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Anomalous enhancement of terahertz radiation from semi-insulating GaAs surfaces induced by optical pump

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Femtosecond pump-generation studies of terahertz emission from semi-insulating GaAs surfaces have been investigated in detail. By changing the time delay between the pump and the generation pulses, we can follow the time evolution of the carrier transport with picosecond resolution. Abnormal increase in the amplitude of terahertz radiation subsequent to the excitation of pump pulse is observed, and it arises primarily from carrier capture in the trapping states with a time constant of about 20 ps. Furthermore, the corresponding amplitude spectrum, pump power-dependent measurements, and temperature-dependent measurements are also discussed. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2338805]

Measurements of the dynamics of hot electrons in semiconductors are extremely important for advancing the physics of nonequilibrium phenomena in these materials. Since terahertz signals are very sensitive to carrier density and mobility, pulsed terahertz systems are a promising tool for obtaining information concerning the ultrafast carrier dynamics in semiconductors.[1–10] In the past decade, several groups used terahertz time-domain spectroscopy systems to study the nonequilibrium electronic transport in semiconductors, such as the screening dynamics of local electric field,[1–3] magnetic field dependence or temperature dependence of carrier dynamics,[4–7] intervally scattering processes,[8,9] coherent phonons effect,[10] etc. While all these studies have provided a more complete understanding of carrier dynamics in semiconductors, very few results, which treat the processes of carrier capture in semi-insulating (SI) GaAs surfaces, are found in literatures.

In this letter, we have applied a developed pump-generation technique to study the temporal and spectral developments of the ultrafast dynamics of photogenerated carriers in undoped 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, after a few picoseconds following the incidence of the pump pulse on the emitter, the terahertz radiation is obviously enhanced. We propose that this abnormal behavior is due to carrier trapping by the EL2 native defect deep donors. In addition, the corresponding amplitude spectrum, pump power-dependent measurements, and temperature-dependent measurements are also discussed to reveal the more complete studies of the dynamics of carrier transfer in the GaAs surfaces.

The experimental setup is illustrated in Fig. 1. A pump-generation 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 was 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 undoped SI GaAs surface normal, and the spot sizes are 1.5 and 4.0 mm in diameter, respectively. Each beam has an average power of 180 mW, corresponding to an initial carrier density of approximately 10^{16} cm^{-3}. The terahertz radiation is detected by free-space electro-optic sampling[11] 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, so that only the terahertz signal produced by the generation pulse is detected, while the pump pulse merely injects carriers into the semiconductor. The terahertz beam path from the transmitter to the receiver is enclosed and vacuumized to prevent absorption by atmospheric humidity.

In Fig. 2, we show the temporal wave forms of the radiated terahertz pulse from the SI GaAs at a series of pump-generation delay times. The inset shows the peak amplitude of the radiated terahertz pulse as a function of delay times with a pump power of 180 mW, and the solid curves are calculated from the simple model to be described in this letter. From this figure, we can see that the signal obviously decreases at a short positive delay of subpicosecond, when the field screening induced by the pump pulse is predominant. The screening process usually takes place in a time scale of several picoseconds following the optical excitation,[12] but only dominates terahertz signal during the first picosecond of the pump-generation delay time. However, following the initial signal reduction, there is a remarkable increase of the signal.

According to the classical electrodynamics, terahertz emission from (100)-oriented GaAs results completely from a surface depletion field generation mechanism,[13] and the radiation amplitude can be deduced from $E_{\mathrm{THz}} \sim dJ/dt$. Here, $J=-env$, where $-e$, n, and v are the electronic charge, photocarrier density excited by the generation beam,[12] and ve-

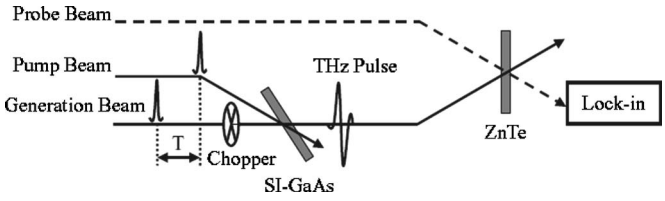

FIG. 1. Experimental setup for the pump-generation measurements.

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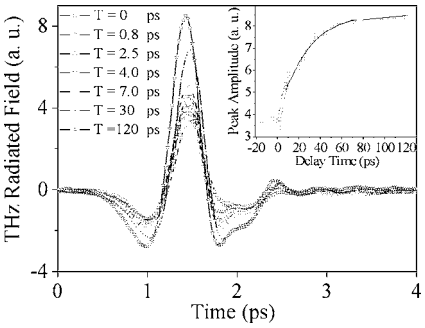

FIG. 2. Wave forms of terahertz radiation observed at a series of pump-generation delay times. The inset shows the peak amplitude of the radiated terahertz pulse as a function of the delay time; the solid curve is the calculated data using the model described in the text. To obtain this agreement, we use a trapping time constant of 20 ps.

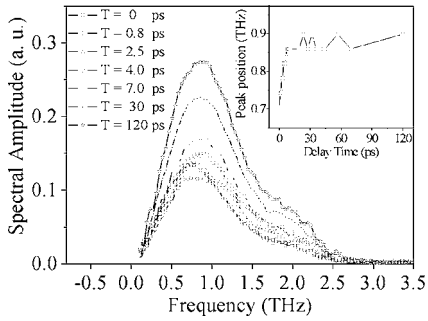

FIG. 3. Fourier spectra of the terahertz pulses at a series of pump-generation delay times deduced from Fig. 2; the inset shows the peak position of the emission spectra as a function of delay time.

locity of electrons, respectively. Thus we can speculate that terahertz field is proportional to the carrier acceleration.

The motion for the photoexcited carriers is treated with a Drude approximation, in which the carrier velocity is governed by

$$m^* dv/dt = -eE_{\mathrm{surf}} - m^* v/\tau, \tag{1}$$

where m^*, E_{surf}, and τ are the effective electron mass in GaAs ($0.067m_0$), built-in surface electric field, and momentum relaxation time, respectively. We neglect the motion of holes because the effective mass of hole is much larger than that of electrons. Imposing the initial condition $v(t=0)=0$, we then find for the carrier acceleration as

$$a(t) = (eE_{\mathrm{surf}}/m^*)\exp(-t/\tau). \tag{2}$$

For this configuration, the radiated terahertz field is given by

$$E_{\mathrm{THz}}(t) \sim E_{\mathrm{surf}} \exp(-t/\tau). \tag{3}$$

With respect to Eq. (3), there are two possible origins of the observed terahertz radiation enhancement: increased built-in surface field E_{surf} and reduced momentum relaxation rate τ^{-1}. The former reason can be ruled out because the surface field E_{surf} is always lowered by the space-charge field generated by the separating electron-hole pairs.[1,2] Therefore, we can only attribute the enhancement of terahertz radiation to an enhanced $\exp(-t/\tau)$ term, which can be caused by a reduced momentum relaxation rate. This means that the energy-dependent momentum relaxation process due to carrier scattering in the GaAs is slowed down, so that more energy can be coupled into tetrahertz radiation process.

We expect many different scattering processes in GaAs. For example, scattering occurs from impurities, heavy holes, acoustic phonons, and LO phonons. The momentum relaxation time τ can be obtained through

$$\begin{aligned}\tau^{-1} &= \tau_{\mathrm{imp}}^{-1} + \tau_{\mathrm{ap}}^{-1} + \tau_{\mathrm{op}}^{-1} \\ &= aNT^{-3/2} + bT^{3/2} + c[(e^{h\upsilon_l/k_0T} - 1)]^{-1},\end{aligned} \tag{4}$$

where τ_{imp}^{-1} is the ionized impurity scattering rate, τ_{ap}^{-1} is the acoustic phonon scattering rate, τ_{op}^{-1} is the the LO phonon scattering rate, and N is the ionized impurity density. The momentum relaxation process is strongly affected by the ionized impurities and defects in the GaAs, such as EL2 native defect deep donors (located approximately 0.8 eV below the conduction band edge).

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.[14] After excitation at a photocarrier density of $n_{\mathrm{exc}}=10^{16}$ cm^{-3} by the pump pulse, carriers could be trapped at a high speed. This process neutralizes the ionized scattering centers, thus the ionized impurity concentration N is reduced considerably. The carriers generated by the later coming generation pulse will suffer less scattering due to the decrease of the Coulomb field of the EL2 deep donors after trapping electrons, and thus have a higher mobility. Also, the neutralization could lead to a redistribution of the local electric field, which will be discussed in the future work. Consequently, tetrahertz radiation enhancement from the GaAs surfaces could be observed.

Because of the capture effect, the ionized impurity density N_d^+ varies with the pump-generation delay time T as

$$N_d^+(T) = N_d^+(0)\exp(-\beta T) = N_d^+(0)\exp\left(-\frac{T}{\tau_c}\right), \tag{5}$$

where β is the capture rate, which is related to the pump power, and τ_c is the trapping time of the EL2 centers, respectively.

Equations (3)–(5) give full information about the radiated tetrahertz pulse and the carrier dynamics. By adjusting the scattering time τ and the trapping time τ_c, we obtained very good agreement between the measured tetrahertz signal and the tetrahertz field obtained from those equations, as shown in the inset of Fig. 2. We found that a trapping time of 20 ps reproduced the measurements. For undoped SI GaAs, ionized EL2 deep centers are usually known as the dominant deep electron traps and have a concentration of 2.8×10^{16} cm^{-3} (Ref. 15) and an ionization degree of 10% at room temperature. Based on the approximation of thermal velocity of electrons $\upsilon \approx 10^8$ cm/s, trapping time $\tau_c \approx 20$ ps, we obtained an electron capture cross section of $\sigma_s \sim 10^{-13}$ cm^2. We note that the data in the literature are also of this order.[14]

The highly resolved and reproducible corresponding Fourier spectra are also observed to change with the delay time T as indicated in Fig. 3; the inset shows the peak positions of the emission spectra as a function of delay time. We can see that the peak amplitudes of the spectra have very similar behaviors as that in the time-domain spectroscopy; however, the peak positions of the emission spectra behave quite differently. The spectrum moves to higher frequency slightly within the first 7 ps and then stops moving for larger

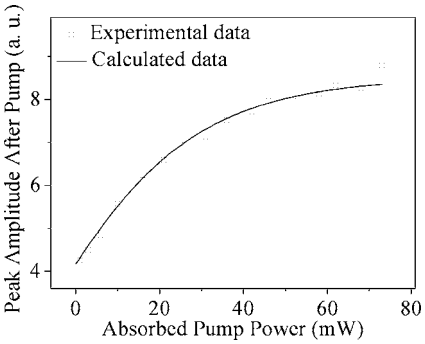

FIG. 4. Peak amplitude of the radiated terahertz pulse as a function of the pump power absorbed by the GaAs. The solid curve is the calculated data.

delay times. This result can be interpreted as follows. Since the tetrahertz radiation increases with a time constant of 20 ps, while the spectrum moves to the higher frequency within 7 ps, the conclusion is what decides the phase of the spectra is quite different from what decides the amplitude of tetrahertz signals. We propose that in the spectrum, it is no longer carrier trapping process, but the field screening that decides the maximum value position. After optical excitation, electron-hole separation due to field-induced carrier drift currents rapidly decreases the surface field. This fast screening of the initial surface field will lead to a sudden decrease of the depletion width, and the rate at which the strength of the surface depletion field changes with respect to a change in the relative distance will increase. Thus the carrier will have a faster change of acceleration and cause an enhanced high-frequency component in the spectrum. Consequently, the Fourier spectrum of the tetrahertz signal must be broadened and shifts towards a higher frequency. However, at longer time delays the surface field will reach a quasiequilibrium, which is governed by the balance between the drift and diffusion currents, and thus the spectrum's peak position will stop moving. Such nonequilibrium carrier transport dynamics can also be confirmed by Monte Carlo simulations.[12]

In order to investigate the influence of the pump pulse in detail, we measured the pump power dependence of the tetrahertz radiation signal at the pump-generation delay time of 120 ps, as shown in Fig. 4. Note that the radiated tetrahertz signal increases with the absorbed pump power by the GaAs. This increase can also be explained by the carrier capture model and this phenomenon agrees with the observation reported previously by Nakajima and Hangyo.[16] As the pump power increases, the EL2 centers can be filled by the pump injected photocarriers more effectively, and then the carriers generated by the generation pulse would get a much higher mobility and exhibit much larger tetrahertz signal. Applying this model by using Eqs. (3) and (5), we calculated the peak tetrahertz signal as a function of the carrier density excited by the pump pulse, which is proportional to the pump power absorbed by the GaAs. The solid curves in Fig. 4 are calculated data. However, this enhancement was not observed in the case of *n*-type and *p*-type GaAs.[16] This is because the EL2 ionized impurity density N_d^+ ($\sim 10^{15}$ cm^{-3} at 300 K) is small compared to the doping density (5×10^{17} and 3.5×10^{17} cm^{-3} for *n*- and *p*-type GaAs, respectively) and the capture effect is shielded.

In contrast to the previous observation by other groups,[1,2] which only observed the field screening effect, our result is quite different. The most likely explanation for this is the difference of the material properties and experimental conditions, such as EL2 deep centers' concentration and the applied field. In our unbiased sample, carrier capture is the dominant mechanism because of the high trap site density, whereas in the samples of other groups, screening played a major role because the pump generated photocarriers were accelerated along the sample surface by the biased field and it was hard for them to meet the EL2 centers, so they cannot be trapped efficiently.

The following temperature-dependent observations support our interpretation that ultrafast trapping accounts for the increased tetrahertz radiation. As the temperature is decreased, the tetrahertz wave forms slightly sharpen and increase in amplitude.[4] However, at the same pump-generation delay times, such as at the delay time of 120 ps, the variation between the wave forms that radiated from the sample with and without pump excitation at 77 K is lesser than the variation measured at 300 K. This behavior may be accounted for by a 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. The relatively low density of trap site at 77 K cannot trap carriers as efficiently as that at 300 K, and then will lead to a smaller increase of the tetrahertz emission.

In conclusion, we report the anomalous enhancement of tetrahertz emission from SI GaAs surfaces after excitation with a pump pulse. Based on our experimental and numerical results, we attribute the strong enhancement both at 300 and 77 K to ultrafast trapping of carriers. A tetrahertz probe is sensitive to the carrier density and mobility. Therefore, this tetrahertz pump-generation technique may be a useful alternative approach for elucidating the trapping process in GaAs and other materials where trapping plays a role.

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