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Anomalous behaviours of terahertz reflected waves transmitted from GaAs induced by optical pumping*

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Femtosecond pump-terahertz probe studies of carrier dynamics in semi-insulating GaAs have been investigated in detail for various pump powers. It is observed that, at high pump powers, the reflection peaks flip to the opposite polarity and dramatically enhance as the pump arrival time approaches the reflected wave of the terahertz pulse. The abnormal polarity-flip and enhancement can be interpreted by the pump-induced enhancement in the photoconductivity of GaAs and half-wave loss. Moreover, the carrier relaxation processes and surface states filling in GaAs are also studied in these measurements.

Keywords: terahertz, carrier dynamics, reflected wave
PACC: 7830, 7220, 4225B

1. Introduction

Carrier dynamics and relaxation processes in semiconductors have attracted much attention due to their wide physical interest and potential device applications. Since terahertz signals are very sensitive to carrier density and mobility, the pulsed terahertz system is a promising tool for obtaining information concerning the ultrafast carrier dynamics in materials.[1−14] In the past decade, much work has been done by using femtosecond pump-terahertz probe (FPTP) techniques to investigate nonequilibrium dynamics in photoexcited materials.[4−7] In these experiments, the time evolution of the carrier transport with picosecond resolution can be followed by changing the time delay between the pump and the terahertz pulses, and the time-resolved, frequency-dependent photoconductivity can be fully characterized.[4,5] While the FPTP experiments have been conducted on a variety of semiconductors,[4−6] and provide a more complete understanding of photoexcited carrier dynamics, very few studies treating the abnormal behaviours of terahertz reflected waves are found in the literature.

In this work, we have applied a developed FPTP technique to study the temporal development of the ultrafast dynamics of photogenerated carriers in undoped semi-insulating (SI) GaAs. The time resolved terahertz field amplitude radiated from the sample serves as a probe for the momentary charge acceleration in the sample. It is observed that the carrier relaxation time is correlated with photo-injected carrier density. Moreover, polarity-flip and abnormal enhancement behaviours in the terahertz reflected waves are also observed. These effects can be fully explained in terms of the dynamics of carrier transfer in GaAs. Picosecond time constants are found for these processes.

2. Experimental details

The experimental setup is illustrated in Fig.1. A pump-probe measurement was performed by using a Ti:sapphire regenerative amplifier delivering ultrashort optical pulses with a duration of 100 fs and a central wavelength of 800 nm at a repetition rate of 1 kHz. The output of the laser has an average power of 0.9 W,

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and is divided by beam splitters into three portions (pump, generation, and probe). Terahertz generation occurs through nonresonant optical rectification[15] of a visible pulse in a 1-mm-thick (110) ZnTe crystal (II-IV Inc.). The incident angle of the pump beam is 25° with respect to a ⟨100⟩ oriented undoped SI GaAs surface normal, the spot size is 4.0 mm in diameter, and the average pump power varies from 1.3 mW to 130 mW using an adjustable optical attenuator. The terahertz radiation is detected by free-space electro-optic sampling[16] in a 1-mm thick ⟨110⟩ ZnTe crystal with the probe pulse. The signal is collected with a lock-in amplifier phase-locked to an optical chopper which modulates the terahertz generation beam. The terahertz beam path from the transmitter to the receiver is purged by nitrogen to prevent absorption by atmospheric humidity.

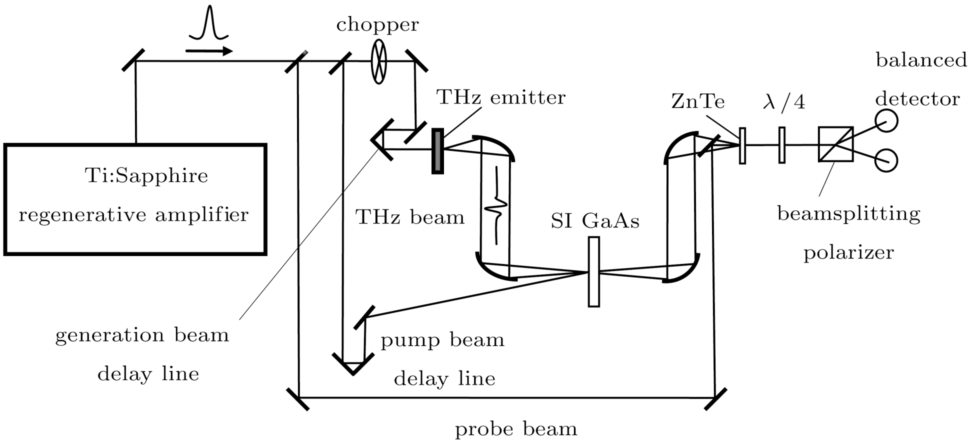

Fig.1. Experimental setup for the femtosecond pump-terahertz probe (FPTP) measurements.

3. Discussion and results

Since no significant pump-induced phase change of the terahertz pulse through the excited sample is observed, the decay dynamics can be described by the peak transmission. Figure 2(a) depicts the relative change of the terahertz peak transmission as a function of the delay time between the pump and the terahertz pulses for various pump powers. We can see that the pump-induced change of terahertz transmission increases with pump power. For GaAs the total measured $\varDelta T/T$ is proportional to the absorption coefficient α, which is related to the mobility of the photoexcited electrons.[4] The photoconductivity σ in this way is related to the differential transmission, $\varDelta T/T_0$, through[17]

$$\begin{aligned}\sigma &= (N+1)(Z_0\delta)^{-1}\frac{\varDelta T}{T}\\ &= (N+1)(Z_0\delta)^{-1}\left[\frac{1}{1-|\varDelta T/T_0|}-1\right],\end{aligned} \qquad (1)$$

where N is the index of refraction of the substrate, $Z_0 = 377\ \Omega$ is the impedance of free space, and δ is the pump penetration depth (taken to be 1 μm here, as for bulk GaAs).[4] The photoconductivity σ can also be given by

$$\sigma(t) = en(t)\mu(t), \qquad (2)$$

where $n(t)$ is the photoexcited carrier density, $\mu(t)$ is the carrier mobility, and e is the electron charge. We neglect the motion of holes because the effective mass of the hole is much larger than that of the electron. As seen from Eqs.(1) and (2), larger photo-injected carrier density or higher mobility may result in greater $|\varDelta T/T_0|$. At low photocarrier density, most of the electrons locate at the $\varGamma$ minimum, so we use a simple model in which carrier mobility μ is regarded to be time-independent. From Eqs.(1) and (2), the pump-injected carrier density can be extracted from Fig.2(a) by using

$$n(t) = \frac{N+1}{Z_0 e\delta\mu}\left[\frac{1}{1-|\varDelta T/T_0|}-1\right], \qquad (3)$$

as plotted in Fig.2(b). The inset of Fig.2(b) shows a nearly-linear relationship between the maximum-injected carrier density ($n_{\max}$) and the pump power.

In this way, from Eq.(3), there should be a significant non-linearity between the measured change in transmission ($|\Delta T/T_0|$) and the pump power. We also see that the carrier densities decrease after several hundred picoseconds, and this decrease is usually attributed to the carrier recombination.

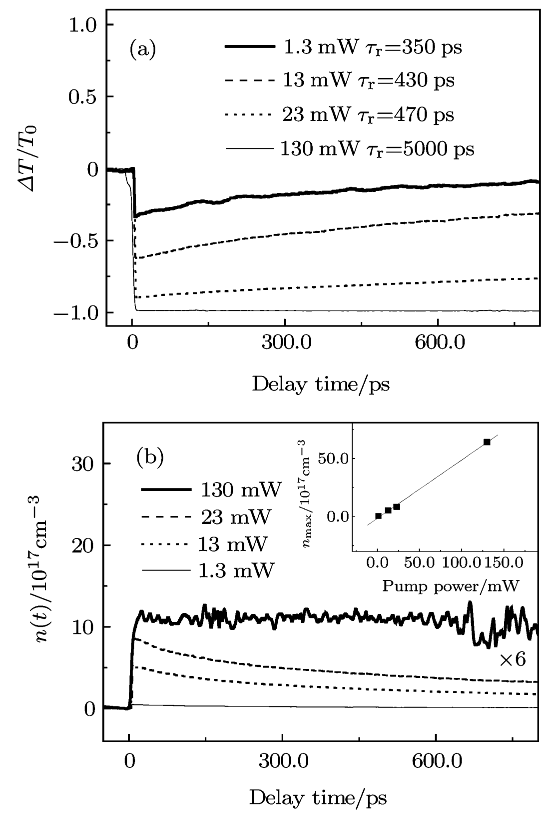

Fig.2. (a) Relative change in terahertz transmission ($|\Delta T/T_0|$) for various pump powers. (b) Time-dependent carrier density obtained from Eq.(3), the inset shows the maximum-injected carrier density ($n_{\max}$) as a function of pump power.

In Fig.2(a), it can also be seen that, at low excitation pump power (1.3 mW and 13 mW), the differential signal $\Delta T/T_0$ decays exponentially; as the pump power is increased (23 mW), the relaxation slows and exhibits a linear decay; at the highest pump power (130 mW), the decay has a slight outward curvature. Fitting an exponential function to the carrier decay curves, we extract the carrier relaxation time τ_r, as shown in Fig.2(a). The relaxation time is found to increase with pump power, from 350 ps to 5000 ps. This trend is expected for trap filling, which may inhibit carrier recombination.[18] Previously, Lloyd-Hughes *et al* have shown that passivating surface states on SI GaAs can lead to changes in carrier lifetime.[17] The results in our experiment are qualitatively similar. For undoped SI GaAs, after excitation at a photocarrier density of $n_{\rm exc} \sim 10^{17}$ cm^{-3} by the pump pulse, carriers could fill these surface states considerably and the carrier decay will then slow down.[18,19]

The measuring methods of the terahertz reflection pulses and main pulse are exactly the same. By measuring the terahertz waveforms, we can see that, following the main pulse, there is a relatively small pulse, which is the reflection pulse and is caused by the reflection in the GaAs layer. In Fig.3, we show the temporal terahertz waveforms, which contain the main and reflected waves, transmitted though SI GaAs at different pump-probe delay times for pump powers of approximately (a) 130 mW, (b) 23 mW, and (c) 1.3 mW. For each pump power, it can be seen that if the terahertz pulse is ahead of the pump pulse, i.e., the delay time is negative, the terahertz main wave is unchanged; when the delay times are positive, the transmitted terahertz main waves decrease due to the pump generated carriers in GaAs. However, it is surprising to see that, for each pump power, the reflected signals at negative and positive delay times exhibit an opposite polarity. For high pump powers (130 mW and 23 mW), as the delay time is 10 ps, the reflected waves increase dramatically, even much larger than the main waves; at the delay time of 100 ps, the reflected waves decreased distinctly.

In order to reveal more complete studies of the dynamics of carrier transfer in GaAs, we investigated the one-dimensional pump scans by fixing the time point of the terahertz beam at the main peaks and the reflection peaks, respectively, and changing the delay time of the pump beam,[10] as shown in Fig.4. At high pump powers, the reflection peaks flip to the opposite polarity and dramatically enhance as the pump arrival time approaches the reflected wave of the terahertz pulse; at the delay time of 15.2 ps, equal to the time space between the main and reflected waves (as shown in Fig.3), the reflection peaks recover to a small value, still showing the opposite polarity. However, at low pump power (1.3 mW), we observed that the reflected wave also flips over at positive delay times, but the peak value of the reflected waves only shows a reduction, instead of enhancement, and remains almost unchanged at large delay times.

The polarity-flip and enhancement of the reflected waves can be interpreted by the schematic of transmitted and reflected terahertz fields in the photoexcited

GaAs depicted in Fig.5. Since the main wave is ahead of the reflected wave for 15.2 ps, the pump pulse encounters the reflected wave first. Concentrating on the pump scans of reflection peaks, for the period of delay time from 0 to 15.2 ps, the pump pulse has already met the reflected wave but has not met the main wave yet, in this way, most of the terahertz field can transmit the GaAs surface (process I); as the reflected wave reflects at the photoexcited surface (process II),

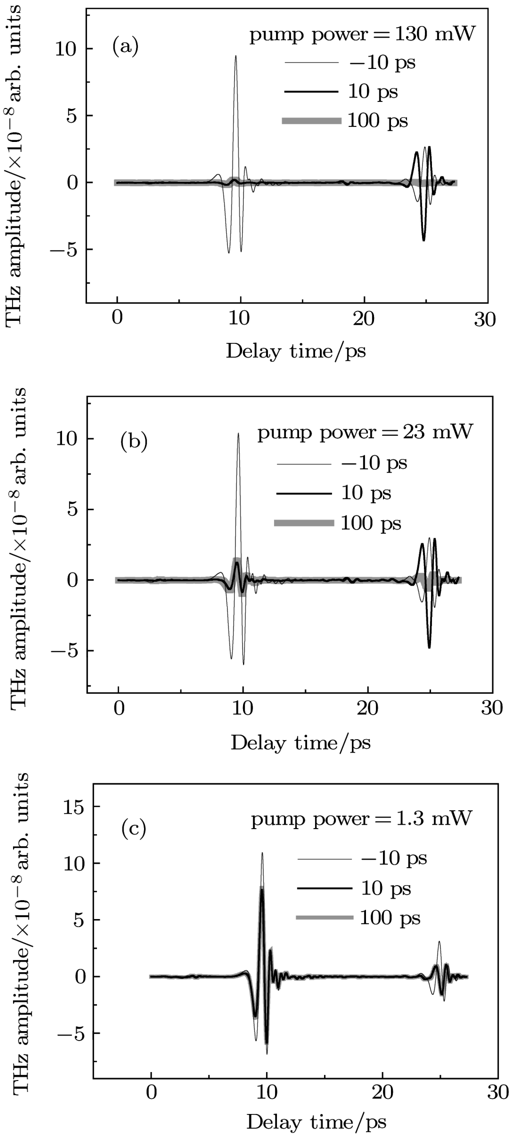

Fig.3. Main and reflected waves of terahertz waveforms transmitted though SI GaAs at different pump-probe delay times for pump powers of approximately (a) 130 mW, (b) 23 mW, and (c) 1.3 mW.

the pump pulse has already arrived, and the reflected wave will be affected by the photogenerated carriers in the surface layer. After excitation at a photocarrier density of $n_{\rm exc} \sim 10^{17}$ cm^{-3} by the pump pulse, from Eq.(2), the surface photoconductivity increases, and for the reflected wave, the photoexcited layer becomes a denser medium, the dielectric constant and

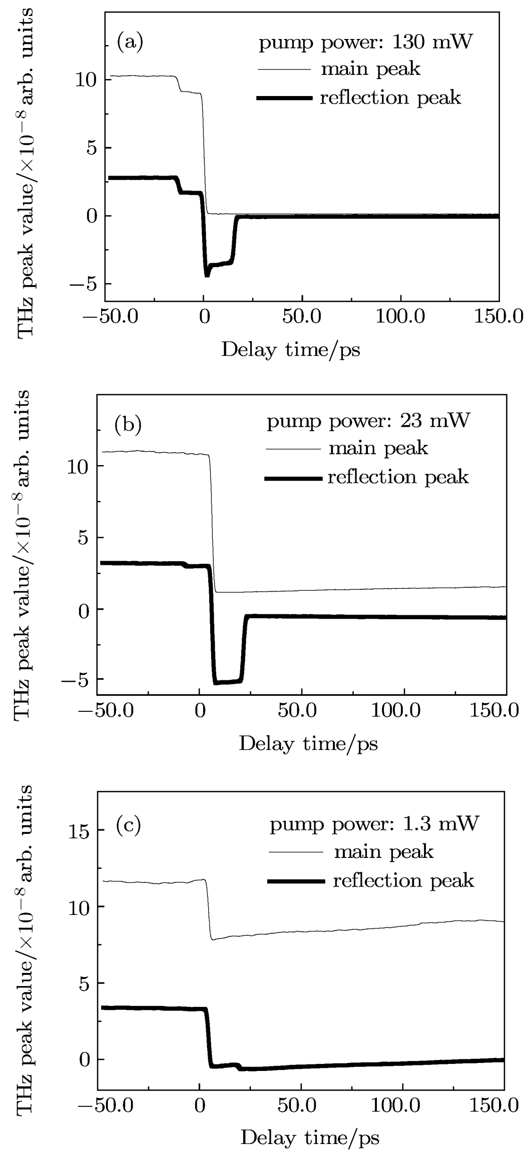

Fig.4. One-dimensional pump scans of terahertz transmission for both the main peaks and the reflection peaks at pump powers of (a) 130 mW, (b) 23 mW, and (c) 1.3 mW.

refraction coefficient of the layer increase too. As the reflected wave is incident on the photoexcited layer, more of the wave is reflected and less is transmitted. The reflected pulse has a much larger amplitude than that without pump excitation. When an incident wave strikes a boundary of a denser medium, 180° phase shift happens to the reflected wave, thus half-wave loss occurs, and the terahertz reflected wave flips over.

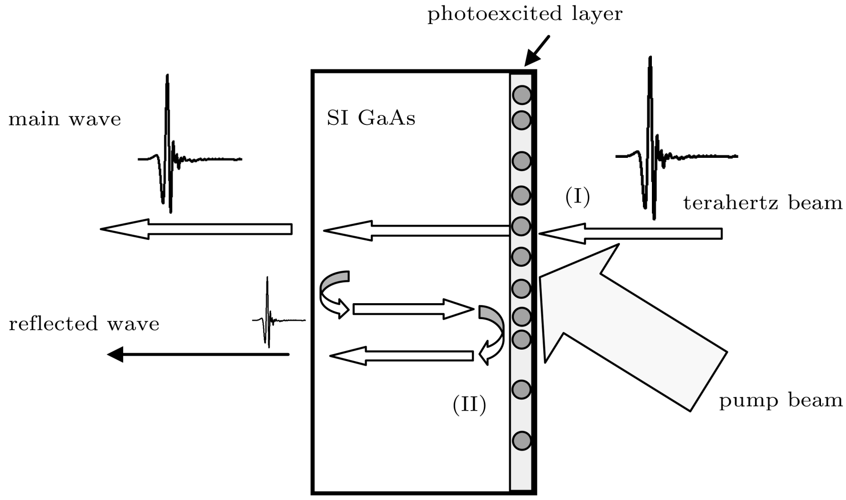

Fig.5. Schematic of transmitted and reflected terahertz fields in SI GaAs photoexcited by a pump beam. The signs of (I) and (II) denote two different processes: (I) indicates that the terahertz pulse transmits the GaAs surface, showing an inward direction; (II) indicates that part of the transmitted terahertz waves reflects at the GaAs surface.

At the delay time of 15.2 ps, the pump pulse reaches the main wave, less terahertz field can transmit the surface layer (process I), and thus the amplitude of both main and reflected waves decreases dramatically. However, for a pump power of 1.3 mW, only a few carriers are excited in the surface layer compared to that at high pump powers, so the enhancement and recovery of the reflected peak value cannot be seen at low pump powers.

4. Conclusion

In conclusion, we used a direct, noncontact method to investigate the influence of optical pump power on the photogenerated carrier relaxation process and effective mobilities. Abnormal reversal and increase of terahertz reflected waves transmitted from GaAs are observed. The measurements provide a direct insight into the ultrafast electronic transprot in SI GaAs. Since a terahertz probe is sensitive to the carrier density and mobility, this terahertz pump-probe technique may be a useful alternative approach for elucidating the ultrafast carrier dynamics in GaAs or other similar materials.

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