstationary to stationary behavior by investigating the degree of stationarity during a short time window. A parameterization of the statistics using a gamma distribution provides a quantifiable measure of the approach to stationarity. Our predictions are in good agreement with experimental observations. © 2006 Optical Society of America

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The ubiquity of multiply scattered photons has motivated the investigation of the statistical properties of electromagnetic waves that propagate through random media. Such studies have led to the development of a variety of techniques for imaging in the presence of multiple scattering.[1–6] Speckle correlation spectroscopy can be used to characterize the random medium itself.[7–9] Moreover, the statistics of the diffusive wave provide a key indicator of the onset of localization.[10,11] The majority of such studies have employed narrowband sources, either at optical or microwave frequencies. Recently, however, the case of broadband excitation has been of increasing interest.[12–18]

In this paper we examine the statistics of the electric field of multiply scattered broadband pulses in the time domain. Most previous studies have considered these statistics in the frequency domain, which is appropriate for the case of narrowband excitation.[19–21] A general assumption has been that the processes under investigation are stationary; that is, that the statistical properties are independent of the absolute time at which the measurement is performed. This assumption is implicit in the use of a frequency-domain analysis, since the basis functions in a Fourier decomposition have infinite duration. However, due to the broad bandwidth and the transient nature of the source in pulsed experiments, the statistics of the diffusive wave necessarily evolve over time and therefore are nonstationary.[14] Because pulsed techniques are widely employed for the study of multiple scattering,[12,14,16,22–24] we are motivated to consider the implications of nonstationary scattering processes by examining the evolution of the statistics of the field and intensity with time. We find that the nonstationary nature of the statistics are a consequence of the time dependence of the photon time-of-flight (TOF) distribution, and that the statistical properties of the random field can be predicted if this quantity is known. Because the TOF distribution can in certain cases be computed directly, this result implies that it should be possible to compute the statistics in an *a priori* fashion.

Our experimental setup has been described previously.[12] We use terahertz time-domain spectroscopy to generate single-cycle pulses of terahertz radiation, to direct these pulses at a random medium, and to detect the scattered electric field with subcycle temporal resolution. These pulses are approximately 1 ps in duration, with spectral content ranging from 50 GHz to 1 THz. The medium consists of a dense collection of randomly positioned Teflon spheres of 0.794 mm in diameter, held in a $(4\ \mathrm{cm})^3$ Teflon box at a volume fraction of 0.56 ± 0.04. In these samples, the mean free path of the radiation varies dramatically within the bandwidth of the terahertz pulse by a factor of ~ 70.[25] We measure the scattered electric field $E_{sc}(t)$ at 90° to the incident beam for numerous different manifestations of the disorder. In our earlier work, we developed a phenomenological model for understanding the departures from Gaussian statistics that are a consequence of the broad spectral bandwidth.[12] This model was implemented in the frequency domain using the spectral components $E_{sc}(\omega)$ obtained from the measured time-domain waveforms by Fourier analysis.

Here, we consider the statistics of the electric field in the time domain $E_{sc}(t)$. Because the sample is excited with a short pulse, the statistics of the scattered field are nonstationary. As a result, it is necessary to consider the changing distribution of the scattered electric field of the diffusive wave with time. When the incident pulse enters the random medium, it can scatter many times before exiting. At a position far from the sample and at 90° to the incident beam, the measured field consists of the superposition of many scattered wavelets. For a particular time delay τ and configuration of scatterers, the measured field can be described as the summation of a large number of independent fields with random phases and amplitudes. According to the central limit theorem, the sum of a large number of independent identically distributed random

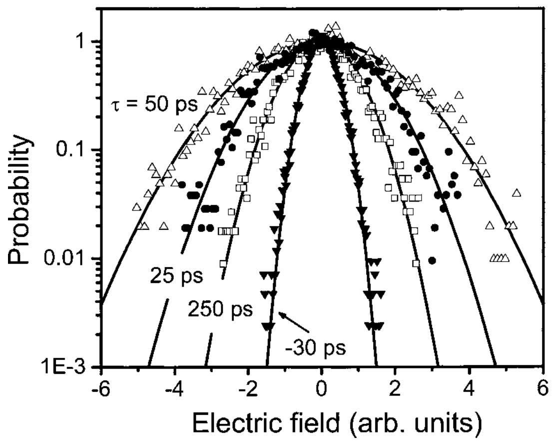

Fig. 1. Probability distribution of the electric field at several different time delays (as labeled), each normalized to unity at its peak. The solid curves are zero-mean Gaussians, computed with $\sigma^2(\tau)=\langle I_{sc}(\tau)\rangle$, the average intensity. The variance $\sigma^2(\tau)$ depends on the time delay τ, demonstrating that this statistical process is nonstationary. The $\tau=-30$ ps result corresponds to a delay when no photons have yet reached the detector. This characterizes the measurement noise, which is also Gaussian distributed.

variables is Gaussian distributed. Thus, the distribution for the scattered electric field $E_{sc}(\tau)$ can be expressed as

$$P(E|\tau)=\frac{1}{\sqrt{2\pi\langle I_{sc}(\tau)\rangle}}\exp\left[\frac{-E^2}{2\langle I_{sc}(\tau)\rangle}\right], \tag{1}$$

where the variance of the Gaussian is equal to the configurationally averaged intensity $\langle I_{sc}(\tau)\rangle$; here, the angled brackets denote averaging over all the manifestations of the disorder. This result is analogous to the random phasor sum model used to describe the probability distribution of the complex parts of the scattered field in the frequency domain.[12,19,21,26]

Figure 1 shows the measured probability distribution of $E_{sc}(\tau)$ at several different time delays. Each distribution is obtained by measuring $E_{sc}(\tau)$ at a particular time delay τ for numerous different configurations of the random medium, and computing a histogram of the acquired values. Here and throughout this paper we define the zero of the time-delay axis by the delay at which the first measurable signal is detected emerging from the random medium. At all measured delays, the data are Gaussian distributed with zero mean and with variance $\sigma^2(\tau)=\langle I_{sc}(\tau)\rangle$, as described by Eq. (1).

Because of its quadratic relationship to $E_{sc}(\tau)$ and the Gaussian statistics of $E_{sc}(\tau)$, the intensity $I_{sc}(\tau)$ must follow an exponential distribution.[19] The exponential distribution is a special case of the more general gamma distribution[18] given by

$$P(I|\tau)=\frac{I^{\alpha(\tau)-1}\exp[-I/\beta(\tau)]}{\beta(\tau)^{\alpha(\tau)}\Gamma[\alpha(\tau)]}, \tag{2}$$

where $\alpha(\tau)=1$ and $\beta(\tau)=\langle I_{sc}(\tau)\rangle$.

The choice of this particular generalization of the exponential distribution is a motivated one. It is relatively straightforward to demonstrate that the probability distribution function of the integrated intensity for a polarized thermal light source is described by Eq. (2). In that case, the parameter α has a physical interpretation: It is the number of coherence intervals that influence any one particular measurement of the intensity. This quantity can never be less than unity, because a measurement always samples at least one coherence interval, no matter how short the integration time.[19] However, these conclusions hinge on the assumption that the process is stationary. In the case of a nonstationary process, the definition of the coherence interval becomes unclear, because the coherence function depends not merely on the time difference between two measurements t_2-t_1, but on the individual times t_1 and t_2. In this case, any statistical property can depend on the particular observation time τ at which a measurement is made. The results of Fig. 1 clearly show that the stationary assumption does not apply to pulsed experiments such as the ones described here. So it is necessary to further explore the statistics of nonstationary processes.

The nonstationary nature of the diffusive waves is reflected in the dependence of the probability distributions $P(E|\tau)$ and $P(I|\tau)$ on time delay τ. Because the variance of $P(E|\tau)$ is proportional to the configurationally averaged intensity $\langle I_{sc}(\tau)\rangle$, this quantity is key to describing the statistics of the field and the intensity. Figure 2 shows the estimate of $\langle I_{sc}(\tau)\rangle$. This is determined by first taking the Hilbert transform of the measured time-domain waveforms to obtain the complex field envelopes, and then averaging the squared magnitude of these over all configurations of the random medium. Clearly, $\langle I_{sc}(\tau)\rangle$ varies substantially within the measured time window. Because the input pulses are extremely short (~1 ps), this quantity is essentially the impulse response of the random medium. As a result, $\langle I_{sc}(\tau)\rangle$ is proportional to the photon TOF distribution, the probability of a photon arriving at time τ.[26,27] In certain geometries, such as when the mean free path is much smaller than the sample thickness, it is possible to predict the TOF distribution using the diffusion equation.[9,23] Because in our case the source has a large bandwidth and the mean free path varies substantially within this bandwidth, it is not possible to apply a simple diffusion model to predict $\langle I_{sc}(\tau)\rangle$ for our

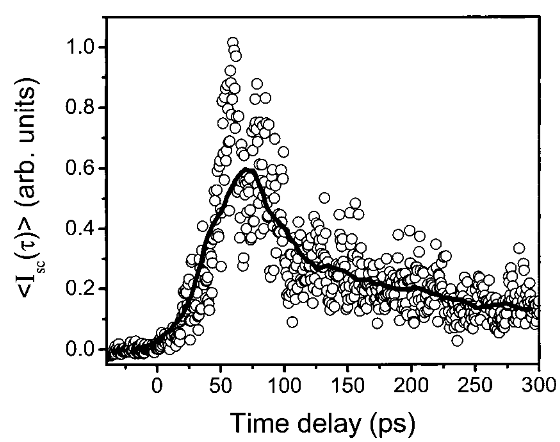

Fig. 2. Configurationally averaged intensity $\langle I_{sc}(\tau)\rangle$, proportional to the photon TOF distribution. This is computed by averaging the measured intensities over all configurations of the random medium. The solid curve shows the result of adjacent averaging of $\langle I_{sc}(\tau)\rangle$ using a sliding 50 ps time window.

situation. Nevertheless, the variation of $\langle I_{sc}(\tau)\rangle$ with time delay shown in Fig. 2 is reminiscent of the $t^{-3/2}\exp[-\text{constant}/t]$ behavior expected for diffusive propagation.[23]

As noted above, at any particular time delay τ, the probability distribution for the intensity is exponential with a rate given by the configurationally averaged temporal intensity, $\langle I_{sc}(\tau)\rangle$. To determine the distribution of the intensity for all delays within the measurement window, we integrate Eq. (2) over τ [using $\alpha(\tau)=1$] and normalize by the time window:

$$p(I)=\frac{1}{\Delta t}\int_{t_1}^{t_2}\frac{\exp[-I/\langle I_{sc}(\tau)\rangle]}{\langle I_{sc}(\tau)\rangle}\mathrm{d}\tau. \tag{3}$$

Equation (3) can be interpreted as a superposition of a large number of exponential distributions with a mean and standard deviation that is controlled by the TOF distribution. We note that this expression is valid even in the case of a band-limited field that exhibits temporal correlations. These correlations are embodied in the dependence of $\langle I_{sc}(\tau)\rangle$ on τ. This can be seen from the fact that, in the extreme case of a perfectly correlated intensity such that $\langle I_{sc}(\tau)\rangle \to I_0$, Eq. (3) reduces to $P(I)=\exp[-I/I_0]$, which is the expected result. The probability distribution of the intensity of the diffusive waves, shown in Fig. 3, is clearly nonexponential. As above, these are computed from the complex field envelopes, obtained via the Hilbert transform. The solid curve is computed from Eq. (3) using the data in Fig. 2 for $\langle I_{sc}(\tau)\rangle$. The result is in excellent agreement with the experimental data.

In a similar fashion, the probability distribution of the real electric field over a finite time window can be expressed as

$$P(E)=\frac{1}{\Delta t}\int_{t_1}^{t_2}\frac{1}{\sqrt{2\pi\langle I_{sc}(\tau)\rangle}}\exp\left[\frac{-E^2}{2\langle I_{sc}(\tau)\rangle}\right]\mathrm{d}\tau. \tag{4}$$

As above, this can be interpreted as a superposition of Gaussians, with variance determined by the TOF distribution. Figure 4 shows the comparison between the predictions of Eq. (4) and the experimentally determined statistics, again showing that this non-Gaussian distribution can be modeled using only $\langle I_{sc}(\tau)\rangle$. We emphasize the crucial result demonstrated here, which is that the statistics of the field and the intensity depend solely on the photon TOF distribution.

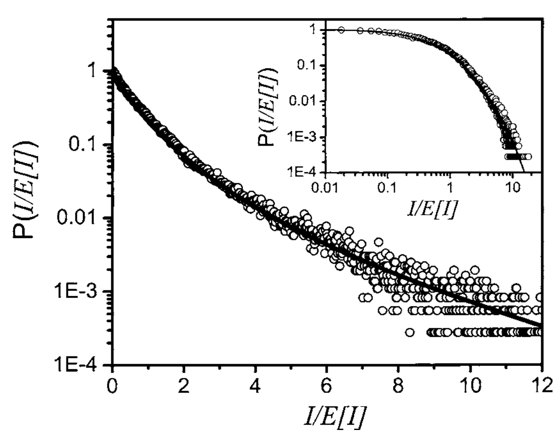

Fig. 3. Probability distribution of $I/E[I]$, the intensity normalized by the mean intensity. The plot shows the data and the prediction computed using Eq. (3). The inset shows the same result on a log–log scale.

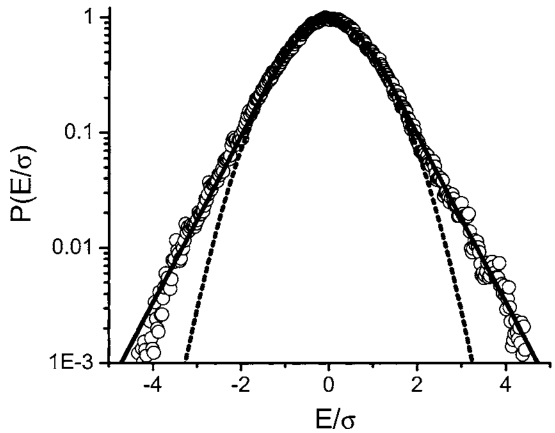

Fig. 4. Probability distribution of the (real) electric field normalized by its standard deviation. The plot shows the data (circles) and the prediction computed from Eq. (4). The dashed curve shows a Gaussian, which does not fit the wings of the distribution.

The introduction of the gamma distribution [Eq. (2)] suggests that one can quantify the degree of stationarity using the parameters of this distribution. If the statistics of a particular set of measured data are stationary, then the probability distribution of the intensities should follow an exponential distribution, that is, a gamma distribution with $\alpha(\tau)=1$. Departures of α from unity, therefore, are a measure of the nonstationary nature of the process. We can test this notion using a narrow time window, short enough so that $\langle I_{sc}(\tau)\rangle$ can be assumed to be nearly constant. The idea of time-windowed processing of nonstationary signals has been used previously to extract time-shifted field correlations.[28] To test this idea here, we extract from our measured waveforms the probability distribution of the intensities within a 50 ps time window centered at τ and determine the α parameter by fitting to a gamma distribution [Eq. (2)] using a least-squares curve-fitting routine with the scale factor β held constant. We can estimate the value of β at each value of τ by smoothing $\langle I_{sc}(\tau)\rangle$ with a 50 ps window, as shown by the solid curve in Fig. 2. The deviation of $\alpha(\tau)$ from unity is a measure of the extent to which the statistics are nonstationary within a 50 ps window centered at delay τ. Figure 5 shows the extracted values for $\alpha(\tau)$ as a function of the position of the center of the time window τ, with error bars denoting the 95% confidence intervals. We find that $\alpha(\tau)$ is significantly smaller than 1 at early times when $\langle I_{sc}(\tau)\rangle$ varies rapidly, but converges to unity at later times when $\langle I_{sc}(\tau)\rangle$ varies more slowly. When there is less variation in $\langle I_{sc}(\tau)\rangle$ within the time window, the assumption of a constant $\beta(\tau)$ is more valid and therefore the statistics can be considered nearly stationary within the 50 ps window. This analysis offers a new approach to understanding the transition from nonstationary to stationary behavior in time-domain statistics.

In conclusion, we have addressed the time-domain statistics of diffusively propagating waves. As a result of the transient nature of the source, stationarity is no longer a

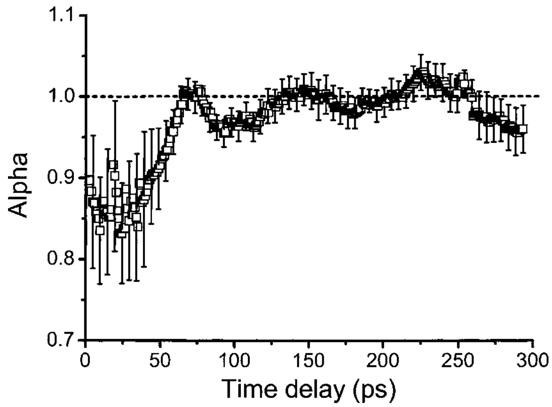

Fig. 5. $\alpha(\tau)$ parameter of a gamma distribution [Eq. (2)] extracted using the fitting procedure described in the text, as a function of the position of the center of the 50 ps time window τ. In fitting to the gamma distribution, $\beta(\tau)$ is set equal to the smoothed average intensity $\langle I_{sc}(\tau)\rangle$, the solid curve in Fig. 2. The error bars denote 95% confidence intervals.

valid assumption and the statistics must be expected to evolve with time. We examine the probability distribution of the electric field and the intensity at particular time delays. The statistics are found to depend only on the photon TOF distribution $\langle I_{sc}(\tau)\rangle$, which under certain circumstances can be accurately predicted using the diffusion equation, and which is usually more easily measured than the scattered electric field.[26] Using $\langle I_{sc}(\tau)\rangle$, we can accurately model the probability distribution of both the field and the intensity. These results have important implications for optical pulsed experiments, where $\langle I_{sc}(\tau)\rangle$ is measurable but the electric field is generally not accessible.[22–24] In these situations, it is still possible to determine the probability distributions for the temporal intensity and the field.

We also demonstrate a new approach to understanding the transition from nonstationary to stationary behavior, based on the parameterization of the intensity statistics using a gamma distribution. This analysis confirms the expected result that the statistics can be assumed to be stationary within a small time window, if $\langle I_{sc}(\tau)\rangle$ is nearly constant within that window. Here, we can point out an analogy with the situation encountered in many frequency-domain studies. In these previous measurements, the assumption of stationarity is implicit in the use of a narrowband (and therefore temporally invariant) source. In this case, stationarity can be assumed if the properties of the medium (such as the photon mean free path) do not vary within the narrow bandwidth of the source. Similarly, the time-domain statistics are stationary if one studies a narrow temporal window, during which the photon TOF distribution is approximately constant. Just as the definition of "narrowband enough" may depend on the central frequency of the spectral band in question, we also find that the definition of "short enough" depends on the position of the narrow temporal window, relative to the peak of $\langle I_{sc}(\tau)\rangle$. For our experimental situation, a 50 ps window is short enough, except at early times when the TOF distribution is rapidly varying.

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