RAPID COMMUNICATION

Picosecond terahertz pump–probe realized from Chinese terahertz free-electron laser*

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Electron energy relaxation time τ is one of the key physical parameters for electronic materials. In this study, we develop a new technique to measure τ in a semiconductor via monochrome picosecond (ps) terahertz (THz) pump and probe experiment. The special THz pulse structure of Chinese THz free-electron laser (CTFEL) is utilized to realize such a technique, which can be applied to the investigation into THz dynamics of electronic and optoelectronic materials and devices. We measure the THz dynamical electronic properties of high-mobility n-GaSb wafer at 1.2 THz, 1.6 THz, and 2.4 THz at room temperature and in free space. The obtained electron energy relaxation time for n-GaSb is in line with that measured via, *e.g.*, four-wave mixing techniques. The major advantages of monochrome ps THz pump–probe in the study of electronic and optoelectronic materials are discussed in comparison with other ultrafast optoelectronic techniques. This work is relevant to the application of pulsed THz free-electron lasers and also to the development of advanced ultrafast measurement technique for the investigation of dynamical properties of electronic and optoelectronic materials.

Keywords: free-electron laser, ultrafast measurements, picosecond phenomena

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1. Introduction

Ultrafast pump and probe has been a powerful and popularly used optical technique in the investigation into, *e.g.*, dynamical properties of electronic and optoelectronic materials and devices.[1] With the development of femtosecond (fs) laser technology, nowadays it has become possible to achieve fs pump and probe system which has been widely used to study ultrafast dynamics of many electronic material systems.[2] It should be noted that at present, the most popularly used fs laser sources possess radiation wavelength of about 780 nm (*e.g.*, Ti:sapphire laser) and 1.56 μm (*e.g.*, fs fiber laser). Thus, the fs pump and probe system is often based on optical pumping and probing in the visible and infrared bandwidth.[3] Moreover, with the rapid development of terahertz (THz) technology, now we are able to achieve the ultrafast optical pump and THz probe (or OPTP)[4,5] based on the high-power fs laser pulses and THz time-domain spectroscopy (TDS).[6] In these OPTP systems, the optical pumping sources are often in the visible and infrared regime. From a view point of physics, most of electronic and optoelectronic materials (such as metals, semiconductors, oxides, superconductors, *etc.*) have electronic band gaps in the visible-to-infrared regime. As a result, the visible and infrared optical pumping can often result in the excitation of photo-induced carriers in the material systems. Hence, currently the fs pump–probe and OPTP are normally applied to the study of electronic dynamical properties relevant to photo-induced carriers and associated excitonic effects.[7]

As is well known, in the conventional metals, semiconductors, oxides, superconductors, *etc.*, the electronic kinetic energy, the Fermi energy, the shallow impurity binding energy, the phonon energy, the plasmon energy, *etc.* are normally on an meV/THz energy/frequency scale. The electrons in these materials can therefore interact strongly with THz radiation field via photo-assistant electronic momentum and energy excitation and relaxation due to electron coupling with scattering centers and to elementary electronic excitations.[8] Moreover, in these materials the electronic momentum and energy relaxation time, τ, are normally on a picosecond (ps) temporal scale.[9,10] Under THz irradiation, the condition $\omega\tau \sim 1$ can be satisfied for electrons in these materials, where ω is

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the THz photon frequency. Thus, THz radiation can modulate strongly the momentum and energy excitation and relaxation of electrons in electronic and optoelectronic materials. More particularly, because THz photon energy is much less than the electronic band gaps and band valleys of these materials, THz techniques can be applied to the study of electronic dynamical properties relevant to free-carriers within conduction or valance band. Hence, the most desirable experimental setup for the investigation of free-electron dynamics in electronic and optoelectronic materials should be the ps THz pump–ps THz probe system. It should be noted that the popularly used THz TDS[11,12] and THz pump–THz probe (or TPTP)[13] based on high power THz source generated via intense fs laser pumping on, *e.g.*, $LiNbO_3$ crystal, belong to ps THz pump and ps probe techniques. However, the THz sources generated via *e.g.*, fs laser pumping in these systems basically possess the nature of broadband. Thus, the results obtained from THz TDS and TPTP measurements can normally be analyzed in frequency-domain through Fourier transformation of the experimental data measured in time domain. If we can achieve monochrome ps THz pump and ps THz probe, we are able to study THz dynamics of free-electrons in a material directly in time domain, and thus to obtain some results which cannot be directly measured from THz TDS and TPTP. This becomes the prime motivation of the present research work. In this study, we intend to establish such an advanced optical measurement facility by using Chinese THz free-electron laser (CTFEL).

2. Facility parameters of the CTFEL

As is well known, the THz free-electron laser (FEL) can provide high-power, high-linear-polarization, high-degreeof-monochrome, and frequency-tunable THz laser radiation.[14] In particular, the CTFEL achieved a saturation output in August 2017[15] and now is ready for user application. In contrast to other THz FELs running presently, such as UCSB (USA), FELIX (Holland/UK), NovoFEL (Russia), FELBE (Germany), *etc*., the current facility of the CTFEL has some unique features of THz laser pulse structures. At present, the CTFEL can provide THz radiation with an average output power up to 50 W and with 1 THz–3 THz frequency tunability. Particularly, the THz laser beam from CTFEL possesses a macro- and micro-pulse structure (see Fig. 1). The repetition rate of the macro-pulses is about 1 Hz and the pulse width is tunable in a range from 0.2 ms to 1.2 ms. Thus, the duty ratio of THz macro-pulses for CTFEL is rather low (less than 0.1%), which implies that the CTFEL is a good pulsed THz laser source. The repetition rate of THz micro-pulses is 54.17 MHz (or the time spacing between two THz micro-pulses is about 18.5 ns). Importantly, the THz pulse width of CTFEL is tunable from 10 ps to 20 ps, which has been examined via *e.g.*, the autocorrelation measurement of the THz pulses from CTFEL (see Fig. 2). The peak intensity of single THz micro-pulse can reach up to the order of μJ. Hence, the CTFEL is an ideal THz laser source to achieve the monochrome ps THz pump–probe system in which the THz pumping frequency and intensity are both tunable.

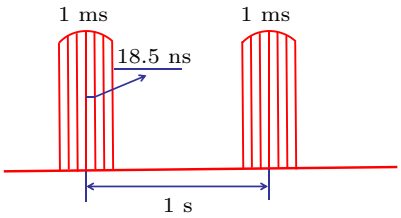

Fig. 1. Schematic diagram of THz macro- and micro-pulse structure of CTFEL.

3. Sensitive detection of ps THz pulses

One of the key technical issues to realize the ps THz pump–probe system using pulsed THz source for the study of a material is the detection of THz beams transmitted through or reflected from a sample in transmission or reflection experiment. In this study, we employ a highly sensitive THz detector realized from GaN heterojunction[16] for autocorrelation measurement (see Fig. 2) with a response width of about 100 MHz, while an InSb THz detector (QMC, UK) working at liquid helium temperature possesses 10-MHz response width for pump–probe measurement. Because the repetition rate of THz micro-pulses from CTFEL is 54.17 MHz, the GaN heterojunction detector (InSb detector) can be used to measure the profile of the micro-pulses (macro-pulses) transmitted through an experimental sample. Although the InSb detector cannot measure the single THz pulse with about 10 ps–20 ps pulse width, it is enough to be applied to the measurement in the ps THz pump–probe experiment where the THz signals induced by the micro-pulses can be detected with ps time delay and the detected signals are the results of the time accumulation.

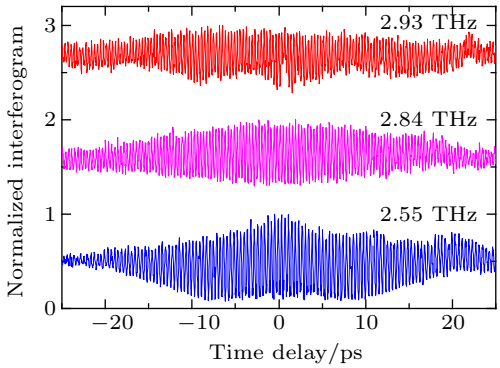

Fig. 2. Autocorrelation measurement of THz pulses of the CTFEL at three different frequencies. Together with Fourier transformation of these data, we can determine the width of THz pulses in both time- and frequency-domains.

4. Experimental setup

The principle and optical path for the ps THz pump–probe realized from the CTFEL is illustrated in Fig. 3. In the present

study, the facility is established for THz transmission measurement. (i) The CTFEL beam is split into two beams by using an Si wafer or a mylar film as a beam splitter. One beam with stronger intensity is for THz pumping and another with weaker intensity is for probing. (ii) The pump THz beam is focused on the surface of the sample. The intensity of the pump radiation can be changed via varying the CTFEL output power and/or, more conveniently, adding Si wafer or mylar films in the optical path to reduce the radiation intensity. (iii) The probe THz beam is first delayed via time delay stage then is focused at the same spot as the pump beam on the sample surface. The focusing spot of the probe beam on the sample should not shift as delay stage moves. Because the ps probe is required, the lengths of the optical paths for the pump THz beam and probe THz beam should be roughly the same and the difference between them should be within the range of the movement of the time delay stage (Thorlab, USA). (iv) The probe beam transmitted through the sample is focused on the sensitive THz detector for the probe measurement and the signals are recorded by an oscilloscope with 300 MHz band width (Tektronix MSO 3034). (v) The pump THz beam and the probe THz beam are focused on the same spot of the sample surface but in a small angle included between them. Thus, the pump beam can be blocked so that it cannot enter into the THz detector, in order to protect the THz detector from being optically damaged. (vi) In this experimental setup, we can measure the transmissions of the THz beam through the sample as a function of delay time at different THz pump intensities and frequencies. It should be noted the above scheme of the ps THz pump–probe system is similar optically to the nanosecond (ns) THz pump–ns THz probe system proposed and realized by NovoFEL (Russia).[17] Hence, by taking the advantage of THz pulse structure of the CTFEL, we are able to achieve a monochrome ps THz pump–ps THz probe system. To our knowledge, this is the first report on the realization of such an advanced optical measurement facility.

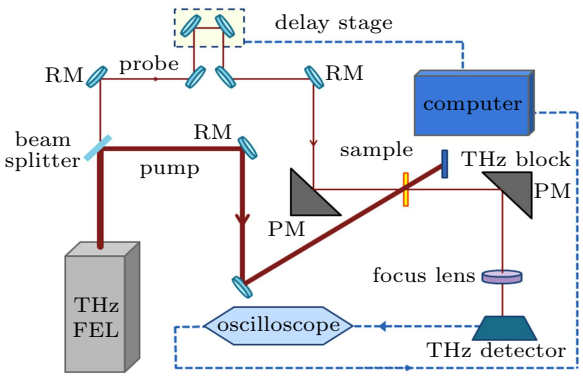

Fig. 3. Schematic diagram of ps THz pump–probe system based on CTFEL.

5. Results from measurement of GaSb

Using the ps THz pump–probe measurement facility realized from the CTFEL, in the present study we measure and examine the electronic dynamical properties of high mobility n-type GaSb. The GaSb possesses a single crystal structure of sphalerite, its lattice constant is 0.61 nm, and the effective electron mass is about $m^* = 0.04m_0$ with m_0 being the electron rest mass. Due to relatively small electron mass, GaSb normally has a high electron mobility.[8] The sample used in this study is grown by molecular beam epitaxy (MBE) and is weakly n-type doped during the growth. The III–V compound GaSb has a band gap of about 0.71 eV.[8] Thus, THz light radiation cannot generate photon-induced carriers in the sample system. The sample parameters of the GaSb wafer used in this study are as follows. i) The sample possesses a diameter of 5.0 cm; ii) The thickness of the sample is about 500 μm; iii) the incident surface of the sample is highly polished. And iv) the room-temperature electron mobility of the sample is about 2500 $cm^2/V{\cdot}s$, obtained via transport measurement.

In this study, we measure the intensity of THz transmission through the n-GaSb wafer as a function of delay time, respectively, for 1.2 THz, 1.6 THz, and 2.4 THz, at room-temperature and in free space. The width of the THz macro-pulse and micro-pulse are fixed respectively at about 0.9 ms and at about 20 ps. The transmission signals of the macro-pulses are recorded, where each delay time measured with a time spacing of about 30 fs by delay stage corresponds to the result from summing and averaging over 1000 points collected by the oscilloscope. We find that the excitation and decay signals cannot be markedly detected at 1.2 THz. This is because the incident surface of the sample is highly polished and, as a result, the reflection of THz probe beam from sample surface is very strong due to surface plasmonic effect.[18] The raw data of the experimental results are shown in Fig. 4. As we can see from Fig. 4, the excitation and decay phenomena become more pronounced with THz pumping frequency increasing. It should be noted that in contrast to time dependence of transmission in the fs pump–probe system,[5] the effect of electronic excitation can be clearly measured by using the ps pump–ps probe system here, which implies that the electronic excitation time and relaxation time are both on a ps time scale in n-GaSb at room-temperature.

Now we generalize the commonly used analytical approach for ultrafast pump and probe[19] to analyze the experimental data obtained from this study. Generally, the ps THz pump–probe system used in the present study is an optical pump (or input)→ sample → probe (or measurement)→ record (output) signal system. Here, i) the input signal, $f(t)$, corresponds to the detail of the pumping THz pulse (*e.g.*, pulse intensity, frequency, width, profile, *etc.*), ii) the response of the sample to the pumping light, $S(t)$, depends on the physical

and material parameters of the sample, iii) the probed signal transmitted via the sample, $R(t)$, is determined by the measurement system including detector, optical path, time delay stage, *etc*., and iv) the output signal, $O(t)$, is the overall contributions from the above signals mentioned in i), ii), and iii). Because this is a cascade system, the response function of the system,[19] $I(t)$, is the convolution of $S(t)$ and $R(t)$, while the output $O(t)$ is the convolution of $f(t)$ and $I(t)$. What we are to measure in the ultrafast pump–probe experiment via transmission measurement is basically the response function $I(t)$.[19] This is because in the measurement the pump and probe pulses coincide at $t = 0$, and the transmission signals are recorded for probe beam with the time delay during which the pump pulse is absent.

It should be noted that under THz pumping, the electronic excitation and relaxation in n-GaSb are achieved mainly through intra-band and intra-valley electronic transition channels or namely, through free-electron transition events. Thus, we can model the excitation-decay in pump–probe experiment by using a single exponential decay with a decay constant τ (or electronic relaxation time) via

$$S(t) = A_1 \mathrm{e}^{-t/\tau}. \tag{1}$$

We assume that the time resolution of the THz detection system (including detector, optical path, time delay stage, *etc*.) can be modelled by a Gaussian function with a full width half maximum as σ (or time resolution of the measurement system), which reads

$$R(t) = A_2 \mathrm{e}^{-t^2/(2\sigma^2)}. \tag{2}$$

As a result, the response function or the time-resolved transmission intensity in pump–probe measurement can be regarded as the conjugation between the instrumental response $R(t)$ and the response from the sample $S(t)$ via[19]

$$\begin{aligned} I(t) &= \int R(t-t')S(t')\mathrm{d}t' \\ &= I_0 + A\exp\left(\frac{\sigma^2 - 2t\tau}{2\tau^2}\right)\left[1 - \mathrm{erf}\left(\frac{\sigma^2 - t\tau}{\sqrt{2}\sigma\tau}\right)\right], \end{aligned} \tag{3}$$

where I_0 is the intensity of background transmission signal and erf(x) is the error function.

From Fig. 4, we see that the experimental data obtained from the ps THz pump–probe system for n-GaSb fit fairly well to Eq. (3). Through fitting the experimental results to the theoretical formula, we are able to determine the sample and measurement parameters. We find that $I_0 = 0.94$, $A = 3.8$, $\tau = 2.92$ ps, and $\sigma = 60.0$ ps at 1.6 THz and that $I_0 = 1.3$, $A = 19.2$, $\tau = 2.32$ ps, and $\sigma = 60.8$ ps at 2.4 THz.

It should be noted that like other THz FELs running at present, the CTFEL has output power varying with radiation frequency. In our measurements, the output power is about 10 W at 1.2 THz and 1.6 THz and about 25 W at 2.4 THz. The relaxation time $\tau = 2.92$ ps at 1.6 THz is slightly longer than $\tau = 2.32$ ps at 2.4 THz. This implies that under the intense THz FEL radiation, hot-electron effect or nonlinear electronic response can be achieved so that a shorter relaxation time can be measured at a stronger radiation intensity. In the presence of intense THz radiation and at room-temperature, the hot-electron effect can be induced by electron–phonon scattering.[8] Owing to this scattering mechanism, electrons gain the energy from radiation field and lose the energy via emission of phonons (or lattice vibrations). When the energy loss rate by phonon emission is smaller than energy gain rate via the absorption of photons, the electrons in the material can be heated, as a result, the electronic relaxation time decreases.

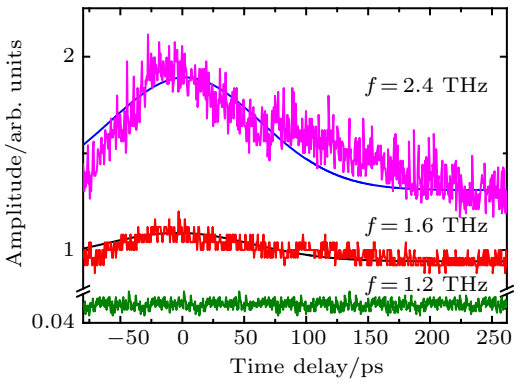

Fig. 4. Plots of transmission intensity *versus* delay time for different pumping frequencies as indicated. Smooth curves are the results given by Eq. (3) for 1.6 THz and 2.4 THz and coarse curves are the raw data of experimental results.

In the present study, we measure the transmission intensity of the sample. The electronic relaxation time determined experimentally should correspond to the energy relaxation time which measures the electronic energy loss rate induced by electron coupling with scattering centers such as impurities, phonons, surface roughness, *etc*.[20] The electron energy relaxation time τ_E for n-GaSb was measured optically via four-wave mixing (FWM).[21] It was found[21] that for an n-GaSb sample with electronic mobility 2540 cm^2/V·s at 77 K, the electronic momentum relaxation time is about $\tau_\mathrm{M} \sim 0.2$ ps at 300 K (obtained by optical absorption) and τ_E can be larger than 1.5 ps at 300 K. The $\tau_\mathrm{E} > 2.3$ ps obtained from this study is in line with the result determined by FWM experiment.[21] We mention further that the electron mobility for our sample (about 2500 cm^2/V·s at room-temperature) is larger than that used for FWM measurement. A longer τ_E for our sample can be expected.

We note that the full width half maximum σ measures the time resolution of the THz detection system in a Gaussian function like time response. It therefore depends not only on the detector itself but also on the intensity and frequency of the radiation field. We find that $\sigma = 60.0$ ps at 1.6-THz frequency

and 10-W power is slightly smaller than $\sigma = 60.8$ ps at 2.4-THz frequency and 25-W power. It implies that the better time resolution of our THz detection system can be reached with relatively lower radiation intensity. When σ does not depend on the radiation frequency nor intensity, the linear response of the detection system is achieved.

6. Further discussion

From Fig. 4, we note that the time decay signal obtained from transmission measurement via the ps THz pump–ps probe system is weaker than those via the fs pump–probe system[3] and OPTP[22] with 780-nm fs laser pumping. This is mainly because the ps THz pump–ps probe technique measures the consequence of the free-electron dynamics in an electronic material. The time decay signals are therefore induced mainly by electronic momentum and energy relaxation within the conduction band. In the sharp contrast case, the fs pump–probe and OPTP using 780-nm laser pumping can induce photo-excited carriers and associated excitonic effect. The opening up of inter-band electronic transition channels and excitonic relaxation in the presence of 780-nm laser pumping can result in more mechanisms for electronic relaxation. Thus, the stronger decay signals can be observed in fs pump–probe and OPTP measurements.

Because the width of the THz micro-pulses for the CTFEL is in a range of about 10 ps–20 ps, the essential condition under which the obvious excitation and decay phenomena can be observed in the ps THz pump–probe experiment is that the electronic energy relaxation time τ_{E} should be large, namely, high electron mobility sample is required. It has been demonstrated experimentally[21] that τ_{E} can be 10 times larger than τ_{M} for semiconductors such as GaAs and GaSb. Therefore, the ps THz pump and ps probe realized from the CTFEL can be applied to the investigation of dynamic properties of high mobility electronic materials such as semiconductors, two-dimensional electronic systems, superconductors, *etc*.

It should be noted that although the width of the THz pulse from CTFEL ranges from about 10 ps to 20 ps, they still can be applied to the measurement of the sample with several ps energy relaxation time in the pump–probe experiment. This is due to the fact that during the measurement of the transmission with the time delay of the probe THz beam, the pump pulse is absent. From Eq. (3), we see that the relaxation time in the response function is multiplied by the delay time. When the delay time is long enough, the decay signals can be measured.

The raw data shown in Fig. 4 look quite coarse. The major error items to the recorded transmission signals come from: i) the absorption of the THz pulse waves by the moisture in air because the measurement is conducted in free space, ii) the turbulence of the THz pulses during the measurement, and iii) the instability of the circuits in THz detector under intense THz irradiation. We find that the InSb THz detector and the GaN heterojunction detector used in the present study become saturated quickly with THz radiation power increasing and, as a result, the response of the detector to the radiation field becomes nonlinear. In this work, we try to measure the dependence of the transmission on THz radiation power by using high-resistance Si wafer (half transmission and half reflection) to attenuate the radiation intensity. However, when the THz radiation intensity is relatively low, we cannot obtain the reliable results because the signals are too noisy. The main reason behind this is that the THz detector used in this study responds nonlinearly to the THz radiation intensity. Furthermore, for fitting Eq. (3), we need to assum an exponential decay of the electronic relaxation process and a Gaussian-function-like time resolution of the THz detection system. These assumptions may not be fully true for the case of intense THz radiation, especially when the response of the THz detector becomes nonlinear and saturated.

7. Conclusions

In this work, we establish a monochrome picosecond THz pump–probe measurement facility by utilizing the special THz pulse structures of the Chinese THz free-electron laser (CTFEL). This technique can be applied to the investigation of free-electron dynamics of electronic and optoelectronic materials and devices. Compared with the fs pump–probe and OPTP using 780-nm laser pumping, this facility can measure the pure consequence of the momentum and energy excitation and relaxation of free-electrons, where almost no effects of photon-induced carriers and associated excitons are involved. Compared with the THz TDS and TPTP, this technique can provide monochrome THz pump and probe so that there is no need to perform the Fourier transformation of the measured data for analyzing the experimental results. Together with frequency tunability of the FEL, this facility can achieve selective radiation frequency for THz pump and probe. Therefore, this facility can be expected to possess wide applications.

We investigate the THz dynamical properties of high mobility n-GaSb and develop an analytical model for analyzing the experimental data. Through fitting the experimental data to the theoretical formula, we can obtain the electronic energy relaxation time τ_{E} for the sample. The obtained τ_{E} from this study is in line with that measured via *e.g*., four-wave mixing technique. Finally, it is noticed that at present, the CTFEL is at its preliminary stage for user applications. We hope that the results shown and discussed in this article can provide some inspiration for the application of the pulsed THz FELs and also for the development of advanced ultrafast measurement technique applied to the investigation of electronic and optoelectronic materials and devices.

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