Spectral shifts as a signature of the onset of diffusion of broadband terahertz pulses

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We describe measurements of polarization dynamics as a probe of multiple scattering of photons in a random medium by use of single-cycle terahertz pulses. We measure the degree of polarization and correlate it directly with the single-scattering regime in the time domain. We also measure the evolution of the temporal phase of the radiation and show that the average spectral content depends on the state of polarization. In the case of broadband radiation, this effect can be used to distinguish photons that have been scattered a few times from those that are propagating diffusively. © 2004 Optical Society of America

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The polarization of multiply scattered light waves has been a topic of considerable interest recently. When the propagation of light in a random medium is considered, a natural length scale is l^*, the transport mean free path, which is the mean propagation distance required for randomizing the direction.[1] In a study of depolarization, Bicout *et al.*[2] introduced a second characteristic length ξ, which is the propagation distance for randomization of the polarization. In an isotropic random medium this depolarization length is generally comparable with or somewhat larger than l^*, indicating that more than one scattering event is required for randomizing the polarization.[2-5] As a result, it is not surprising that in time-resolved studies a delay in the emergence of cross-polarized radiation from a random medium illuminated with linearly polarized light has been observed. This delay is due to the larger average number of scattering events experienced by this component.[6-9] This result has inspired a number of new imaging schemes based on polarization gating or polarization contrast.[10-14]

We point out that the majority of these studies have employed narrowband radiation, for which the spectral dependence of the properties of the medium (e.g., the mean free path) can be neglected. Here we explore the opposite situation, that of extremely broadband light. We show that in this case the spectral content of the radiation emerging from the random medium is also related to the number of scattering events. This is an alternative measure of the loss of spatial coherence of broadband waves.

For these measurements we use terahertz time-domain spectroscopy, which permits the generation of single-cycle pulses of radiation with a spectral content spanning more than 1 order of magnitude in frequency (50 GHz–1 THz). The radiation can be detected coherently, so both the field amplitude and the temporal phase are measured with subcycle time resolution. As a result, we can experimentally determine the precise limits of the single-scattering regime in the time domain.

The experimental setup is similar to the one described in Ref. 15. Single-cycle terahertz pulses are focused at the edge of the sample scattering medium,

and the emerging radiation is measured at an angle of 90° to the incident pulse direction. The medium consists of a dense collection of Teflon spheres [refractive index $n = 1.4330$ (Ref. 16)] with a diameter of 0.794 ± 0.025 mm. The spheres are poured into a cubic Teflon cell with dimensions of $(4\ \text{cm})^3$ at a measured volume fraction of 0.56 ± 0.04. The incident radiation has almost a pure horizontal linear polarization, with a small vertical component[17] that is filtered out by a wire grid polarizer placed before the sample. The detector is placed behind a second polarizer to ensure that only the desired polarization is measured. The detector and the second polarizer are rotated by either 0° or 90° to measure the electric field $E_{\perp,\parallel}(t)$ of each polarization component separately. Great care is taken to ensure that the position of the detector does not change during this rotation, such that the temporal delay calibration is maintained. In these samples, the mean free path of the radiation varies dramatically within the bandwidth of the terahertz pulse, by a factor of ~ 70.[18] We measured $E_{\perp}(t)$ and $E_{\parallel}(t)$ for 33 different manifestations of the disorder.

Figure 1 shows the photon time-of-flight distributions for both polarizations $\langle I_{\parallel}(t) \rangle$ and $\langle I_{\perp}(t) \rangle$, as well as for the sum $\langle I_{\text{Total}}(t) \rangle$, averaged over all measured realizations. The perpendicular radiation exits the medium approximately 30 ps later than the parallel radiation. This delay results from the larger average number of scattering events experienced by the perpendicular radiation and is consistent with previous reports in which it was found that $\xi > l^*$.[6-9] We may also compute the time-resolved degree of polarization. As the measured fields $E_{\perp}(t)$ and $E_{\parallel}(t)$ are mutually incoherent, this quantity is given by[19]

$$P(t) = \frac{\langle I_{\parallel}(t) \rangle - \langle I_{\perp}(t) \rangle}{\langle I_{\parallel}(t) \rangle + \langle I_{\perp}(t) \rangle}. \quad (1)$$

For early times, before the arrival of any photons, $P(t)$ is undefined. After ~ 510 ps the polarization begins to decay, until it essentially vanishes after ~ 600 ps. At much later times, when $P(t) = 0$, the propagation is purely diffusive, as is shown in the inset of Fig. 1
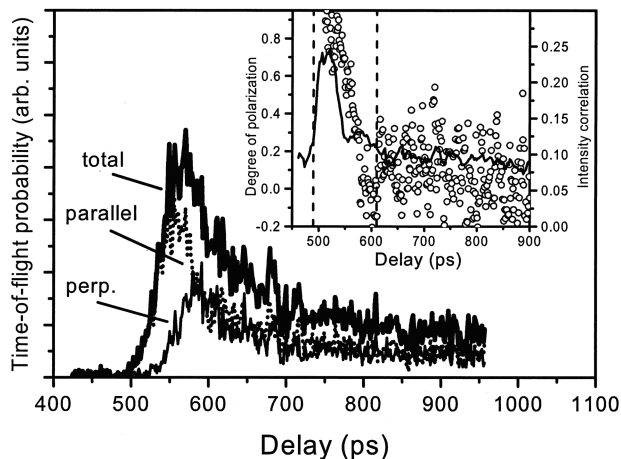
Fig. 1. Measured time-of-flight distributions of the parallel polarization $\langle I_{\parallel}(t) \rangle$, the perpendicular polarization $\langle I_{\perp}(t) \rangle$, and the sum $\langle I_{Total}(t) \rangle$. Inset, degree of polarization, $P(t)$ (open circles) and also time-windowed intensity correlation function C_I (solid curve). The latter is computed by windowing $I_{\perp}(t)$ and $I_{\parallel}(t)$ in the time domain by use of a 100-ps square window and computing the correlation as a function of the window position as described in Ref. 20. Vertical dashed lines, range of times during which singly scattered photons arrive, as determined from Fig. 3 below. Note that the time axis here and throughout this Letter has an arbitrary time of $t = 0$, which is determined by the absolute transmitter-to-receiver distance and is the same for all our measurements.

along with the averaged intensity cross correlation $C_I = \langle I_{\parallel}(t) I_{\perp}(t) \rangle$ computed by use of a sliding time window.[20] This time-windowed cross correlation, which shows the temporal evolution of the degree of correlation between the two intensities, also shows an enhancement before 600 ps.

The high time resolution of our measurements permits us to directly measure the time window during which single-scattered photons emerge from the medium. Without changing any of the other parameters of the measurement, we placed a long aluminum rod vertically into the random medium, as illustrated in Fig. 2. Because aluminum is such a strong reflector, the intensity of radiation scattered only from the rod is much larger than the radiation scattered from the random medium. Thus the signal measured with the rod in place is dominated by the radiation scattered directly from this metal object. By moving the rod to many different locations in the path of the incident radiation, it is possible to determine the maximum and minimum transit times for singly scattered photons. This procedure essentially permits us to calibrate the arbitrary delay axis in terms of the earliest- and latest-arriving singly scattered photons.

Figure 2 illustrates the two rod locations that correspond to the longest and shortest photon paths, P_L and P_S, respectively. The measured waveforms that correspond to those limits, Figs. 2(b) and 2(c), exhibit clear signatures of the single scattering of radiation from the rod and are therefore unambiguous markers of the limits of the temporal window that correspond to single scattering. The dashed vertical lines in Fig. 1 (inset) show that these limits correspond well

to the temporal window in which $P(t)$ is nonzero and also to the window during which correlation $C_I(t)$ is enhanced. This result suggests that only two scattering events are needed to randomize the polarization, so $\xi / l^* < 2$. We note that most earlier methods for determining this depolarization rate relied on computation of the scattering cross section by use of Mie theory,[2,4,5,8,12] which is difficult to apply in the case of irregularly shaped or densely packed scatterers.[21]

For broadband waves, the evolution of the spectral content of the radiation can be another useful quantity for distinguishing the ballistic from the diffusive regime. To extract this spectral evolution we perform a Hilbert transformation on the measured waveforms to obtain a complex representation of $E(t)$. The angle of this complex quantity is the temporal phase $\phi(t)$, which is related to the instantaneous frequency by $d\phi / dt = 2\pi v_{inst}(t)$. We compute the average phase function for both parallel and perpendicular polarizations, $\langle \phi_{\parallel}(t) \rangle$ and $\langle \phi_{\perp}(t) \rangle$, by configurationally averaging the phase functions of all the individual waveforms. The results are shown in Fig. 3.

These data show several interesting features. First, the phase functions are both slightly nonlinear with a positive chirp, indicating that, on average, the later portions of the waveforms have a higher average frequency. This can be qualitatively understood from the frequency dependence of the mean free path[18]; as the higher-frequency components scatter much more readily, it is expected that these components would have longer average paths in the medium and therefore would emerge later. Second, $\langle \phi_{\perp}(t) \rangle$ is essentially
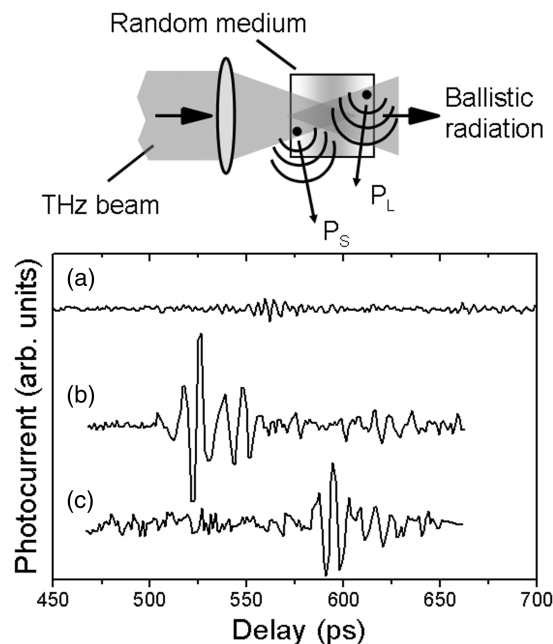

Fig. 2. Top, a schematic illustrating the method that we used to determine the arrival times of singly scattered photons. A metal rod is placed at several different locations in the medium, giving rise to a strong single scattering signature. Bottom, (b), (c) waveforms that correspond to the locations of the metal rod that resulted in the earliest (P_S) and the latest (P_L) arrival times. Waveform (a) was measured without the rod present.
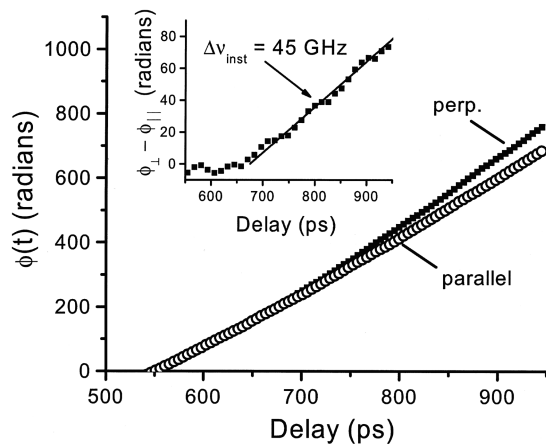
Fig. 3. Average phase functions for parallel $\langle\phi_\parallel(t)\rangle$ and perpendicular $\langle\phi_\perp(t)\rangle$ polarizations. Inset, difference between these two functions. A linear fit to this difference, at later delays, yields the average frequency difference between the two polarizations' components in the diffusive regime.

equal to $\langle\phi_\parallel(t)\rangle$ until roughly $t = 650$ ps, after which it increases more rapidly. The larger slope indicates a higher average frequency for the perpendicular component. Because the perpendicular component must have experienced (on average) more scattering events, it is reasonable to expect that it comprises those photons that are more likely to scatter. The inset of Fig. 3 shows the difference $\langle\phi_\perp(t)\rangle - \langle\phi_\parallel(t)\rangle$, which shows a nearly linear increase for later times. The slope of this line gives the average frequency difference Δv_{inst} between the two perpendicular field components at these later times, when the propagation is completely diffusive. Such spectral shifts are not unknown in multiple scattering,[22] but this is the first demonstration to our knowledge of a polarization-dependent frequency shift.

In conclusion, we have observed that, in the diffusive regime, the average spectral content of the two orthogonal polarization components is distinct. This divergence occurs *after* the regime of single scattering, where both the degree of polarization and the intensity correlation have vanished, and is therefore a feature of diffusive broadband radiation. It arises from the frequency dependence of the scattering parameters such as the mean free path, which vary strongly within the bandwidth of the incident radiation. We note that this frequency shift goes hand in hand with the delayed emergence of the perpendicular component of the ra-

diation, which gives rise to the variation of $P(t)$: Both are a consequence of the larger number of scattering events experienced, on average, by the perpendicular component.

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