Statistics of Multiply Scattered Broadband Terahertz Pulses

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We describe the first measurements of the diffusion of broadband single-cycle optical pulses through a highly scattering medium. Using terahertz time-domain spectroscopy, we measure the electric field of a multiply scattered wave with a time resolution shorter than one optical cycle. This time-domain measurement provides information on the statistics of both the amplitude and phase distributions of the diffusive wave. We develop a theoretical description, suitable for broadband radiation, which adequately describes the experimental results.

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The propagation of classical waves in the presence of random scattering is a topic of considerable interest in many research communities. In particular, scattering of electromagnetic waves can lead to a rich array of phenomena [1,2]. In a random medium, the propagating field can be described as a superposition of unscattered and scattered waves. Diffusive propagation occurs after the incident wave travels a distance much larger than the Boltzmann transport mean free path (l_{tr}) [1]. In this case, the incident light beam is completely randomized, and only multiply scattered photons are transported through the medium. This diffusive wave is of much interest because it can be used for locating and imaging objects buried in the random medium [3–5]. In addition, the statistics of the diffusive wave can be used to extract information on the nature of the random medium [6] and are a key indicator of the onset of localization [7]. Much of the research on diffusive optical waves has concentrated on the case of monochromatic or narrow-band waves [3,8–14]. Several authors have used short optical pulses as a means for separating the diffusive portion of the wave from the ballistic light [7,15–18]. Others have used low-coherence interferometry to extract relative phase information [19–23]. Short acoustic pulse propagation is also extensively studied in the context of seismic tomography [24]. However, the treatment of the statistics of broadband fields in random media remains largely unaddressed.

Here, we report measurements of the electric field of multiply scattered broadband optical pulses. We compute the statistics of these random fields and demonstrate the connections to the case of monochromatic radiation. These measurements employ terahertz time-domain spectroscopy (THz-TDS), in a configuration quite similar to the one described previously [25–27]. Using this technique, it is possible to generate pulses with a fractional bandwidth in excess of 100% (50 GHz–1 THz). Furthermore, the coherent measurement of the electric field permits extraction of both the amplitude and phase of the field, with a temporal resolution better than one optical cycle, without the use of interferometric techniques. As a result, one directly observes, among other things, the distribution of photon transit times (i.e., the path-length distribution function). This quantity usually must be extracted from time-integrated measurements using spatial intensity correlations [14]. Finally, we emphasize that these measurements have been performed in a three-dimensional sample, rather than in the waveguide geometry customarily employed in microwave measurements [28].

The experimental setup is illustrated in Fig. 1. Single-cycle terahertz pulses are focused into a random medium, and the emerging radiation is measured at an angle of 90° with respect to the incident beam direction. This setup ensures that no ballistic radiation reaches the detector. The model random medium consists of a large number of Teflon spheres with a diameter of 0.794 ± 0.025 mm. Teflon is an excellent material for these measurements because of its low absorption and because the refractive index of Teflon, $n = 1.4330$, is nearly independent of frequency throughout the spectral range of the measurements. The spheres are poured into a cubic Teflon cell with dimensions of $(4\ \mathrm{cm})^3$. The volume fraction of the spheres in the sample cell is measured to be 0.56 ± 0.04. Our

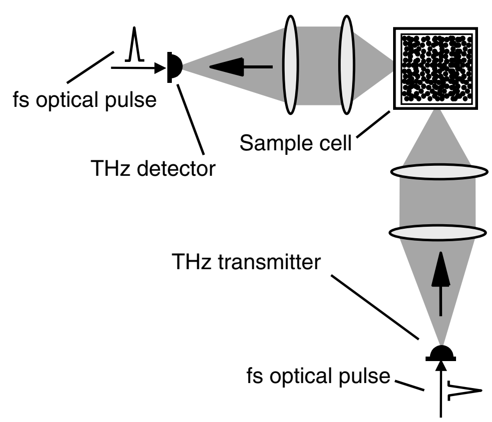

FIG. 1. A schematic of the diffusion experiment. The detector is situated at 90° to the input beam direction, in order to avoid measuring ballistic terahertz photons.

previous measurements indicate that the mean free path in these samples ranges from ~1 to ~70 mm within the bandwidth of the terahertz pulse [26,27].

Figure 2 shows several representative terahertz waveforms. Each waveform corresponds to a realization of a unique configuration of the random medium. These waveforms have been spectrally filtered at both low and high frequencies to improve the signal to noise, which is about 10:1 at the spectral peak. For reference, the signal to noise for a measurement of the incident single-cycle pulse exceeds 20 000 after equivalent spectral filtering. We measured waveforms for 22 different sample configurations. The result of the multiple scattering is a randomization of the phase, which produces the complex structure shown in Fig. 2. By taking the Fourier transform of these waveforms, we can extract both the real $r = \mathrm{Re}[E(\omega)]$ and the imaginary $i = \mathrm{Im}[E(\omega)]$ parts of the scattered electric field $E(\omega)$. From these measurements, we are able to obtain the probability distribution of the real and imaginary parts of the transmitted electric field, $P(r)$ and $P(i)$.

If we assume that the complex electric field component at a given frequency is the sum of a large number of random phasors, then the central limit theorem predicts that the scattered field should obey Gaussian statistics [28,29]. Assuming that the phase is uniformly distributed, the joint probability distribution of the real and imaginary parts at a given frequency ω can be considered zero mean, jointly Gaussian variables, and therefore

$$P(r, i|\omega) = \frac{1}{2\pi\sigma_\omega(\omega)^2}\exp\left[-\frac{r^2 + i^2}{2\sigma_\omega(\omega)^2}\right], \qquad (1)$$

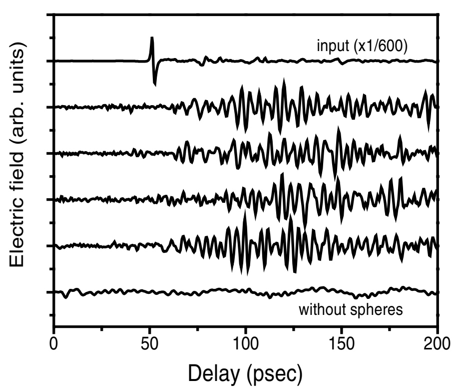

FIG. 2. Several typical terahertz waveforms measured using the setup shown in Fig. 1. Each waveform represents a unique configuration of the random medium. For reference, the upper curve shows the single-cycle pulse incident on the random medium. This curve has been scaled by a factor of 600 and is shown on a shifted time axis. The bottom curve shows a measurement with no scatterers in the sample cell, and is therefore an indication of the noise level. Only the first 200 psec of each waveform is shown, although the measurable signals extend beyond 600 psec.

where the variance $\sigma_\omega(\omega)^2 = \langle I(\omega)\rangle/2$. $\langle I(\omega)\rangle$ is the spectral intensity of the diffuse light averaged over all configurations of the medium, and it is dependent on the input pulse and the scattering parameters of the random medium. To determine the joint distribution of the real and imaginary parts within a finite frequency range $\Delta\omega = \omega_2 - \omega_1$, we integrate (1) over ω and normalize by the bandwidth $\Delta\omega$,

$$P(r, i) = \frac{1}{\pi\Delta\omega}\int_{\omega_1}^{\omega_2}\frac{1}{\langle I(\omega)\rangle}\exp\left[-\frac{r^2 + i^2}{\langle I(\omega)\rangle}\right]d\omega. \qquad (2)$$

This expression may be interpreted as the sum of a large number of zero-mean Gaussian distributions (one for each spectral component), each with a unique variance proportional to $\langle I(\omega)\rangle$. The marginals $P(r)$ and $P(i)$ are equivalent to each other and can be computed as

$$P(a) = \frac{1}{\Delta\omega}\int_{\omega_1}^{\omega_2}\frac{1}{\sqrt{\pi\langle I(\omega)\rangle}}\exp\left[-\frac{a^2}{\langle I(\omega)\rangle}\right]d\omega \qquad (3)$$

with $a = \{r, i\}$ and with variance $\sigma^2 = \frac{1}{2\Delta\omega}\int_{\omega_1}^{\omega_2}\langle I(\omega)\rangle d\omega$. The variance of $P(r, i)$ is proportional to the integrated average intensity of the diffuse light. This is analogous to diffuse monochromatic waves where the variance is proportional to the average intensity [28]. We extract the complex parts of $E(\omega)$ over the 50–500 GHz spectral range, where there is an appreciable signal in the measured waveforms. The probability distributions of the normalized real and imaginary parts $P(r/\sigma)$ and $P(i/\sigma)$ are shown in Fig. 3. The real and imaginary parts are zero mean and have nearly identical distributions as predicted by (2). As expected, the Gaussian distribution predicted for the case of monochromatic illumination [28] (dashed line) does not accurately fit the data. In order

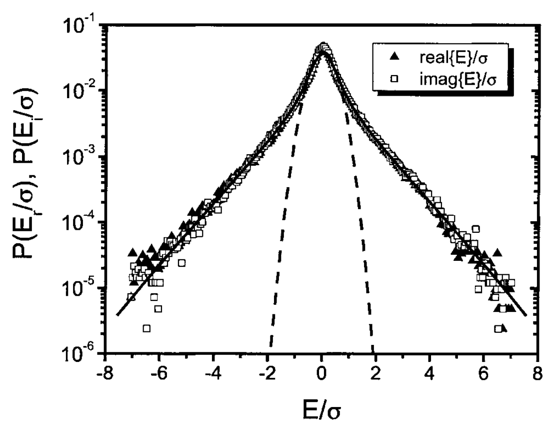

FIG. 3. The probability distribution of the normalized real (triangles) and imaginary (open squares) parts of the complex scattered electric field, $P(r/\sigma)$ and $P(i/\sigma)$, plotted on a log scale. The dashed line shows the Gaussian distribution, which is the result expected for monochromatic radiation [28]. The solid curve is the prediction of Eq. (3), using an experimentally determined estimated for the mean spectral intensity.

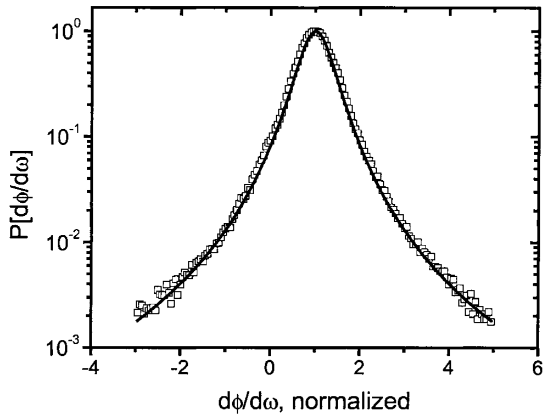

FIG. 4. The probability distribution of the normalized spectral phase derivative $\hat{\phi}'$, plotted on a log scale. The solid line is the probability distribution given in Eq. (4), equivalent to the monochromatic case [30], with $Q = 0.234$.

to compare to the predicted result [Eq. (3)], we extract an estimate of the average intensity $\langle \hat{I}(\omega) \rangle$ by averaging the frequency-dependent intensity spectrum over the 22 measured waveforms. By substituting $\langle \hat{I}(\omega) \rangle$ for the average intensity in (3), we can numerically calculate $P(a/\sigma)$. The result (solid line) is in excellent agreement with the experimental data.

The statistics of the phase derivative $d\phi/d\omega \equiv \phi'$ are also of great importance. In the case of narrow-band wave packets, the ensemble average of this quantity is inversely proportional to the transport velocity, so it can be interpreted as a time delay for photons in the medium. For broadband waves, its connection to the concept of a delay time is questionable, because of the randomization of the spectral phase. Nevertheless, it is instructive to investigate the statistics of ϕ', because of its relevance in the study of higher-order (i.e., C_2) correlations [30]. For narrow-band wave packets, the probability distribution for the normalized phase derivative has been derived within the Gaussian approximation as [12,30]

$$P\left(\hat{\phi}' \equiv \frac{\phi'}{\langle \phi' \rangle}\right) = \frac{1}{2} \frac{Q}{[Q + (\hat{\phi}' - 1)^2]^{3/2}}, \qquad (4)$$

where $\hat{\phi}'$ is the phase derivative normalized to its ensemble-averaged mean, and where Q is a parameter related to the absorption length and the sample geometry [31]. For broadband waves, $P(\hat{\phi}')$ can be derived by integrating [4] over frequency with an appropriate weighting function, as in Eqs. (2) and (3) above. However, because in our measurements the absorption length is approximately constant over the entire bandwidth of the radiation, Q should not vary much as a function of frequency. Since this is the only parameter, the distribution of the phase derivative for broadband waves should also be given by (4). Figure 4 shows the probability distribution for $\hat{\phi}'$, extracted from the Fourier transforms of the measured waveforms. The solid curve is the predicted result [Eq. (4)], with $Q = 0.234$. As anticipated, the theoretical expression derived for the monochromatic case can also accurately predict the statistics of the broadband wave packet.

In conclusion, we report the first use of terahertz time-domain spectroscopy in the study of diffuse waves. The direct measurement of the multiply scattered electric field allows for the computation of statistics for both amplitude and phase. We have extended the theoretical framework, developed for monochromatic waves, to the broadband case, and found excellent agreement with our measured results. Using these time-resolved measurement techniques, it should also be possible to extract information on the nature of specific scattering events within the random medium.

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