Ultrafast carrier dynamics and terahertz conductivity of photoexcited GaAs under electric field

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The ultrafast carrier dynamics and terahertz conductivity in semi-insulating GaAs have been investigated under electric field (E) by using optical pump-terahertz probe technique. The measurements indicate that the terahertz transmission change induced by the pump pulses at high E is smaller than that without E. We attribute this phenomenon to carrier scattering into the L valley, which leads to a drop in carrier mobility. The calculated transient photoconductivities fit well with the Drude–Smith model, being consistent with our intervalley scattering model. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2980026]

Ultrafast carrier dynamics in semiconductors has attracted much attention due to the application in high speed devices.[1] Many optical techniques have been used previously to investigate carrier transport in semiconductors. Compared to the conventional experimental techniques, such as the time-resolved optical transmission technique[2] and the all-optical pump-probe spectroscopy,[3] the optical pump-terahertz probe (OPTP) spectroscopy has a plethora of advantages to provide the ability to temporally resolve phenomena at the fundamental time scales of carrier motion.[4] OPTP is such a technique that the optical beam induces a change in the sample and the resulting change in the terahertz transmission is measured. The distinct advantage of OPTP is being able to directly measure the photoinduced changes in the photoconductivity, which contains the information of carrier density and mobility, with a temporal resolution of subpicosecond.[5] Recently, this time-resolved terahertz spectroscopy has dramatically advanced in its utility, and has been widely used in probing the dynamics of photogenerated electrons, including carrier relaxation process and carrier-phonon scattering mechanism in semiconductors,[4,6,7] nanomaterials,[8] and other materials.[9,10] However, very few results about photogenerated carrier dynamics and transient photoconductivity under high electric field are found in literature.

In this letter, the behavior of terahertz transmission has been studied under different electric fields with an unchanged pump power irradiating on the GaAs surface. We have calculated the photoconductivities of the excitation layer in GaAs at the applied fields of 0 and 15 kV/cm, respectively. The frequency-dependent photoconductivity data are well fit with the Drude–Smith model.

The experimental setup of OPTP system is presented schematically in Fig. 1. A Spectra Physics regenerative amplifier system produces 800 nm pulses of 100 fs duration with 1 kHz repetition rate. The source beam is split into three portions, corresponding to terahertz generation, probe, and pump beams, respectively. The terahertz wave generation is assigned to the four-wave mixing in the air plasma produced by a type-I beta-BaB_2O_4 crystal.[11] The terahertz radiation is detected by free-space electro-optic sampling in a ⟨110⟩ ZnTe crystal.[12] Then, the signal is collected by a lock-in amplifier with phase locked to an optical chopper. The path with terahertz radiation is enclosed and purged with dry nitrogen. In our experiment, the sample is a semi-insulating GaAs wafer in the ⟨100⟩ orientation with thickness of 0.5 mm. The pump-beam energy is 11.5 mW and the spot size is at least two times larger than that of the terahertz beam to ensure uniform pump-beam illumination. The electric field is applied to the GaAs surface in the same plane. The polarization of the terahertz probe beam is parallel to the applied electric field. All experiments are performed at room temperature. The data can be acquired by one-dimensional and two-dimensional pump-probe scans.[4] The former is to scan the pump delay line while the terahertz generation delay line is fixed at the peak of the terahertz pulse. The latter refers to obtaining the terahertz waveforms by scanning the generation delay line at a series of pump delay times.

Figure 2 is a one-dimensional pump scan at the maximum value of the terahertz pulse under electric fields of 0, 6, and 15 kV/cm, respectively. For each curve, it can be seen that if the terahertz signal is ahead of the pump pulse, i.e., the delay time is negative, the terahertz peak value is unchanged. When the terahertz pulse begins to encounter the pump pulse, the terahertz transmission decreases dramatically due to the photogenerated carriers in GaAs. As the delay time Δt between pump and terahertz pulses exceeds about 3 ps, terahertz peak value commences to recover gradually as a result of the carrier recombination. However, it is surprising to see that the external E applied to the sample modulates the transmission of terahertz signal evi-

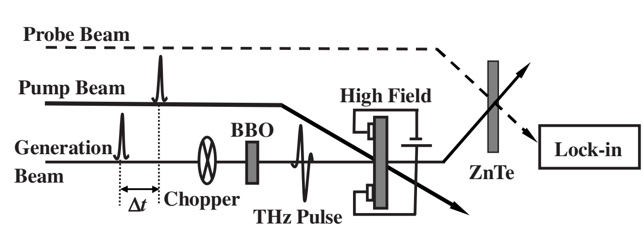

FIG. 1. Experimental apparatus used to collect OPTP spectra.

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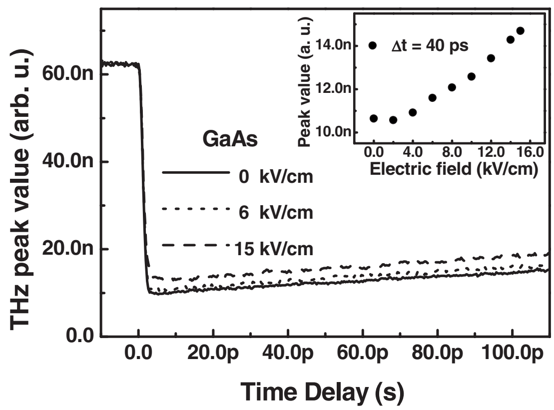

FIG. 2. One-dimensional pump scans at E of 0, 6, and 15 kV/cm, respectively. Inset: E dependence of the terahertz peak value at delay time of 40 ps.

dently at positive delay time, showing a significant increase in transmission with E. Since the carrier density is very low before the pump pulse arrives, the terahertz peak value does not change with E at negative delay time. The inset of Fig. 2 exhibits the E-dependent change in the terahertz peak value at the delay time of 40 ps. It is obvious that the threshold value of E, which begins to enhance the transmission, is about 3–4 kV/cm.

The E-dependent phenomenon can be interpreted by the schematic GaAs band structure depicted in Fig. 3. The band gap of GaAs is about 1.42 eV between the top of valence band and the bottom of Γ valley of the conduction band. The energy difference is about 0.29 eV between the Γ and L minima.[13] The 800 nm (1.55 eV) pump pulse excites the electron directly into the Γ valley. As the effective mass of hole is much larger than that of the electron, here we only consider the electron motion. The electrons injected into the Γ valley accelerate efficiently under E due to their small effective mass of $m_\Gamma = 0.067 m_0$ in GaAs. When the bias field is increased to the Gunn threshold, the electrons may reach an energy comparable to the energy level of the side valley L, and enter into the L valley. At very high E, a certain fraction of electrons may reach the X valley of the conduction band, whose bottom has about 0.48 eV energy higher than that of the Γ valley.[13] The mobilities in those satellite

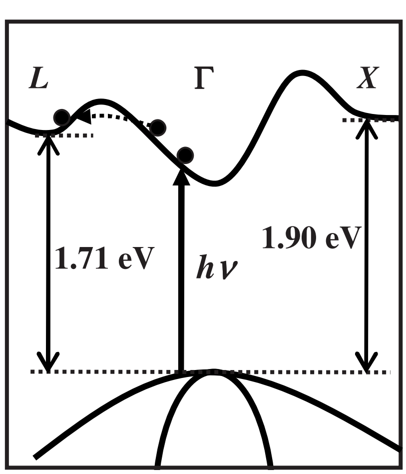

FIG. 3. The schematic GaAs band structure. Optical photon excites electrons directly into the Γ valley. A fraction of electrons subsequently transfer into the L valley at high E.

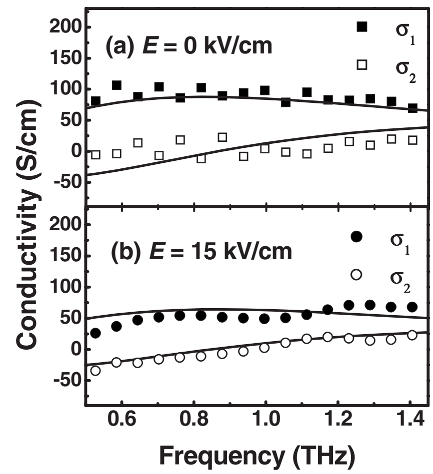

FIG. 4. The complex photoconductivity under E of 0 kV/cm (a) and 15 kV/cm (b), respectively. The solid lines are the fitting curves using the Drude–Smith model.

valleys are very low due to the large effective masses. It is known that if the carrier density remains unchanged, the absorption of the terahertz radiation depends upon the carrier mobility, exhibiting a drop in absorption with the decreased mobility.[4] Hence, the transmission will increase at high E. The threshold E of 3–4 kV/cm observed in the inset of Fig. 2 is consistent with the Gunn threshold (reported about 3 kV/cm in GaAs).[4] In addition, the Franz–Keldysh effect,[14] which increases the absorption coefficient and affects the refractive index at high E, also might influence the terahertz signal. In this way, the transmitted terahertz signal through the GaAs sample should decrease with the increasing absorption coefficient of pump excitation. However, the terahertz peak value is enhanced at high E in our experiment so the pump effect is dominated by the intervalley scattering process.

To further investigate the transport properties in the photoexcited GaAs with E, we can obtain the transient frequency-dependent complex photoconductivity by measuring the transmitted terahertz waveform with ($\tilde{E}_{\text{pump}}$) and without ($\tilde{E}_{\text{without pump}}$) the optical excitation at a specific pump time dalay.[15] The ratio of Fourier transform of these two waveforms are related to the complex conductivity $\tilde{\sigma}(\omega) = \sigma_1(\omega) + i\sigma_2(\omega)$ through[16,17]

$$\frac{\tilde{E}_{\text{pump}}(\omega)}{\tilde{E}_{\text{without pump}}(\omega)} = \frac{N+1}{N+1+Z_0 d\tilde{\sigma}(\omega)}, \tag{1}$$

where d is the thickness of the photoexcited layer (estimated to be 1 μm based on the penetration depth of the 800 nm pump in GaAs), $Z_0 = 377\ \Omega$ is the impedance of free space.[4] $N \approx 3.48$ is the index of refraction in the terahertz range of the unexcited GaAs wafer calculated from our experimental data (not shown).

Figure 4 shows the calculated photoconductivity with the real part σ_1 and the imaginary part σ_2 at the time delay of $\Delta t = 1.33$ ps under $E = 0$ (a) and $E = 15$ kV/cm (b) in 0.5–1.4 THz region, respectively. It is clear that the photoconductivity with $E = 15$ kV/cm is lower than that without E. Generally, the real part of photoconductivity corresponds to the absorption coefficient so the lower photoconductivity with high E indicates that the terahertz signal has a higher transmission. Furthermore, we find that the calculated photoconductivity is deviated from the Drude conductivity, which shows maximum value at zero frequency in the real part and positive value in imaginary part.[18] However, in our case, the real conductivity has a maximum at nonzero frequency and the imaginary part has some negative values. Therefore we use the Drude–Smith model,[19] which attributes the negative imaginary conductivity to the backward scattering of electrons as a result of the electron reflecting from surface or grain boundaries or defects or Coulombic force, to fit the photoconductivity. This model is derived based on the impulse response approach and Poisson statistics, and could mimic the infrared properties of poor metals that display a minimum in the optical conductivity at zero frequency. Many studies indicated that the Drude–Smith model could provide a superior fit to both real and imaginary parts of conductivity for many materials.[16,18,20] The Drude–Smith model is given by[19]

$$\tilde{\sigma}(\omega) = \frac{ne^2\tau/m^*}{(1-i\omega\tau)}\left[1 + \sum_{j=1}^{\infty}\frac{c_j}{(1-i\omega\tau)^j}\right]. \qquad (2)$$

The summation in Eq. (2) is often truncated after the first term, where the parameter c_1 is a measure of persistence of velocity and its negative value implies a predominance of backscattering. The parameter c_1 can vary between 0 and -1, corresponding to the Drude conductivity for $c_1=0$, and complete carrier backscattering or localization for $c_1=-1$. Additionally, n is the electron density, e is the elementary charge, m^* is the electron effective mass, and τ is the characteristic scattering time. The effective mass of electrons in the L valley is $0.55m_0$, which is much larger than that in the Γ valley. Hence, the photoconductivity is mainly determined by the electrons in the Γ valley. In our fitting, we assume that the τ of the Γ valley electrons is unchanged by the electric field, and that the carrier density n changes. The solid lines in Fig. 4 are the fitting curves with parameters of $c_1=-0.98$, $n=2.2\times10^{17}$ cm^{-3}, $\tau=190$ fs with $E=0$ and $c_1=-0.96$, $n=1.7\times10^{17}$ cm^{-3}, $\tau=190$ fs with $E=15$ kV/cm, respectively. The negative value of c_1 in our fitting implies that a fraction, but not all, of the backward scattering is a result of the electron reflecting from surfaces. It could also result from a Coulombic scattering between carriers. Due to the low mobilities of electrons in the L valley, the average mobility of all electrons will decrease under high E.

In conclusion, the transport properties of the photoexcited GaAs have been studied under electric field. It can be found that the reduced terahertz transmission due to the occurrence of the photogenerated carriers can be enhanced under the high fields. This phenomenon attributes to some electrons scattering into the L valley, which induces a drop in their mobilities. The transient complex photoconductivities with and without E have been calculated, fitting well with the Drude–Smith model. Our investigation suggests that the OPTP technique is a very promising method for detecting the ultrafast dynamics in those materials.

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