Examination of the spatial and temporal field distributions of single-cycle terahertz pulses at a beam focus

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Measurements of spatially resolved beam profiles of single-cycle terahertz pulses are presented. In the approach here the terahertz beam is scanned over the stationary electro-optic detector using a gimbal-mounted mirror. A detailed study of the temporal- and frequency-dependent field in the region of the terahertz focus reveals field patterns which are dominated by diffraction and absorption. The observed field distribution is reproduced in model calculations. The exceptionally high signal to noise ratio and the high frequency and spatial resolution of the experiment here facilitate the observation of minute field distortions caused by the anomalous dispersion of absorbers in the beam path from the spatially resolved field pattern.
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The spatial profile of single-cycle terahertz pulses is greatly affected by optical elements which modify the temporal characteristics and thus the bandwidth of the pulse. Spatiotemporal reshaping is mainly associated with diffraction on apertures and refraction, scattering, and absorption in optical components or samples. Although a detailed understanding of the characteristics of a terahertz system and its components would in principal require the knowledge of the spatially resolved field distribution both in the time and the frequency domain, experimental studies have been sparse due to the difficulties involved in such measurements. Either a single-spot detector was scanned through the beam[1,2] or the two-dimensional (2D) field was recorded electro-optically in a nonlinear crystal using a charge coupled device camera.[3–5] Whereas the first approach requires careful re-alignment of the laser after each step or the use of fiber-coupled detector systems,[6–10] the latter typically suffers from a small dynamic range and low signal to noise ratio.

In this letter we report an approach based on rotating a gimbal-mounted terahertz mirror with respect to a stationary detector. Using this system we perform measurements of the temporal and frequency profiles of the spatial electric field distribution in the focus of a terahertz pulse and discuss a range of pulse forming effects.

A standard terahertz spectroscopy setup[11,12] based on terahertz emission from a photoconductive antenna and electro-optic detection was modified to spatially resolve the terahertz field in the focus, as illustrated in Fig. 1. Briefly, the output of a mode-locked Ti:sapphire laser is split into an excitation and a detection beam. The excitation pulses are focused into the 80 μm gap between the electrodes of a biased photoconductive GaAs antenna. The emitted terahertz radiation is coupled out of the substrate by a collimating silicon lens with a diameter of 10 mm in contact with the emitter.[1,8] Two planoconvex Teflon lenses, L1 and L2, each with a nominal focal length of $f=100$ mm generate a terahertz focus at the image plane which is located at the position of the electro-optic detector. A focused detection beam illuminates a 500 μm diameter spot on a 1 mm thick (110)-ZnTe crystal to detect the terahertz field.[13,14] The polarization of the probe beam and the crystal axis are oriented to detect the x component of the linearly polarized terahertz field which is also polarized in the x direction due to the orientation of the electrodes. In order to enable spatially resolved measurements we introduce into the terahertz beam path a gimbal-mounted plane mirror with a pinhole of 1 mm diameter for the probe beam. The gimbal pivot axis is located at the position of its aperture (see inset of Fig. 1). In this configuration a rotation of the mirror moves the terahertz focus with respect to the fixed position of the probe beam and the detector and thus allows us to spatially resolve the electric field of the terahertz pulse. The maximal range of rotation was ±7° which corresponds to a scan range of 18 mm in the image plane. By using a gimbal mount with the rotation axis defined by the intersection of the mirror's front surface with the plane orthogonal to the probe beam, we minimize shifts of the focal plane in the z direction. We estimate a maximum time delay of 1.8 ps for the pulses recorded at the limits of our scan range. Rotating the mirror around two orthogonal axes in the x and y directions intersecting at the center of the

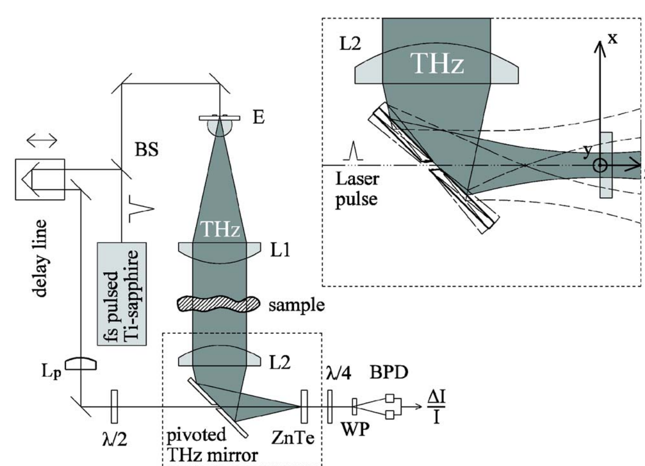

FIG. 1. (Color online) Experimental setup. The inset shows the spatial displacement induced by rotating the gimbal-mounted plane mirror. (BS, beam splitter; E, terahertz emitter; L_p, optical lens; WP, Wollaston polarizer; BPD, balanced photodiodes.)

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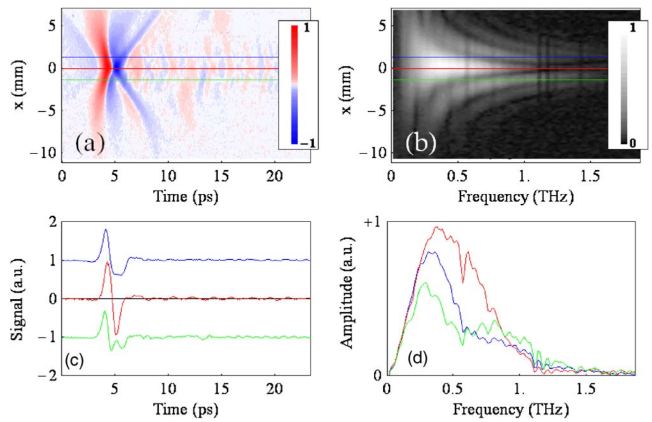

FIG. 2. (Color online) Line scans along the x axis of the spatial field distribution measured in the terahertz focus in the (a) time domain and (b) frequency domain. (c) Corresponding terahertz wave forms measured at the locations of the lines in (a). The curves are offset vertically for clarity. (d) Terahertz spectra corresponding to cross sections through the radiation pattern in (b) as indicated by the lines.

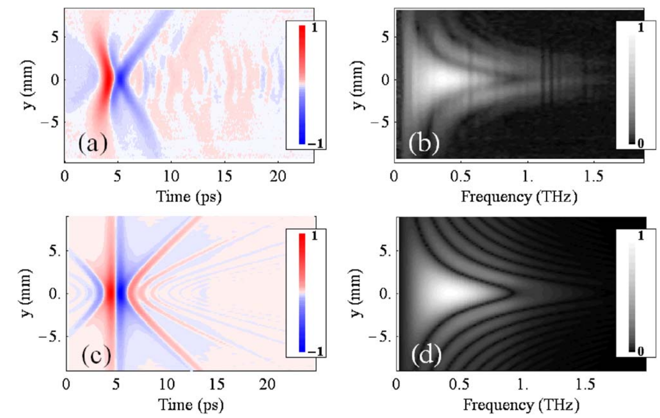

FIG. 3. (Color online) Line scans along the y axis of the spatial field distribution measured in the terahertz focus in the (a) time domain and (b) frequency domain. (c) and (d) are simulations of the field distribution.

mirror would in principle enable 2D scans of the time-dependent terahertz field $E(t,x,y)$ in the focal plane. We exclusively rotated around one axis which produces a single line scan. Then, after rotating the emitter and detector components by 90° around the optical axis we were able to scan the focal plane along the perpendicular axis. Due to the rotational symmetry of the setup the two scans represent cross sections through the focus parallel and perpendicular to the terahertz polarization corresponding to line scans along the x and y axes.

In Fig. 2(a) we plot the spatially resolved electric field $E(x,t)$ at the terahertz focus scanned along the x direction. Figure 2(c) shows cross sections along the three lines drawn in Fig. 2(a) representing the terahertz wave forms at positions $x=+1.5$, 0, and -1.5 mm with respect to the origin located at the center of the focus. Fourier transformation of the time-resolved wave forms yields the frequency-dependent field amplitudes and phase at each spatial point. Figure 2(b) shows a grayscale image of the spatial dependence of the normalized field amplitude $E(x,f)$ around the focus. Again, in Fig. 2(d) we plot amplitude spectra corresponding to the three locations around the origin. In the time-domain plot we observe a characteristic X-shape pattern of the field maxima. Similar radiation patterns were reported previously and have been shown to originate from geometric diffraction at the apertures of the optical components in the terahertz beam.[1,4,10] This characteristic field distribution in the time domain leads to an asymptotic pattern of field minima in the frequency domain. In our case these patterns are directly associated with geometric diffraction on the clear apertures of the lens system consisting of L1 and L2 as we will demonstrate below. In Fig. 2(a) the main field maxima are followed by periodic oscillations due to resonant excitation of rotational transitions in water molecules in the beam path.[15] The transitions appear as vertical absorption lines in the 2D frequency plots, in analogy to Fraunhofer lines in the optical spectrum of the sun.

Assuming a cylindrically symmetric emission pattern of our antenna, we expect rotational symmetry of the radiation pattern around the beam axis (z direction). In fact the line scan in the y direction given in Figs. 3(a) and 3(b) essentially shows the same features as the x scan in Fig. 2, not unexpected due to our rotationally invariant imaging setup. Slight variations in the frequency distribution, in particular, the minimum for $y=0$ at a frequency of 0.9 THz in Fig. 3(b), can be attributed to an offset of the scan axis with respect to the origin. This is due to a small misalignment of the setup as we will show in the following.

In order to model the spatial frequency distribution at the focus, we approximate our system by a single focusing lens with a circular aperture and calculate the diffraction pattern for each frequency using a plane wave approximation. For this greatly simplified case, the electric field distribution in the focal plane generated by Fraunhofer diffraction from the lens aperture is given by[16]

$$E(x,y,\lambda) = E_0(\lambda)\left|\frac{2J_1(\pi Dr/\lambda f)}{\pi Dr/\lambda f}\right|, \quad r=\sqrt{x^2+y^2}, \qquad (1)$$

where λ is the wavelength of the incoming plane wave, D and f are the diameter and the focal length of the lens, and J_1 is the first order Bessel function. $E_0(\lambda)$ is the spectral amplitude at a given wavelength of the incident terahertz wave. For our simulations we used the spectrum measured at the center of the beam focus for the incident spectral distribution. Note that Eq. (1) is invariant under rotation around the z axis and therefore the simulated spectra also show rotational symmetry. In order to incorporate misalignment effects we introduced an adjustable vertical offset through the transformation $x \mapsto x - x_0$. Figure 3(d) shows the result of a simulation as a cross section along the y axis. We achieve the best agreement between the measured pattern in Fig. 3(b) and our simulation with a vertical offset of $x_0=900$ μm and a ratio $D/f=0.27$, different from the nominal ratio of our optical part parameters, $D/f=0.48$. We attribute this discrepancy to the fact that in contrast to the model assumption of a single lens, we have an optical system consisting of the silicon substrate lens, and L1 and L2. It has been shown that spherical substrate lenses show frequency-dependent angular radiation patterns.[1,7,10] Therefore in a more realistic model the characteristic emission pattern and diffraction on the apertures of L1 and L2 would have to be included. Furthermore, the frequency-dependence of the beam diameter in the region between L1 and L2 (Ref. 10) and the limited spatial resolution of the experiment (probe beam diameter of 500 μm) would have to be taken into account. Nevertheless, in spite of these shortcomings our simple model already reproduces

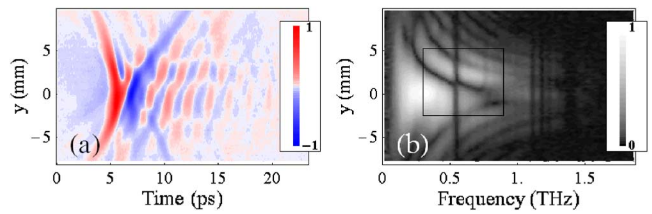

FIG. 4. (Color online) (a) Spatiotemporal line scan of the terahertz focus with an irregularly shaped α-lactose sample in the beam path. (b) Corresponding radiation pattern in the frequency domain.

all the characteristic features of the measured radiation pattern. To convert the simulated frequency distribution into the time domain by an inverse Fourier transformation, we assume a linear phase of the form $\phi(\nu)=a\nu+b$ with $\nu=c/\lambda$. Whereas variation of parameter a introduces a temporal delay and phase shift of the terahertz wave form, changing parameter b modifies the pulse amplitude and polarity. Animations of these effects can be found on our homepage.[17] Figure 3(c) shows the simulated spatio-temporal field distribution where we used the modeled frequency distribution in Fig. 3(d) and adjusted a and b to best reproduce our measurement. The simulation consistently reproduces the X-shape pattern. We conclude that the spatiotemporal and spatiofrequency field distributions at the focus of a terahertz pulse are dominated by diffraction caused by the apertures of the optical components and by resonant absorption of substances in the beam path.

In addition refraction or scattering caused by arbitrarily shaped samples can influence the field pattern as we will demonstrate by the following experiment. For this purpose we introduced an arbitrarily shaped polyethylene (PE) bag filled with polycrystalline α-lactose powder into the beam path between the two Teflon lenses, as indicated in Fig. 1. The mean thickness of the bag was 2 mm. Whereas the PE bag is transparent at terahertz frequencies, the lactose sample displays a number of strong resonances in the far infrared.[18] The resulting time domain plot in Fig. 4(a) shows a distorted X shape followed by periodic oscillations of the field induced by the lactose absorption. Due to the spatial variations of the sample thickness, portions of the terahertz pulse experience an additional phase delay with respect to the others. Subsequent superposition in the terahertz focus generates the observed field distortion and shifts of the wave fronts. The frequency-domain plot in Fig. 4(b) reveals the prominent lactose absorption lines at 0.53 THz and at 1.4 THz. The minima in the region around ~1.2 THz are again due to water vapor absorption. Initially one expects diffraction and absorption to be independent effects.

Interestingly, closer inspection points to a significant displacement of the diffraction minima in the vicinity of the strong lactose absorption line. This effect is clearly visible in the magnification shown in Fig. 5 which represents a zoom into the region indicated by the square in Fig. 4(b). An analogous deformation of the white light line spectrum near the D line of Na vapor in the flame of a Bunsen burner has been observed as early as 1880 by Kundt.[19] In Kundt's case this was the result of wavelength-dependent refraction at the prism-shaped absorption zone generated by the inhomogeneous temperature distribution in the flame. This induces anomalous dispersion close to the strong absorption line.[19,20] Analogous to this effect the terahertz pulse in our experiment encounters a strong variation of the refractive index in the vicinity of the absorption line. The irregularly shaped lactose sample leads to local shifts of the terahertz wave from the beam axis through index-dependent refraction and propagates this effect into spatial displacement of the diffration minima near the absorption minima.

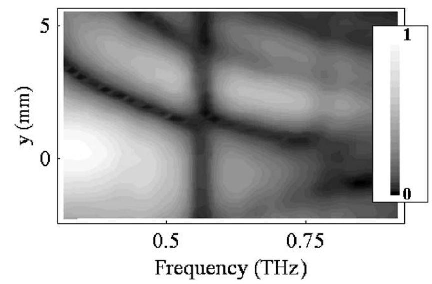

FIG. 5. Zoom into the region indicated by the square in Fig. 4(b).

In conclusion we measured the spatial distribution of the electric field near the focus of a terahertz pulse. For this purpose we developed a system where the terahertz focus is scanned over a stationary detector using a gimbal-mounted mirror. Our measurements have been compared to a simple diffraction model. As a demonstration of the system's performance we report the observation of smallest beam displacements induced by frequency-dependent refraction in an irregularly shaped absorbing sample.

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