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A versatile and reconfigurable setup for all-terahertz time-resolved pump-probe spectroscopy

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A versatile optical setup for all-terahertz (THz) time resolved pump-probe spectroscopy was designed and tested. By utilizing a dual THz pulse generator emitter module, independent and synchronized THz radiation pump and probe pulses were produced, thus eliminating the need for THz beam splitters and the limitations associated with their implementation. The current THz setup allows for precise control of the electric fields splitting ratio between the THz radiation pump and probe pulses, as well as in-phase, out-of-phase, and polarization dependent pump-probe spectroscopy. Since the present THz pump-probe setup does not require specialized THz radiation optical components, such as phase shifters, polarization rotators, or wide bandwidth beam splitters, it can be easily implemented with minimal alterations to a conventional THz time domain spectroscopy system. The present setup is valuable for studying the time dynamics of THz coherent phenomena in solid-state, chemical, and biological systems. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4717732]

I. INTRODUCTION

The study of intricate temporal dynamics of various ultrafast phenomena has become widespread, spanning various fields in physics, chemistry, biology, and many engineering disciplines.[1] The use of femtosecond and attosecond laser pulses, along with the development of configurable time-resolved optical setups and accurate pulse characterizations techniques, were key in the advancement of these fields. These techniques are all-optical in nature, as femtosecond or attosecond temporal resolutions exceed that of the state-of-the-art electronic instrumentations. While the ultrafast time-resolved measurement techniques can be assembled in various optical configurations, they all share a common theme as they all employ pump and probe-type experimental arrangements to infer the temporal dynamics of the phenomenon under study.[2]

Conventional ultrafast time-resolved spectroscopy techniques employ temporally-delayed ultrashort light pulses, referred to as pump or probe pulses, to sweep the ultrashort time interval of interest.[3] In an optical regime type of experiment, a single ultrashort seed pulse, generated from an ultrafast laser source, is split into two pulses (or more for specialized experiments) via passive optical components (e.g., reflection, polarizing, fiber, lamellar mirrors, and grating type beam splitters). These pulses are referred to as the pump and probe pulses. As the pump and probe pulses follow different optical paths, one pulse is directed along a variable path length to vary the relative timing between the pump and probe pulses. The pump pulse, which is the more intense pulse, is used to initiate the time-dependent phenomenon on a sample. The spatial extent of the ultrafast event is then probed, at a predefined temporal delay, by a much weaker probe pulse. Thus, the instantaneous response of the physical phenomenon can be detected at a particular temporal delay. By repeatedly probing the excited location at different time delays and recording the probe pulse response, the underlying time-resolved dynamic in samples can be mapped out as a function of the time delay between the pump and probe pulses.[4] A fraction of a femtosecond time resolution is easily achieved as it is limited only by the duration of the optical seed pulse and the degree of the spatial overlap between the pump and the probe pulses.

Similarly, in optical-pump/terahertz (THz) radiation or THz radiation-pump/THz radiation-probe time-resolved spectroscopy, the intense optical or THz radiation pulse serves to disturb the sample to create the ultrafast event and a synchronically-generated THz radiation pulse is directed and focused on the same spot probing the phenomenon of interest.[5] Precise time delays can be produced with optical path length differences between the pump and the THz radiation probe pulses, thus enabling high precision time-resolved investigation of temporal response at THz frequencies.

A common method of producing two synchronized THz radiation pulses is by dividing a single THz radiation pulse by the use of reflective dielectric beam splitter.[6] However, for THz-pump/THz-probe time-domain spectroscopy (THz-PP-TDS), such an implementation is cumbersome as it often requires positioning the THz radiation beam splitter in the collimated and expanded portion of the THz radiation beam.[7] This in turn necessitates the use of large diameter beam splitters (usually a few inches in diameter), to divide and superimpose the THz radiation pump and probe pulses, and exceedingly larger reflective optics to ensure complete interception of the THz radiation. Another disadvantage in using a beam splitter for such an experimental configuration, especially for co-linear arrangements, is the fact that 50% of the THz radiation intensity is removed and not utilized in the experiment. This arises due to secondary transmissions through the beam splitter since the THz pump and probe pulses are recombined. The loss of half of the THz radiation intensities can be detrimental especially when the seed THz radiation pulse intensity is very weak to start. Other limitations imposed in using THz beam splitters for THz-PP-TDS experiments are: the lack of

the ability to precisely adjust the splitting ratio between the THz radiation pump and probe pulses as such requires the use of a different beam splitter for each ratio, lack of low loss ultrawide bandwidth THz radiation beam splitters,[8] and THz radiation interference effects within the beam splitter especially for ultrashort THz radiation pulses where multiple reflections occur at the dielectric-air interfaces within the beam splitter. In an infrared pump-probe configuration, Joffre *et al.* reported on producing two almost collinear synchronized infrared pulses via optical rectification in a GaAs substrate.[9] Although the beam splitter was eliminated in this configuration, however, the setup had several limitations such as: low degree of co-linearity, inaccessibility to near-zero delay time, limited pump-probe modulating frequencies, and the inability of independently controlling the electric field polarization of the pump and the probe.

In this article, we demonstrate a versatile THz time domain spectroscopy (THz-TDS) system that can be configured for either a traditional THZ-TDS or a THZ-PP-TDS arrangement. The setup overcomes the abovementioned inherent constraints and limitations in traditional THz-PP-TDS systems. The setup utilizes a dual pulse generator module based on semiconductor photoconductive emitters to generate two independent but synchronized THz radiation pump and probe pulses. Since the present THZ-PP-TDS setup eliminates the use of a beam splitter, it allows for 100% utilization of the THz radiation pulses generated at the source for time-resolved experiments. The THz setup allows for precise control of the splitting ratio between the THz radiation pump and probe pulses by independently controlling the level of optical excitation of the PC THz emitters. Without the use of additional components, the system has the added capabilities of providing both in-phase and out-of-phase THz-PP-TDS experiments as well as polarization-dependent THz-pump/THz-probe measurements. The setup has a compact footprint, is easy to align, does not require specialized THz radiation optical components, and can be implemented in any THz-TDS system.

II. SETUP DESIGN

In all pump-probe type experiments, a crucial factor is precise synchronization between the pump and probe pulses. Thus, our THz-PP-TDS design setup takes advantage of the THz generation process from the PC emitters. The basic principle behind the THz-PP-TDS is as follows: Since the PC THz radiation pulse generation mechanism is sensitive to the shape of the envelope, not phase, of the excitation optical pulse, synchronized THz radiation pulses can be generated from identical optical pulses which are phase-locked and accurately timed. That is, if two identical PC emitters are placed in very close proximity to each other (i.e., at the collection point of the THz-PP-TDS setup as shown in Figure 1(a)), and are excited by two femtosecond optical pulses (i.e., optical pump 1 and optical pump 2) driven from the same femtosecond laser pulse source, two synchronously phase-locked and amplitude stable THz radiation pulses can be generated. In this situation, one of the THz radiation pulses serves as a pump pulse and the other serves as a probe pulse. By varying the time delay between the two femtosecond optical pump pulses (t_{d2}), the two THz radiation pulses follow the same relative time delay. Thus, for the THz-PP-TDS experiments, instead of scanning the time delay between the pump and probe THz radiation pulses, one can simply delay the femtosecond optical pump pulses exciting the PC emitters and achieve a precise time-resolved response from the THz radiation probe pulse. Clearly, such a design eliminates the use of an optical beam splitter and the cumbersome optics often required to redirect the THz radiation pulses. Furthermore, precise control of the relative electric magnitudes of the THz pump and probe pulses can be simply achieved by either independently varying the bias voltage on the PC THz emitters or by changing the amount of optical energy fluence exciting the PC THz radiation emitters.

Central to the implementation of this THz-PP-TDS design is the ability to independently address the two PC THz emitters. However, having the two PC THz emitters integrated on a common semiconductor substrate and being placed in very close proximity to each other, results in a non-linear coupling between the PC transient currents. In this situation, the two PC THz emitters' response can strongly couple due to photogeneration and diffusion of carriers in the semiconductor substrate. To eliminate current cross-talk and improve the THz radiation pulses' amplitude stability, the two PC THz emitters must be both spatially and electrically isolated from each other.

The THz-PP-TDS system presented here consists of two identical and independent PC THz emitters. Each PC THz emitter is a 2 mm long coplanar transmission line type consisting of two 10 μm wide transmission lines and a 70 μm gap. The emitters were fabricated by means of photolithography on a 500 μm thick semiconductor (100) GaAs substrate having a resistivity of 2.5×10^7 Ω cm. Thin layers (20 nm/100 nm) of Cr/Au were sputtered and the coplanar transmission line metallic pattern was transferred to the substrate via a lift-off process. A schematic of the two PC THz emitters' configuration is illustrated in Figure 1(b). While the two PC THz emitters were fabricated on the same semiconductor substrate, they are physically and electrically isolated from each other as shown in Figure 1(b). This is accomplished by cleaving each PC THz emitter chip and placing them in close proximity to within 200 μm. This arrangement guarantees that no transient current flows between the two PC THz emitters. Each PC THz emitter was biased independently via a 20 Vpp sinusoidal voltage waveform delivered from a waveform synthesizer. To eliminate the relative electrical waveform drift between PC THz emitters, the two waveforms' frequencies (f_1 and f_2) were phase-locked to each other.

Depicted in Figure 1(a) is the THz-PP-TDS system consisting of a four 90° off-axis gold-coated parabolic mirror arrangement each of 2.5 cm focal length. The PC THz emitters were illuminated by a Ti:sapphire laser pulses having durations of 20 fs, a central wavelength of 800 nm, and a repetition rate of 80 MHz. For generating the THz radiation pump and the THz radiation probe pulses and for probing the time-resolved electric fields, the 110 mW pulse train from the femtosecond laser was split into three pulse trains. A 20 mW pulse train was used to probe the THz pulses (referred to as an

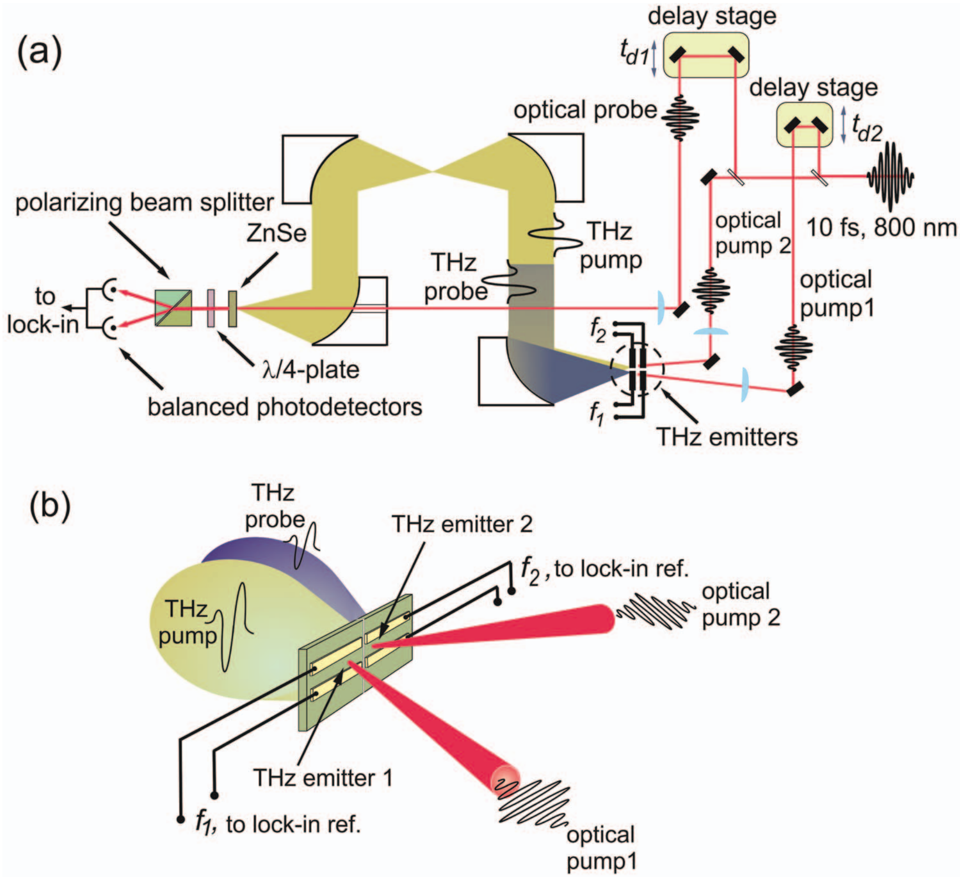

FIG. 1. (a) Schematic of the THz-PP-TDS system. The 10 fs, 800 nm laser pulse train is split into three pulse trains: An optical probe (20 mW), an optical pump 1(60 mW, generating THz pump pulse) and an optical pump 2 (30 mW, generating THz probe pulse). The THz-PP-TDS system consists of four 90° off-axis gold-coated parabolic mirrors, two PC THz emitters, and an electro-optic sampling setup. The generated THz radiation pump pulse and TH radiation probe pulse were collected and co-linearly focused along with the optical probe pulse onto a 500 μm thick ⟨111⟩ ZnSe electro-optic crystal. The THz radiation-induced polarization modulation of the optical probe was extracted via a balanced detection arrangement, and lock-in detection was carried out at the bias voltage modulation frequency. (b) Schematic of the THz emitters. The two PC THz emitters were placed near each at a separation of 200 μm. The bias voltages of the PC THz emitters 1 and 2 were modulated at frequencies f_1 and f_2, respectively. Through phase-locking at the modulation frequency of the biasing voltage, this configuration allows locking at and detecting the THz radiation due to the THz pump signal, or the THz probe signal, or both.

optical probe pulse train) and the other two (30 mW and 60 mW) were used to generate the THz radiation pump and probe pulses. The angle between these two optical pulse trains was set to 20°. To provide the temporal sampling scan of the THz pump pulse, the 60 mW optical pulse laser train was made to traverse a 15 fs resolution optical delay line driven by a stepper motor. Both the 30 mW and the 60 mW laser pulse trains were focused by two 5 cm focal length lenses to spot sizes of 10 μm each between the co-planar transmission line arms.

To perform the electro-optic sampling of the THz radiation electric fields, the generated THz radiation pump pulse and TH radiation probe pulse were collected and co-linearly focused along with the optical probe pulse onto a 500 μm thick ⟨111⟩ ZnSe electro-optic crystal.[10] The THz radiation-induced polarization modulation of the optical probe was extracted via a balanced detection arrangement, and lock-in detection was carried out at the bias voltage modulation frequency. Through phase-locking at the modulation frequency of the biasing voltage, this configuration allows locking at and detecting the THz radiation due to the THz pump signal, or the THz probe signal, or both. Complete information on the time-domain THz electric field amplitude and its phase were obtained by means of this setup.

III. RESULTS AND DISCUSSIONS

Typical electric fields of the THz radiation pump and probe pulses generated and detected by the above mentioned system are presented in Figure 2. Clearly, the presence of the electro-optic signature indicates that there is a good degree of spatial overlap between the two pulses and the femtosecond optical probe pulse. The temporal duration of the THz pulses presented is ∼1 ps. Here, the modulating frequencies of bias voltages applied to both PC THz emitters were set to 81 KHz and the delay between the THz radiation pump and the THz radiation probe pulses was fixed at 5.5 ps. For the purpose of performing THz-PP-TDS experiments, the energy fluence of the femtosecond optical laser pulse train incident illuminating the two THZ PC emitters was set so that the ratio of the electric fields between the two THz radiation pulses is 3:1. This ratio could be varied to any desired value by varying the energy fluence illuminating the emitters or by varying the biasing voltage applied to the emitters. In this THz-PP-TDS system, THz radiation fields' ratios of 1:1 down to 10:1 have been achieved.

Since the two PC THz emitters were separated by ∼200 μm, a major concern was the degree of spatial overlap

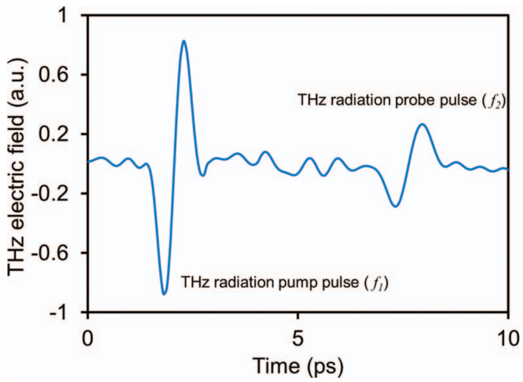

FIG. 2. Electric fields of the THz radiation pump and probe pulses as generated and detected by the abovementioned THz-PP-TDS system. The THz probe pulse is delayed by 5.5 ps with respect to the THz pump pulse. The modulating frequencies (f_1 and f_2) of bias voltages applied to both PC THz emitters were set to 81 KHz. The ratio of the THz pump to THz probe was set to 3:1 in this experiment.

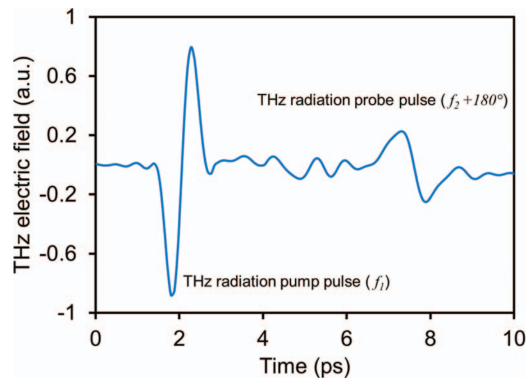

FIG. 4. Electric fields of the THz radiation pump and probe pulses where the polarity of the THz probe pulse is reversed by 180° with respect to the THz pump pulse. The THz probe pulse is delayed by 5.5 ps with respect to the THz pump pulse and the modulating frequencies (f_1 and f_2) of bias voltages applied to both PC THz emitters were set to 81 KHz. The ratio of the THz pump to THz probe was set to 3:1 in this experiment.

between the THz radiation pulse and probe pulse. Using the knife edge technique, both THz radiation pulse spatial profiles were characterized at the focal plane located between the two parabolic mirrors. Here, the edge of a sharp blade was scanned vertically and horizontally, at a 50 μm resolution and the transmitted THz radiation fields were measured as a function of the spatial translations. Using this technique, the spot sizes of both the THz radiation pump and probe pulses were mapped out. As shown in Figures 3(a) and 3(b), the overall full width at half maximum (FWHM) spatial extent of the THz radiation focal region was found to be 510 μm and 740 μm, along the *x*- and *y*-directions, respectively. Clearly, the spot size was slightly asymmetric due to the lateral displacement of the PC THz emitters. In the *x*-direction, both the THz radiation pulse spot and the THz radiation pulse probe were overlapped, whereas along the *y*-direction, the centers of the focal spots were displaced by 200 μm with 90% overlap.

It should be noted that the present THz-PP-TDS setup offers an additional capability to the time-resolved THz experiments, namely a control over the relative polarity between the THz radiation pump and THz radiation probe pulses. Figure 4 illustrates such a feature where by simply advancing the relative phase of the biasing voltages between the two PC THz emitters by 180°, the THz radiation probe pulse reverses its polarity. Such a feature is important for phase sensitive investigations.

Moreover, one of the challenges for THz-PP-TDS is the ability to perform THz time-resolved investigations at various THz radiation probe pulse polarizations. The ability to control the relative polarization angle between the THz radiation pump and probe pulses is essential for most all-THz pump-probe spectroscopy. Without sacrificing any THz radiation pulse power or employing of any optical components, this can be easily achieved with the current setup. The relative polarization state of the THz radiation pulses was attained by rotating one PC THz emitter while keeping the other fixed. The relative polarization state follows this rotation angle and, thus, any desired polarization can be set.

Using such a setup for THz-PP-TDS investigations requires illuminating a sample under study by the THz radiation pump pulse for excitation and intercalating the corresponding response on the THz radiation probe pulse as function of relative time delay. Thus, it is essential to modulate the biasing voltages of both PC THz emitters at different frequencies and phase-lock on one the PC THz emitter's modulation frequency. Such a requirement is easily met with the present THz-PP-TDS. Figure 5 illustrates such an application

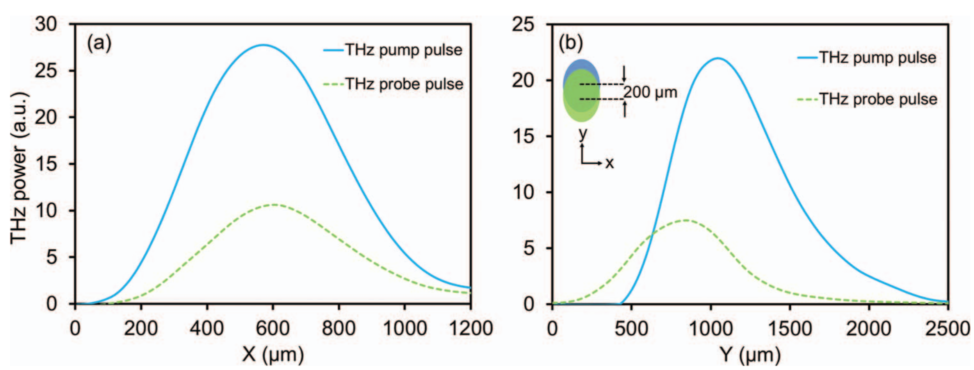

FIG. 3. The spot sizes of both the THz radiation pump and probe pulses were mapped out using the knife edge technique in the: (a) *x*-direction and (b) *y*-direction. The FWHM spatial extent of the THz radiation focal region was found to be 510 μm and 740 μm, along the *x*- and *y*-directions, respectively. In the *x*-direction, both the THz radiation pump pulse and the THz radiation pulse probe overlapped and along the *y*-direction, the centers of the focal spots were displaced by 200 μm with 90% overlap. The *x*- and *y*-directions are depicted in the inset of (b).

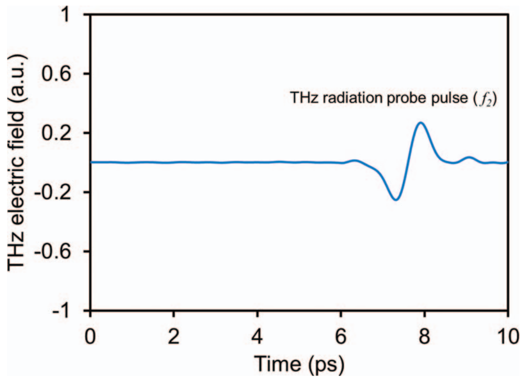

FIG. 5. The THz-TDS probe signal pulse acquired at 40.5 kHz. Here, the PC THz emitter generating the THz radiation pump pulse was modulated at $f_1 = 81$ kHz while the PC THz emitter generating the THz radiation probe pulse was modulated at $f_2 = 40.5$ kHz.

where the PC THz emitter generating the THz radiation pump pulse is modulated at $f_1 = 81$ kHz while the PC THz emitter generating the THz radiation probe pulse is modulated at $f_2 = 40.5$ kHz.

Notably, the choice of these frequencies represents the worst case scenario since f_1 is the second harmonic frequency of f_2. This situation is intended to test the performance capability of the THz-PP-TDS under unfavorable conditions. It can be clearly observed from Figure 5 that by phase locking at f_2, only the THz radiation probe signal is detected. This capability is especially essential for investigating phenomena for which the THz radiation pump pulse and the THz radiation probe are infinitesimally delayed in time or around zero time overlap. Moreover, the signal to noise ratio of the signal in this setup is very high (10^4:1) and thus any change, as long as it is above the noise floor, can be easily detected.

IV. SYSTEM APPLICATION

The capabilities of the THz-PP-TDS setup were confirmed by performing a time dynamics investigation of semiconductor-metal barrier experiments on a THz particle plasmon platform. For this, an ensemble consisting of subwavelength Cu particles having a mean diameter of 100 μm was utilized in the THz-PP-TDS setup. The Cu/Cu_xO interface forms a Schottky junction through creating a space charge region at the particles' surfaces. It has been hypothesized by Baron *et al.* that this space charge region can be modulated when the semiconductor-metal barrier is introduced to terahertz pulses.[11]

In this experiment, a pump THz pulse is utilized to excite the metallic ensemble (via near-field coupling of non-resonant particle plasmons), and the transient charge oscillations on the particle plasmons is probed at various delay times via a THz probe pulse. The ensemble was placed at the focus of two collinear THz beams with an amplitude ratio of 4:1 for the pump and probe pulses, respectively. The transmitted THz probe pulse was detected at delay times of $\triangle t = 0$ and 1 ps relative to the THz pump pulse. A total of four probe time-domain THz electric field signals were acquired and averaged for each time delay to obtain the final results. The results are further analyzed by comparing it to the situations where the THz pump beam is absent. The electric fields of the terahertz pump and probe pulse are 6 V/cm and 1.5 V/cm, respectively. During the course of the experiment, the PC THz emitter generating the THz radiation pump pulse was modulated at $f_1 = 81$ kHz, while the PC THz emitter generating the THz radiation probe pulse was modulated at $f_2 = 40.5$ kHz.

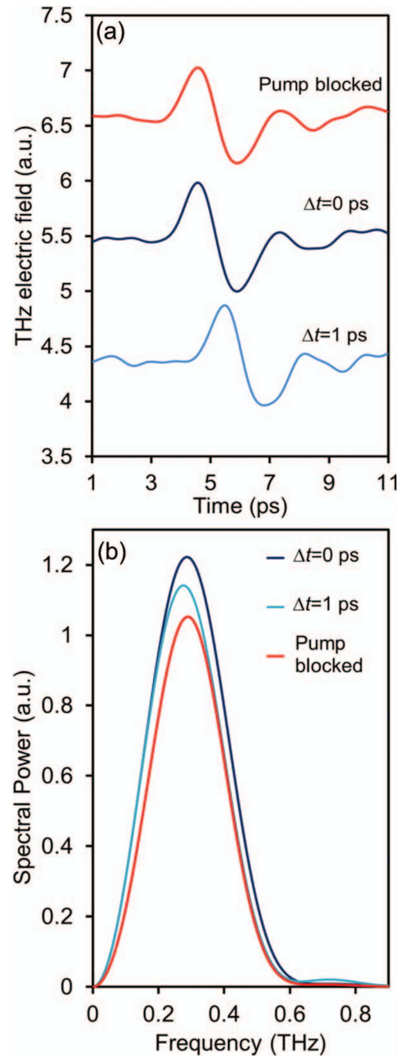

FIG. 6. (a) Time-domain THz probe signals transmitted through Cu/Cu_xO particle sample at various delay times ($\triangle t$), and (b) their corresponding spectral power plots.

Figure 6(a) depicts the time-domain electric field of the THz probe pulse transmitted through the Cu/Cu_xO particles at various time delays ($\triangle t$). The variation of the transmitted probe incident THz electric field as a function of $\triangle t$ can be better observed by exploring the change in the pulse' spectral power density through the evaluation of the Fourier transform of each signal presented in Fig. 6(a). These results are illustrated in Fig. 6(b).

It is evident from Figure 6(b) that the transmitted probe THz pulse power is the lowest when the probe pulse is transmitted through the sample while the pump pulse is blocked. On the other hand, the transmitted THz probe pulse power is the highest when the probe pulse is transmitted through the sample at the same time as the pump pulse (i.e., at $\triangle t = 0$).

For a better comparison of the probe pulse transmitted power, the total integrated spectral powers of each of the spectral power densities were utilized. When compared to the situation where the THz pump beam is blocked, the transmitted probe pulse power at $\triangle t = 0$ is enhanced by ~23%. Interestingly, the transmitted THz probe at $\triangle t = 1$ ps still exhibits an enhancement of ~10% compared to the situation where the THz pump pulse is blocked. This indicates that even at such a low THz pulse electric field, small changes of a few percent in the probe signal can be easily detected using the current THz-PP-TDS setup. These time-resolved experimental findings confirm the early report by Baron *et al.*[11] that a Schottky junction formed at the Cu/Cu_xO interface is modulated via the presence of the plasmonic surface currents generated by weak THz pulses. This alters the propagation of the THz probe beam as it is transmitted through the ensemble of Cu/Cu_xO sub-wavelength particles.

V. CONCLUSION

We have presented a versatile all-THz pump-probe spectroscopy system which is suitable for studying picosecond time scale phenomena. By utilizing two independent photoconductive THz emitters, the need for any specialized THz optical components, such as beam splitters, phase shifters, and polarization rotators were eliminated. This setup can be easily implemented with minimal alteration to conventional THz-TDS system. The possibility of controlling the electric fields splitting ratio between the THz radiation pump and probe pulses makes this system suitable for high resolution spectroscopy. The capabilities of producing in-phase and out-of-phase as well as polarization dependent pump-probe pulses make this system more versatile for many THz radiation coherent phenomena experiments. Furthermore, modulating the bias voltage of each of the PC THz emitters enables detecting the desired THz pump or the probe or both signals for various experiments. This permits the current THz-PP-TDS to measure signals even for infinitesimal time delay between the pump and the prob. This feature is important for solid state, chemistry, and biology related THz radiation spectroscopy.

ACKNOWLEDGMENTS

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