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Transient surface photoconductivity of GaAs emitter studied by terahertz pump-emission spectroscopy*

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This paper investigates the ultrafast carrier dynamics and surface photoconductivity of unbiased semi-insulating GaAs in detail by using a terahertz pump-emission technique. Based on theoretical modelling, it finds that transient photoconductivity plays a very important role in the temporal waveform of terahertz radiation pulse. Anomalous enhancement in both terahertz radiation and transient photoconductivity is observed after the excitation of pump pulse and we attribute these phenomena to carrier capture in the EL2 centers. Moreover, the pump power- and temperature-dependent measurements are also performed to verify this trapping model.

Keywords: terahertz, ultrafast carrier trapping, surface photoconductivity
PACC: 7830, 7210F, 7860

1. Introduction

Carrier dynamics and relaxation processes in semiconductors have attracted much attention due to wide physical interest and potential device applications. Since terahertz signals are very sensitive to carrier density and mobility, pulsed terahertz system is a promising tool for obtaining information concerning the ultrafast carrier dynamics in materials.[1−10] In the past years, much work has been done by using optical pump-terahertz probe techniques to investigate the photoinduced changes in the material photoconductivity, which contains the information of carrier density and mobility, with a temporal resolution of subpicosecond.[1,2] However, very few studies, which analyze the transient surface photoconductivity of terahertz emitters, are found in literature. In our previous paper,[8] we demonstrated that the carrier capture process in unbiased semi-insulating (SI) GaAs could be detected by a terahertz pump-generation technique. The measured capture time constant was found to be 20 picoseconds.

In this paper we further simulate the capture process by analysing the photo-induced change of surface photoconductivity of GaAs based upon the Hertz vector potential. Through theoretical modelling, we find that the terahertz waveforms are correlated with the transient photoconductivity. Experimentally, we also find that the time-resolved conductivity increases with pump-generation delay time and pump power, this fact is fairly consistent with our carrier capture model. Finally, the temperature-dependent measurement is also discussed.

2. Approximate analysis of the emitter's photoconductivity

According to classical electrodynamics, terahertz emission from (100)-oriented unbiased SI-GaAs results completely from a surface depletion field generation mechanism.[11] A model of the radiated field near the unbiased photoconducting emitter can be constructed from the Hertz vector potential with a plane wave approximation and the boundary condition of a current normal to the semiconductor surface.[11]

Figure 1 is the schematic diagram of an unbiased photoconducting antenna. The emitter is illuminated

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by an optical pulse at an incident angle of θ_1. The amplitudes of outward radiated field $\boldsymbol{E}_{\mathrm{r,out}}$, inward radiated field $\boldsymbol{E}_{\mathrm{r,in}}$, and transmitted radiated field $\boldsymbol{E}_{\mathrm{r,t}}$ can be expressed as[11]

$$\boldsymbol{E}_{\mathrm{r,out}} = \eta_2 \boldsymbol{J}_{\mathrm{s}} \frac{\sin\theta_1}{\cos\theta_1 + (n_2/n_1)\cos\theta_2}, \tag{1}$$

$$\boldsymbol{E}_{\mathrm{r,in}} = -(n_1/n_2)\boldsymbol{E}_{\mathrm{r,out}}, \tag{2}$$

$$\boldsymbol{E}_{\mathrm{r,t}} = t(\theta_2)E_{\mathrm{r,in}}, \tag{3}$$

where n_1 and n_2 are the indices of refraction of air and the sample respectively, η_2 is the impedance of the sample, θ_2 is the angle of refraction, $t(\theta_2)$ is the electric field transmission coefficient at the semiconductor/air

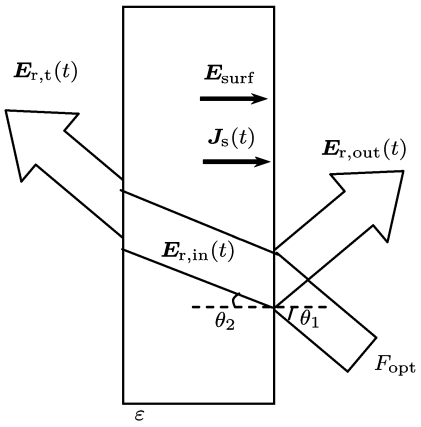


Fig.1. Radiated field from a photoconductor surface optically excited at an incident angle of θ_1.

interface, and $\boldsymbol{J}_{\mathrm{s}}(t)$ is the time-varying surface current density. From Eqs.(1) and (2), we can represent $\boldsymbol{J}_{\mathrm{s}}(t)$ as a function of $\boldsymbol{E}_{\mathrm{r,in}}(t)$ through

$$\boldsymbol{J}_{\mathrm{s}}(t) = -\frac{n_2}{n_1\eta_2} \cdot \frac{\cos\theta_1 + (n_2/n_1)\cos\theta_2}{\sin\theta_1} \cdot \boldsymbol{E}_{\mathrm{r,in}}(t). \tag{4}$$

Thus, the surface current $\boldsymbol{J}_{\mathrm{s}}(t)$ has the same time dependence as that of the inward radiated field $\boldsymbol{E}_{\mathrm{r,in}}(t)$ at the emitter.

From Ohm's law, the surface current $\boldsymbol{J}_{\mathrm{s}}(t)$ is given by

$$\boldsymbol{J}_{\mathrm{s}}(t) = \sigma_{\mathrm{s}}(t)(\boldsymbol{E}_{\mathrm{surf}} + \boldsymbol{E}_{\mathrm{r,in}}(t) \cdot \cos\theta_2), \tag{5}$$

where $\sigma_{\mathrm{s}}(t)$ is the surface conductivity of the emitter which is defined as[12]

$$\sigma_{\mathrm{s}}(t) = nq\mu = \frac{e(1-R)}{\hbar\omega} \int_{-\infty}^{t} \mathrm{d}t' \mu(t,t') \cdot I_{\mathrm{opt}}(t') \times \exp\left[\frac{-(t-t')}{\tau_{\mathrm{car}}}\right], \tag{6}$$

where e is the electron charge, R is the optical reflectivity of the illuminated surface of the photoconductor, $\hbar\omega$ is the photon energy, $\mu(t)$ is the carrier mobility as a function of time, and τ_{car} is the lifetime of the excited carriers. $I_{\mathrm{opt}}(t')$ is the optical pulse intensity, which can be expressed as

$$I_{\mathrm{opt}}(t') = I_0 \exp\left(-\frac{t'^2}{\tau_0^2}\right), \tag{7}$$

where τ_0 is the optical pulse duration.

It is important to note that $\mu(t)$ takes into account the relatively slow increase in the conductivity in comparison with that of a possibly rapidly varying optical intensity. Then we can use the mobility with the following form for the description of carrier transport processes when GaAs is excited by the ultrashort optical pulse[13]

$$\mu(t,t') = \frac{\mu_{\mathrm{f}} - (\mu_{\mathrm{f}} - \mu_{\mathrm{i}})\exp[-(t-t')/\tau_{\mathrm{m}}]}{1 + B\int_{-\infty}^{t} I_0 \exp(-t''^2/\tau_0^2)\exp[-(t-t'')/\tau_{\mathrm{car}}]\mathrm{d}t''}, \tag{8}$$

where τ_{m} is the carrier relaxation time of the initial hot carriers, μ_{i} is the mobility of carriers immediately after excitation, and μ_{f} is the final value of mobility in a time τ_{m}, the parameter B is introduced to include the effect of the decrease in the carrier mobility, which is caused by the electron-electron and electron-hole collisions in the case of high carrier density.[13]

From Eqs.(4) and (5), we can express the inward radiated field $\boldsymbol{E}_{\mathrm{r,in}}(t)$ at the emitter as

$$\boldsymbol{E}_{\mathrm{r,in}}(t) = -\frac{\sigma_{\mathrm{s}}\boldsymbol{E}_{\mathrm{surf}}}{\sigma_{\mathrm{s}} \cdot \cos\theta_2 + \dfrac{n_2}{n_1\eta_2} \cdot \dfrac{\cos\theta_1 + (n_2/n_1)\cos\theta_2}{\sin\theta_1}}. \tag{9}$$

Thus, as shown in Eqs.(6) and (9), as the carrier mobility increases, the surface conductivity increases, and thus, the inward and outward radiated fields are also predicted to enhance. As discussed below, the amplitude of the far-field radiated electric field should have the same dependence on carrier mobility.

By making use of Maxwell's equations and Fresnel laws, the transmitted electric field $\boldsymbol{E}_{\mathrm{THz}}(t)$ in the far-field limit is given by[14]

$$\boldsymbol{E}_{\mathrm{THz}}(t) = -\frac{A}{4\pi z\varepsilon_0 c^2}\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{J}_{\mathrm{s}}(t), \tag{10}$$

where ε_0 and c are the dielectric constant and the speed of light in the free space, A is the illuminated area of the emitter, z is the axial distance of the detector away from the emitter. Since the photoexcited carrier density is low in our experiment ($\sim 10^{15}$ cm^{-3}), the screening effect of carriers on the surface electric field is not dominant. In order to simplify the model, we assume that $\boldsymbol{E}_{\mathrm{surf}}$ is independent of time. Using Eqs.(4), (9) and (10), we can obtain the terahertz radiated field as

$$\boldsymbol{E}_{\mathrm{THz}}(t) = -\frac{A}{4\pi z\varepsilon_0 c^2}\cdot\frac{\mathrm{d}}{\mathrm{d}t}\left[\frac{\boldsymbol{E}_{\mathrm{surf}}\sigma_{\mathrm{s}}}{1+\dfrac{n_1\eta_2}{n_2}\cdot\dfrac{\sin\theta_1\cdot\cos\theta_2}{\cos\theta_1+(n_2/n_1)\cos\theta_2}\cdot\sigma_{\mathrm{s}}}\right] = -C\cdot\frac{\dfrac{\mathrm{d}\sigma_{\mathrm{s}}}{\mathrm{d}t}}{(1+D\cdot\sigma_{\mathrm{s}})^2}, \tag{11}$$

where

$$C = \frac{A\boldsymbol{E}_{\mathrm{surf}}}{4\pi z\varepsilon_0 c^2},$$

$$D = \frac{n_1\eta_2}{n_2}\cdot\frac{\sin\theta_1\cdot\cos\theta_2}{\cos\theta_1+(n_2/n_1)\cos\theta_2}$$

are constants. The negative sign means that the direction of the radiated electric field is opposite to the direction of the surface electric field.

By solving Eq.(11), we can also calculate the time-varying surface photoconductivity from the emitted terahertz waveforms by

$$\sigma_{\mathrm{s}}(t) = \frac{1}{D}\cdot\left(\frac{C}{C - D\int \boldsymbol{E}_{\mathrm{THz}}(t)\mathrm{d}t} - 1\right). \tag{12}$$

In this way, we can observe the dependence of the photoconductivity on the pump-generation delay time and pump power.

3. Experimental details

The experimental setup is illustrated in Fig.2. A pump-emission measurement was performed by using a mode-locked Ti:sapphire laser delivering ultrashort optical pulses with a duration of 100 fs and a central wavelength of 800 nm at a pulse repetition rate of 83 MHz. The output of the laser is divided by beam splitters into three pulses (pump, generation, and probe). The incident angles of generation and pump beams are 45° and 25° with respect to a (100)-oriented unbiased SI–GaAs surface normal, respectively. The terahertz radiation is detected by free-space electro-optic sampling[15] in a 1-mm thick (110) ZnTe crystal with the probe pulse. It is important to note that the signal is collected with a lock-in amplifier phase-locked to an optical chopper which modulates the terahertz generation beam, so that we can only detect the terahertz signal and the photoconductivity produced by the generation pulse. Therefore, the photoconductivity mentioned in this paper refers to the photoconductivity that originates from the generation beam, while that arising from pump-generated carriers cannot be detected. The terahertz beam path from the transmitter to the receiver is enclosed and vacuumized to prevent absorption by atmospheric humidity.

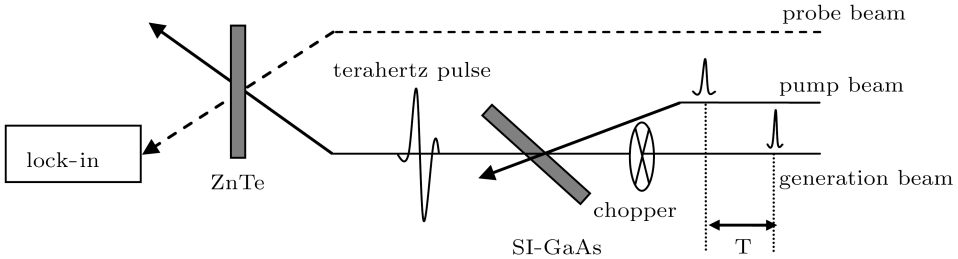


Fig.2. Experimental setup for the terahertz pump-emission measurements.

4. Results and discussion

To perform a comparison with experimental result, we first carry out the calculation according to experimental parameters. Using Eqs.(6), (7), (8), and (11), we obtain the temporal waveforms of radiated terahertz pulse emitted from unbiased SI-GaAs and LT–GaAs, as shown in Fig.3(a). Since the relaxation from satellite valleys will not happen, so it is reasonable to let $\mu_{\rm f} = \mu_{\rm i}$. Because of the very low excitation fluence used in the experiment, we assume $B = 0$. Curves 1 and 2 are for SI–GaAs ($\tau_{\rm car} = 100$ ps) with $\mu_{\rm f} = 4000\,{\rm cm}^2/{\rm V{\cdot}s}$ and $\mu_{\rm f} = 3000\,{\rm cm}^2/{\rm V{\cdot}s}$, respectively; curve 3 is for LT–GaAs ($\tau_{\rm car} = 1$ ps) with $\mu_{\rm f} = 2000\,{\rm cm}^2/{\rm V{\cdot}s}$. The inset shows the calculated time-varying surface photoconductivity of the emitters. We can see that the photoconductivity decays exponentially with a time constant $\tau_{\rm car}$, and both the surface photoconductivity and radiated terahertz signal increase with the increase of carrier mobility. For SI–GaAs, the radiated terahertz pulse is unipolar and the peak is symmetric. However, for LT–GaAs, following the main positive peak, there is a sharp and negative part, which is caused by the sudden drop in the photoconductivity, and this drop is due to the ultrafast recombination of photogenerated carriers in LT-GaAs.

Figure 3(b) shows the waveforms of terahertz radiation from SI–GaAs observed at different pump-generation delay times. Compared with the calculated waveforms, the detected terahertz signals are broadened and have two small negative peaks beside the main positive peak. This distortion is primarily caused by the screening effect on the surface field and the system response function, and the latter originates from the propagation of the terahertz field through the whole terahertz system.[16]

Using the data of emitted terahertz waveforms at different pump-generation delay times, we can calculate the time-resolved surface photoconductivity by using Eq.(12). Figure 4(a) depicts the calculated peak conductivity of SI–GaAs emitter as measured via terahertz pump-emission spectroscopy. We observe that both the terahertz signal (shown in Fig.3(b)) and the photoconductivity increase remarkably after the pump excitation. Note that the optical chopper only modulates the generation beam, so that the terahertz signal and photoconductivity arising from pump-induced carriers cannot be detected by the lock-in amplifier at all. In this way, we can only attribute this abnormal enhancement to the increased photocarrier mobility, as indicated by the calculated results in Fig.3(a).

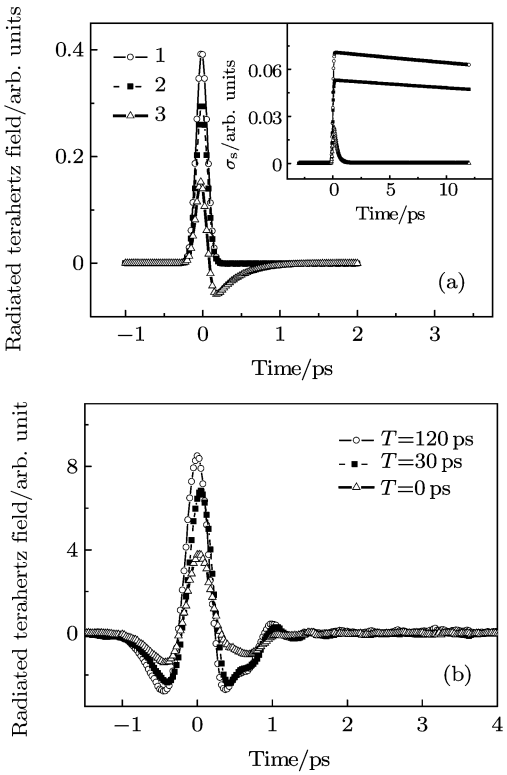


Fig.3. (a) Theoretic waveforms of radiated terahertz pulses emitted from SI- and LT-GaAs calculated from Eqs.(6), (7), (8), and (11); the inset shows the corresponding calculated time-varying surface photoconductivity of the emitter. (b) Waveforms of terahertz radiation observed at different pump-generation delay times.

For undoped SI-GaAs, the ionized EL2 deep centers strongly scatter carriers and reduce the carrier mobility with their powerful Coulomb field. They are also known as the dominant deep electron traps and have a capturing cross section of 10^{-13} cm^2.[17] After excitation by the pump pulse, the pump-injected carriers could be trapped effectively. This process neutralizes the ionized scattering centers. The carriers generated by the later coming generation pulse will then suffer less scattering due to the decrease of the Coulomb field of the EL2 deep donors after trapping electrons, and thus have higher mobilities. Correspondingly, the conductivity increases too. From Eq.(6), the conductivity is related to ionized impurity density $N_{\rm d}^+$ through

$$\begin{aligned}\sigma_{\rm s} &\sim \mu \sim \gamma^{-1} \sim (\gamma_{\rm imp} + \gamma_{\rm ap} + \gamma_{\rm op})^{-1} \\ &= \{a \cdot N_d^+ \cdot T^{-3/2} + b \cdot T^{3/2} \\ &\quad + c \cdot [({\rm e}^{h\nu_l/k_0T} - 1)]^{-1}\}^{-1}, \end{aligned} \tag{13}$$

where γ is the overall carrier scattering rate, $\gamma_{\rm imp}$,

γ_{ap}, γ_{op} are ionized impurity, acoustic phonon, and LO phonon scattering rate, respectively. The ionized impurity density N_d^+ decays exponentially with the pump-generation delay time at the capture rate. The solid curve in Fig.4(a) is the calculated data with an EL2 center trapping time of 20 ps.

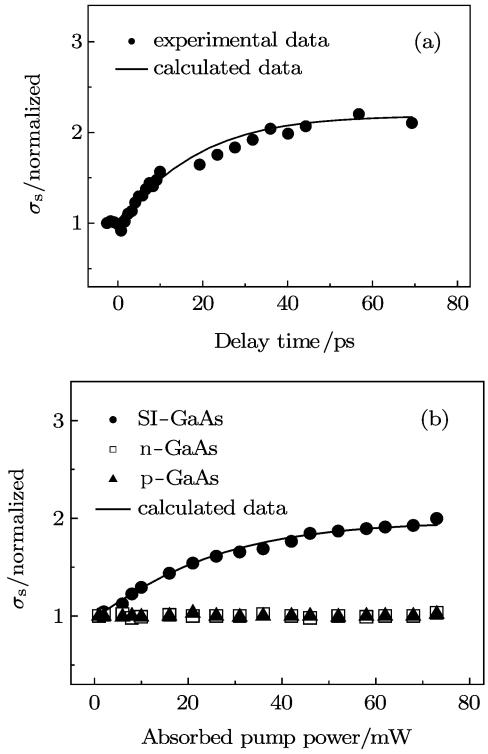


Fig.4. (a) Time-resolved conductivity of SI-GaAs emitter as measured via terahertz pump-emission spectroscopy with a pump power of 180 mW. (b) Conductivities of SI- (dot), n- (square), and p- (triangle) GaAs samples as a function of absorbed pump power at the delay time of 120 ps.

In addition, we observe that, for SI-GaAs, the radiated terahertz signal increases with pump power; but in the case of n- and p-type GaAs, the signal keeps unchanged with pump power (not shown), agreeing well with the observation reported previously by Nakajima and Hangyo.[18] Using the same method as that in Fig.4(a), we extract the conductivity of the emitter as a function of the pump power, as indicated in Fig.4(b). The solid curve is the calculated data by using the capture model. As the pump power increases, the EL2 centers will be filled more effectively by the pump injected photocarriers, and then the carriers generated by the generation pulse will get much higher mobilities and lead to much larger conductivity. However, this phenomenon does not exist in n- and p-type GaAs because the doping density (5×10^{17} cm^{-3} and 3.5×10^{17} cm^{-3} for n- and p-type GaAs, respectively) is much larger than the EL2 ionized impurity density N_d^+ ($\sim 10^{15}$ cm^{-3} at 300 K) and the capture effect is shielded.

The following temperature-dependent observations support our interpretation that ultrafast trapping accounts for the increased terahertz radiation. In Fig.5, as the temperature decreases, the terahertz waveforms slightly sharpen and increase in amplitude.[19] However, at the same pump-generation delay times of 120 ps, the pump-induced enhancement in terahertz radiation at 77 K is less than that measured at 300 K. This behavior may be attributed to the different ionization degree of the impurity in the sample, with a low ionization degree at 77 K and a high ionization degree at 300 K. As a result of the relatively low density of trap site at 77 K, EL2 centers cannot trap carriers as efficiently as they are at 300 K, leading to a smaller increase of terahertz emission.

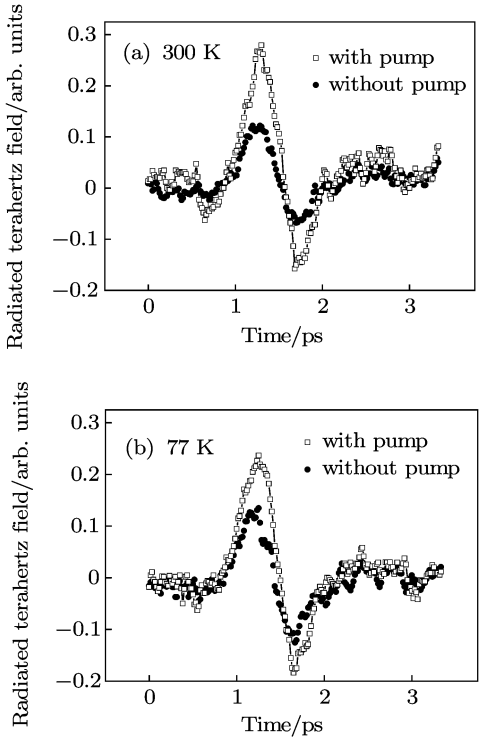


Fig.5. Waveforms of terahertz radiation observed without (dot) and with (square) the pump excitation at (a) 300 K and (b) 77 K at the delay time of 120 ps. Both the pump and generation beam have an average power of 130 mW, and this measurement is performed in a variable temperature cryostat.

5. Conclusion

In conclusionof this paper, we have fully studied terahertz radiation properties and transient surface

photoconductivity of SI-GaAs, by making use of a theoretical model to extract photoconductivity from terahertz waveforms. We found that the temporal waveforms have close relations with the photoconductivity and specific carrier transport processes. Our study shows that this model should be an ideal way to investigate the ultrafast carrier transport in semiconductors.

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