Terahertz vibrational absorption spectroscopy using microstrip-line waveguides

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We demonstrate that terahertz microstrip-line waveguides can be used to measure absorption spectra of polycrystalline materials with a high frequency resolution (~2 GHz) and with a spatial resolution that is determined by the microstrip-line dimensions, rather than the free-space wavelength. The evanescent terahertz-bandwidth electric field extending above the microstrip line interacts with, and is modified by, overlaid dielectric samples, thus enabling the characteristic vibrational absorption resonances in the sample to be probed. As an example, the terahertz absorption spectrum of polycrystalline lactose monohydrate was investigated; the lowest lying mode was observed at 534($\pm$2) GHz, in excellent agreement with free-space measurements. This microstrip technique offers both a higher spatial and frequency resolution than free-space terahertz time-domain spectroscopy and requires no contact between the waveguide and sample. © 2008 American Institute of Physics. [DOI: 10.1063/1.3013349]

Terahertz time-domain spectroscopy (TDS) is widely used to measure the spectral absorption features of crystalline materials across the frequency range from tens of gigahertz to several terahertz.[1] In conventional free-space terahertz-TDS systems, broadband pulsed terahertz radiation is typically generated by sub-picosecond-duration current transients in a photoconductive switch; this radiation is focused onto and transmitted through a sample before being detected coherently at a second photoconductive switch or by an electro-optic crystal. Free-space terahertz TDS has allowed detection of vibrational modes in a wide variety of polycrystalline compounds with typical system bandwidths in excess of several terahertz. Samples need to be sufficiently thick to produce a measurable interaction while nevertheless allowing a detectable portion of the terahertz signal to be transmitted. It has recently been shown that terahertz spectroscopic absorption resonances can also be measured using low-loss free-standing metal wire waveguides[2] and by parallel plate waveguides.[3,4] In these studies, the waveguide confines the propagating electric field, so increasing its interaction with samples.

In this letter, we demonstrate the potential of lithographically defined on-chip microstrip waveguides for measuring the broadband terahertz absorption spectra of polycrystalline materials using integrated pulsed terahertz emitters and detectors. The evanescent terahertz field extending above and propagating along the microstrip line penetrates and interacts with dielectric samples held in close proximity. The propagating electric field can therefore become modified, exhibiting spectral features corresponding to the characteristic vibrational absorption resonances of the sample, which are subsequently determined by Fourier transform of the detected time-domain signal.

We fabricated a 25/250-nm-thick Ti/Au microstrip line on a 6 μm thick benzocyclobutene (BCB) dielectric layer, itself formed on a 25/500 nm Ti/Au coated Si wafer, which

was used as the backplane (Fig. 1). Integrated photoconductive switches for terahertz-bandwidth signal excitation and detection were formed from low-temperature gallium arsenide (LT-GaAs), grown by molecular beam epitaxy on a sacrificial 100-nm-thick AlAs layer, itself grown on a GaAs substrate. Epitaxial lift-off of the 350-nm-thick LT-GaAs layer was achieved using dilute hydrofluoric acid (10%) to remove selectively the AlAs layer, before the transfer of the LT-GaAs onto the BCB, using wax as a support.[5,6]

The width of the microstrip line was chosen to be 30 μm with pulses transmitted over an "active" length of microstrip between the LT-GaAs emitter and detector of 2.8 mm. The "parasitic" length of the microstrip beyond each photoconductive switch was made 7.4 cm long to delay the
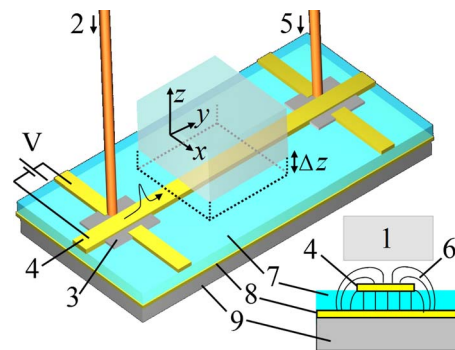

FIG. 1. (Color online) Main figure: schematic of lactose monohydrate samples (1) being investigated using a terahertz microstrip system. An ~12 fs duration, 800 nm laser beam with a repetition rate of 80 MHz (2) is focused on a biased (5–40 V) photoconductive LT-GaAs switch (3). The terahertz pulses generated by photoconduction are coupled directly into a gold microstrip line (4) where they propagate as ~picosecond duration electrical pulses. The time-domain amplitude of the electrical transients is sampled by a time-delayed portion of the femtosecond laser beam (5) focused on a second integrated photoconductive switch. Inset: cross-sectional view of the microstrip line showing a schematic of the electric fields associated with current pulses propagating on the microstrip line. The evanescent field (6) penetrates the lactose sample held above the microstrip, recording its absorption spectra. Under the microstrip signal conductor, a BCB dielectric layer (7) overlays a Ti:Au backplane (8) formed on a Si wafer (9).
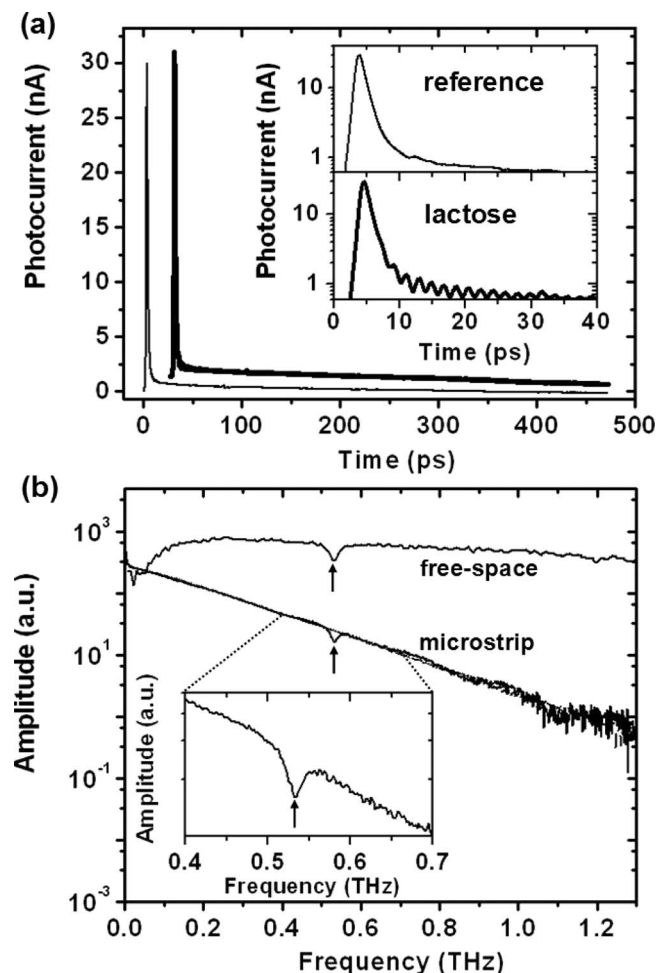
(a)

(b)

FIG. 2. (a) Main figure: time-domain measurements of terahertz pulses transmitted along the microstrip line over a 470 ps time window, yielding a frequency resolution of 2 GHz in the Fourier transform. The bold line indicates data obtained after contacting the lactose sample with the microstrip and is horizontally and vertically offset for clarity. The faint line represents (reference) data obtained in the absence of a lactose sample. Inset: pulsed data over a shorter time window, highlighting the ringing indicative of frequency-specific absorption in the lactose sample. (b) Fourier transforms of the data shown in (a) with data close to the lactose absorption resonance magnified for clarity. Transmission data obtained using a free-space terahertz-TDS system are also shown for comparison. The absorption resonance at 534 GHz is indicated by arrows.

arrival of reflections from either end of the microstrip and thus maximize the sampling time window for the main transmitted pulse arriving at the LT-GaAs detector. The ability to produce a longer reflection-free time window in this way, and therefore a higher frequency resolution Fourier transform, is a significant advantage of our technique compared with those obtained in many other terahertz spectroscopy systems. The total length of the microstrip line was chosen to be 15 cm, as a compromise between maximizing device yield (given the extreme 5000:1 aspect ratio of the microstrip signal conductor so formed) and maximizing the frequency resolution. Measurements were performed using an $\sim$15 mW, $\sim$12 fs pulse duration Ti:sapphire laser, which illuminated the biased (40 V) LT-GaAs switch regions for terahertz pulse emission; an $\sim$15 mW time-delayed portion of the beam was focused onto a second LT-GaAs switch for signal detection.[5] Terahertz pulses were measured at the detection switch over a typical time window of 470 ps [Fig. 2(a)], over which no signal reflection occurred, yielding a

frequency resolution of 2 GHz after Fourier transformation of the pulsed data.

Samples of pure lactose monohydrate (Sigma-Aldrich) were compressed into pellets and then diced into $1 \times 1 \times 0.5$ mm^3 samples. Each sample was mounted on a brass holder using a hard-setting varnish, itself attached to a three-axis linear translation stage (Ocean Optics), to control its position relative to the microstrip; all samples were initially mounted on plane-parallel full contact with the microstrip signal conductor in order to maximize their interaction. All samples measured (five in total) showed a clear absorption resonance at 534($\pm$2) GHz [Fig. 2(b)]. The absorption resonance is also evident in the corresponding time-domain terahertz signal [inset, Fig. 2(a)] as a train of oscillations in the pulse tail.

The frequency position of the 534 GHz absorption resonance was confirmed by direct comparison to spectra recorded in a free-space terahertz-TDS system [also shown in Fig. 2(b) using a 0.38-mm-thick pellet]; it also agrees well with prior experimental data undertaken using a range of techniques.[3,7] The microscopic molecular motion responsible for this mode has recently been the subject of detailed periodic density functional theory calculations,[8,9] which attribute the mode to a hindered external rotational mode (rather than an internal molecular mode), with an unusually long lifetime (and therefore sharp resonance in the frequency domain). By fitting a Lorentzian to our absorption resonance, a full width at half maximum (FWHM) of 22($\pm$4) GHz was calculated, with an equivalent damping period $\tau = (\pi \text{ FWHM})^{-1}$ of $\sim$14 ps, in excellent agreement with data obtained in Ref. 7 using a continuous-wave frequency-multiplier-chain source. We calculate the effective absorption strength of the 534 GHz resonance as $\alpha_{\text{eff}} = 5.26(\pm 0.2)$ cm^{-1} for our microstrip measurements. Our free-space terahertz-TDS measurements show a rather higher value for the same resonance [$\alpha_{\text{eff}} = 88.1(\pm 1)$ cm^{-1}]. We interpret the reduced effective absorption strength measured in microstrip to be caused by the smaller portion of the total propagating electric field interacting with the sample; a similar reduction in α_{eff} measured using microstrip compared with free-space measurements was recently observed for liquid systems.[10]

Further experiments were undertaken to examine in detail the interaction of samples with the evanescent field and to demonstrate that samples do not have to be in direct contact with the microstrip line for the absorption resonances to be recorded in the propagating current pulse; this will be important for potential applications where repeated contact with the samples under test could damage the microstrip line or the sample itself. The x-y-z translation stage was used to vary the separation of the microstrip and lactose sample over the range from 0 μm (full contact) to 200 μm (outside the region of evanescent field as assessed using our simulations described later). The depth of the vibrational resonance rapidly reduced as the sample-to-microstrip distance was increased, falling below the noise floor for separations >100 μm (Fig. 3). The amplitude of the propagating pulse and depth of the absorption resonance measured are controlled by the bias voltage applied to the emitter photoconductive switch (Fig. 3); the bias controls the propagating current density and therefore the intensity and extent of the evanescent field extending above the microstrip line. For all photoconductive switch biases, the depth of the vibrational
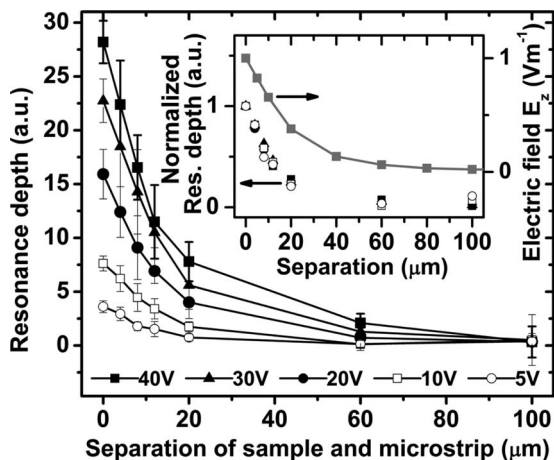
FIG. 3. Data showing the 534 GHz vibrational resonance depth (defined as the maximum difference in amplitude of the frequency domain spectra, at the center of the resonance, from a reference trace taken with no sample present) as a function of the separation between the microstrip line and lactose sample. Data are shown for the range of photoconductive emitter bias voltages as indicated. The progressively larger resonance depths observed, as the bias voltage is increased, are attributed to the increased current density in the microstrip line induced by the larger-amplitude terahertz pulses. Inset: resonant depth of the data for all values of voltage normalized to the value calculated for zero separation, showing collapse of the data onto a single curve. 3D electromagnetic simulation results are also presented for the normalized maximum instantaneous value of the transverse electric field E_z above the center of the microstrip line. All lines are a guide to the eyes.

resonance as a function of sample-to-microstrip distance fell onto a single curve when the data were normalized to the absorption depth observed when the sample was in full contact with the waveguide (Fig. 3 inset).

A numerical, fully three-dimensional (3D), frequency-dependent electromagnetic simulation of the system (undertaken using the Ansoft high frequency structure simulator) provided calculations of the instantaneous electric field strength at arbitrary positions around the microstrip waveguide. The functional form of the maximum instantaneous value of transverse electric field (E_z) intensity at the resonant absorption frequency (534 GHz) at the sample location was found to correspond well with the observed decay of the resonance (Fig. 3 inset). We note that a sample separation of 20 μm yielded very clear absorption resonances above the noise floor, at which separation we estimate from the simulation data (Fig. 3) that only $\sim$37% of the propagating evanescent electron field penetrates the sample. We can use this figure to give an order-of-magnitude estimate for the minimum volume of lactose required in our microstrip setup to observe the 534 GHz resonance above the noise floor; assuming that 37% of the propagating evanescent electric field volume must be perpetrated, we arrive at $\sim 10^{-12}$ m^3, which is of order one-hundredth of the electric field volume interacting with a sample in a typical diffraction-limited terahertz-TDS system.

The frequency resolution ($\sim$2 GHz) of our spectral measurements is over three times smaller than those recently

reported for parallel plate waveguide systems.[4] The high (2 GHz) spectral resolution reported in the present work is expected to be useful in low-temperature measurements of linewidth in polycrystalline solids (as has been the case with high-resolution parallel-plate waveguide experiments[4]); we note that the feasibility of terahertz bandwidth time-domain measurements of microstrip at low temperatures (down to 4 K) has already been demonstrated in our prior work.[11]

We note that our numerical simulations of planar Goubau-line waveguide confirm a higher proportion of electromagnetic field above the waveguide than for microstrip waveguides, which may make them preferable for trace sensing applications that require even higher sensitivity. A limitation of the present measurements is the bandwidth available after radiation loss from our 2.8-mm-long microstrip ($\sim$1.2 THz). Prior experiments using polypropylene as the microstrip dielectric (in place of BCB) showed that this can improve high frequency attenuation;[12] better optimization of LT-GaAs carrier lifetime in combination with use of such dielectrics will be needed to further enhance bandwidth in the present terahertz spectroscopy modality. We note that our existing bandwidth of $\sim$1.2 THz is approximately three times that obtained in suspended wire waveguides,[2] but only one-half of that obtained in parallel-plate waveguide systems.[3,4]

In summary, we have demonstrated the capability of planar microstrip circuits to resolve narrow spectral features of polycrystalline materials in the terahertz frequency range. The broadband spectrum of polycrystalline lactose monohydrate was measured over the frequency range of 0.1–0.8 THz using a terahertz microstrip line, with a resolution of 2 GHz, which is unprecedented for pulsed terahertz techniques.

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