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Transient saturation absorption spectroscopy excited near the band gap at high excitation carrier density in GaAs*

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Transient saturation absorption spectroscopy in GaAs thin films was investigated using femtosecond pump and supercontinuum probe technique at excitation densities higher than $1\times10^{19}\,\mathrm{cm}^{-3}$. The Coulomb enhancement factor of the electron–hole plasma results in a spectrum hole at the pump wavelength. Two distinct transmission peaks at two sides of the pump wavelength are observed, arising from the bleaching of transitions from the heavy- and light-hole bands to the conduction band. The dynamic process of the transient saturation absorption is fitted using a bi-exponential function. The fast decay process is dominated by the carrier-phonon scattering and the slow process may be attributed to the electron–hole recombination.

Keywords: saturation absorption, supercontinuum, Coulomb enhancement factor
PACC: 7847, 7210, 7280

1. Introduction

A detailed understanding of the basic scattering mechanism of highly energetic carriers in semiconductors is essential for various problems in modern physics and technology.[1,2] Ultrafast optical spectroscopy is a major tool for the observation of hot carrier dynamics, since the strong coupling of elementary excitations, a characteristic property of solids, leads to a rapid relaxation typically occurring on a subpicosecond time scale. In intrinsic semiconductors, there are two interaction mechanisms responsible for this ultrafast temporal evolution: carrier–carrier (cc) and carrier–phonon (cp) scattering. Numbers of experiments on hot carrier distributions in GaAs have been performed at excitation densities typically between 5×10^{16} and $1\times10^{19}\,\mathrm{cm}^{-3}$.[3–9,13] In 1996, Leitenstorfer *et al*[10] first observed directly the longitudinal optical (LO) phonon emission of highly excited nonequilibrium electrons at excitation densities of 10^{15}–$5\times10^{16}\,\mathrm{cm}^{-3}$ using a novel two-colour 15 fs Ti:sapphire laser system. Their measurements indicated the time constant of LO phonon emission as being about 220 fs. Recently, Mazur and co-workers[11] have carried out a detailed measurement on the dielectric function and the second-order susceptibility at excitation densities of 3.2×10^{21}–$1.6\times10^{22}\,\mathrm{cm}^{-3}$. Their results indicated a semiconductor-to-metal phase transition which is attributed to a nonthermal structural change arising directly from the excitation. In this paper, we study the transient differential transmission spectroscopy in an intrinsic GaAs thin film at excitation densities from 3.15×10^{19} to $1.26\times10^{20}\,\mathrm{cm}^{-3}$. A two-peak configuration of differential transmission spectroscopy generated by transitions from the heavy-hole and light-hole bands to the conduction band was observed.

2. Experiment

The experimental setup[12,17] for the femtosecond pump and supercontinuum probe technique shown in

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Fig.1 is based on a Ti:sapphire chirped-pulse amplification (CPA) laser system. It (TSA-10, Spectra Physics, USA) delivers laser pulses with wavelength of 807nm and pulse duration of 130 fs. The maximum pulse energy is up to 700μJ. In the experiment the laser pulse is split into two parts by a beam splitter (BS). The reflected beam with 85% energy, after passing a variable optical delay line (ODL) controlled by a computer, is used for optical pumping. The pump beam (beam spot of 3 mm diameter) intensity can be varied from 100 to 500μJ by rotating a half-wavelength plate in front of a polarizer corresponding to carrier excitation densities from 3.15×10^{19} to 1.26×10^{20} cm^{-3}. The transmitted beam is focused into distilled water to generate a supercontinuum probe pulse. This supercontinuum pulse, with an energy of 5μJ, is converged by a lens group and then passes through a white glass plate. The transmission part, probe beam 1, is focused to intersect with the pump beam in the GaAs thin film sample of a diameter of 0.2mm. This probe beam is then detected by a fibre spectrometer (PC2000, Ocean Optics, Inc.) and the transient transmission change of $\Delta T/T$ can be obtained. The reflected probe beam from the plate, probe beam 2, is used as a reference to monitor the energy fluctuation. The time resolution of the system is about 200 fs.[14,15] The sample, a 1μm thick layer of un-doped GaAs,[18,19] was grown by molecular beam epitaxy at a temperature 605°C.

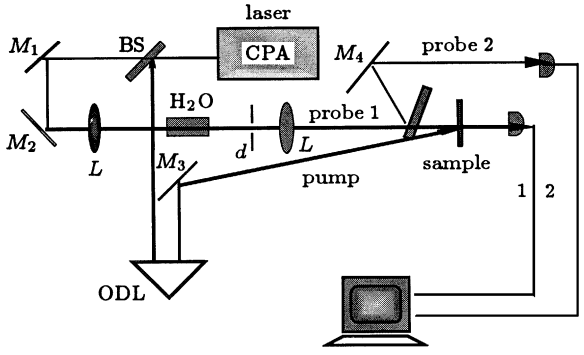


Fig.1. Femtosecond pump and supercontinuum probing (FSCP) system.

3. Results and analysis

The transient differential transmission spectroscopy is shown in Fig.2 at an excitation carrier density 1.26×10^{20} cm^{-3}. We have observed the differential transmission spectroscopy differing from that at general excitation carrier density.[3−9,20] Two distinct transmission enhancement peaks at the two sides of the pump wavelength are observed. In addition, a shoulder appears at the high-energy side of the 760 nm peak (indicated by an arrow). This corresponds to the shoulder in the linear absorption (inset) at the onset of transition from the split-off (SO) valance band (marked by an arrow) to the conduction band.[5]

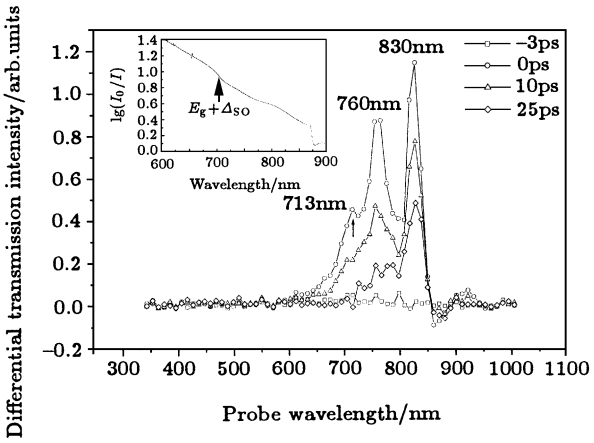


Fig.2. Differential transmission spectrum of GaAs thin film at an excitation carrier density 1.26×10^{20} cm^{-3}. The inset shows the linear absorption. A shoulder is seen in the linear absorption at the onset of transition from the split-off valance band (indicated by an arrow) and an exciton absorption peak is also seen at the absorption edge.

It is well known that the differential transmission, $\Delta T/T$, is directly proportional to $-\Delta\alpha/\alpha_0$ of the change of the sample interband absorption.[3,4,7] The absorption coefficient is expressed as follows:

$$\alpha = \frac{\Sigma}{\nu}\alpha_0 C_\nu(\hbar\omega,\rho)\sqrt{\hbar\omega - E_g(\rho)} \times [1 - f_e(k) - f_h(k)], \quad (1)$$

where α_0 is the interband absorption of the sample without excitation, $\hbar\omega$ is the photon energy, ρ is the plasma density, $C_\nu(\hbar\omega,\rho)$ is the Coulomb or excitonic enhancement factor (CEF), $E_g(\rho)$ is the band gap at a carrier density of ρ, and $f_e(k)$ and $f_h(k)$ stand for the distribution functions of the electron and hole states, respectively. For $\sqrt{\hbar\omega - E_g(\rho)}$, this is the effect of band gap renormalization (BGR), which could be ignored at excitation carrier density below 10^{21} cm^{-3}, according to the result of Mazur and co-workers.[11]

With our incident pump photon energy of 1.55 eV, carriers may be generated via interband transitions from the heavy-hole (point C) and light-hole (point E) valance bands to the conduction band (point A and point B), as shown in Fig.3.

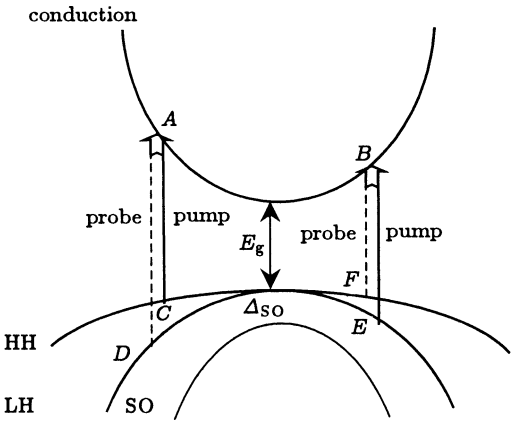


Fig.3. Schematic diagram of the band structure for GaAs (E_g=1.425eV), showing the allowed transitions (line) to the conduction band, from the heavy-hole (HH) (point C), light-hole (LH) (point E) valence bands respectively at pump pulse energy of 1.55eV. The absorption transitions (dashed line) from the light-hole (LH) (point D), heavy-hole (HH) (point F) valence bands are bleaching.

Thus, free electron distributions with high density in the conduction band are produced at 104 meV (point A) and 70 meV (point B) above the band edge, which lead to the distribution functions of the electrons $f_e(k_A)$ and $f_e(k_B)$, respectively. According to Eq.(1), the linear absorption α decreases at the probe wavelength corresponding to the transition energy at k point of k_A and k_B. Hence for the absorption transitions from point D to point A and point F to point B, the differential transmission change of $\Delta T/T$ increases owing to the increase of $f_e(k_A)$ and $f_e(k_B)$ in Eq.(1). This induces the saturation absorption of transitions from point D to point A and from point F to point B, respectively, which correspond to the probe wavelengths of 760 and 830nm in Fig.2.

A dip is observed at the pump wavelength of 807 nm in the transient transmission spectroscopy, as shown in Fig.2. We suggest that the Coulomb enhancement [10,21–23] of $C_\nu(\hbar\omega,\rho)$ at the pump wavelength induced by high-density nonequilibrium electrons is responsible for this phenomenon. Though the mechanism needs further study, one can know from Eq.(1) that when the CEF change ΔC_ν of the electron–hole plasma is large enough to compensate for the distribution function change of carriers, the differential transmission of $\Delta T/T$ at the pump wavelength depends slightly on the electron density. This is different from the cases at wavelengths of 760 and 830 nm, where there are no holes excited at point D and point F in the valance band in Fig.3. Therefore, we find that the bleaching of the probe wavelength at 807 nm is not as high as those at 760 and 830 nm.

When we change the pump intensity, the differential transmission intensity at the two peaks increase with the increase of excitation carrier density. On the other hand, the differential transmission intensity at the pump beam wavelength is nearly independent of the excitation carrier density. By calculation,[21,23] one may obtain that the CEF will reach a maximum at carrier densities 1×10^{20} and 1.7×10^{20} cm^{-3}, respectively, where the dynamic energy of excited electrons (point A and point B in the conduction band in Fig.3) is equal to the chemical potential.[21,23] From Eq.(1), it can be seen that as the CEF change counteracts the distribution function change, it occurs that the transmission signal does not depend on pump intensity at the pump beam wavelength.

Figure 4 shows the transient relaxations of saturation absorption at the probe wavelength of 830 nm with three excitation densities.

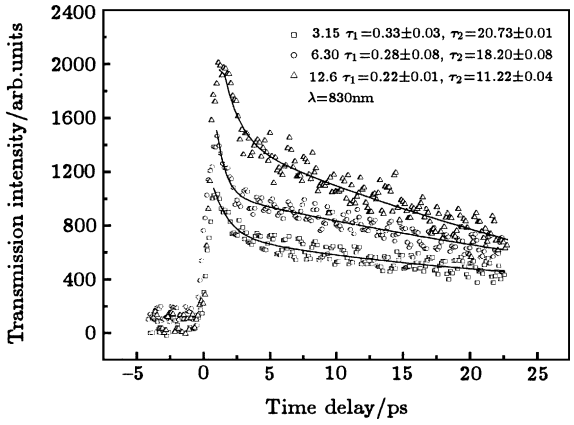


Fig.4. Relaxation characteristic of the probe wavelength 830 nm at the excitation carrier densities 3.15×10^{19}, 6.30×10^{19} and 1.26×10^{20} cm^{-3}.

The decay curve is fitted as a two-exponential relaxation process. Similarly, the decay processes of the whole bleaching spectroscopy include a fast process and a slow process reflecting the evolution of excited carriers, as shown in Table 1.

Table 1. Transient relaxation time constants of the probe wavelengths 760 and 830 nm at the excitation carrier densities 3.15×10^{19}, 6.30×10^{19} and 1.26×10^{20} cm^{-3}.

Carrier density/10^{19} cm^{-3}	τ_1/ps	τ_2/ps
λ_{pr}=760nm		
3.15	0.29±0.08	23.34±0.16
6.30	0.25±0.01	22.35±0.51
12.6	0.20±0.01	11.14±0.02
λ_{pr}=830nm		
3.15	0.33±0.03	20.73±0.01
6.30	0.28±0.08	18.20±0.08
12.6	0.22±0.01	11.22±0.04

For the fast process, as the cc scattering rate is below 200fs,[5−9,21,24] beyond our time resolution of 200 fs, the cp scattering is responsible for the fast dynamics of nonequilibrium carriers through emitting LO phonon by the conduction band electrons in our experiment. This fast decay time constant τ_1 is 200±10 fs at the probe wavelength of 760 nm, and 220±10 fs at the probe wavelength of 830 nm at excitation densities 1.26×10^{20} cm^{-3} in Table 1. From Table 1, we find that the fast decay time constant of the probe wavelength of 770 nm is smaller than that at wavelength of 830 nm. This is because the electrons at wavelength of 770 nm have higher energy than those at wavelength of 830 nm in the conduction band. The slow process with time constant τ_2 in Table 1 is usually attributed to the electron–hole recombination. τ_2 is about tens of picoseconds at low carrier densities. The recombination constant is much smaller at high carrier density than that at low carrier density. In addition, it is noted that for the slow decay process, the time constant decreases with the increase of probe wavelength. It reflects the feature of electron–hole recombination: if closer to the band edge, the recombination occurs more easily.[4,9,25] For both the fast and slow processes in Table 1, we still can see that the higher excitation carrier leads to the faster decay rate, which is in good agreement with the results in Ref.[16].

4. Conclusion

We have investigated the transient saturation absorption spectroscopy of GaAs thin film at the excitation densities from 3.15×10^{19} to 1.26×10^{20} cm^{-3}. Two-peak configuration of differential transmission spectroscopy generated by transitions from the heavy- and light-hole bands to the conduction band was studied. The Coulomb enhancement factor contribution was observed at the pump wavelength.

The relaxation process of excited carriers is fitted as a two-exponential decay. The duration of the fast process is 200–300 fs, which is attributed to the LO phonon emission, while that of the slow process is tens of picoseconds, which corresponds to the electron–hole recombination.

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References

[1] Shah J 1999 *Ultrafast Spectroscopy of Semiconductors and Semiconductor Nanostructures* (Berlin: Springer)
[2] Wang Q M 2001 *Opt. Fiber Commun.* **3** 8
[3] Tommasi R, Langot P and Vallée F 1995 *Appl. Phys. Lett.* **66** 11
[4] Alexandrou A, Berger V and Hulin D 1995 *Phys. Rev.* B **52** 4654
[5] Shi X Z, Wang L and Xia G Q 1998 *Chin. Phys. Lett.* **15** 370
[6] Lian S Y 1998 *Chin. Phys. Lett.* **15** 125
[7] Lin W Z *et al* 1999 *Chin. Phys. Lett.* **16** 758
[8] Lin W Z *et al* 2002 *Chin. Phys. Lett.* **19** 557
[9] Snoke D W, Rühle W W, Lu Y C and Bauser E 1992 *Phys. Rev. Lett.* **68** 990
[10] Leitenstorfer A *et al* 1996 *Phys. Rev. Lett.* **76** 1545
[11] Huang L, Callan P J, Glezer E N and Mazur E 1998 *Phys. Rev. Lett.* **80** 185
[12] Schoenlein R W, Lin W Z, Ippen E P and Fujimoto J G 1987 *Appl. Phys. Lett.* **51** 1442
[13] Othonos A 1998 *J. Appl. Phys.* **83** 1789
[14] Wang D L *et al* 2002 *J. Opt. A—Pure Appl. Opt.* **4** 155
[15] Wang D L *et al* 2002 *Chin. Phys. Lett.* **19** 1115
[16] Lin W Z, Fujimoto J G, Ippen E P and Logan R A 1987 *Appl. Phys. Lett.* **50** 124
[17] Lin W Z, Fujimoto J G, Ippen E P and Logan R A 1987 *Appl. Phys. Lett.* **51** 161
[18] Zhao Q X *et al* 2001 *Phys. Rev.* B **63** 125337
[19] Yuan X Z *et al* 2000 *J. Infrared Millim. Waves* **19** 385
[20] Zimmermann R 1988 *Phys. Stat. Solidi.* B **146** 371
[21] Haug H and Stephan W K 1989 *Phys. Rev.* A **39** 1887
[22] Tanguy C and Combescot M 1992 *Phys. Rev. Lett.* **68** 1935
[23] Hunsche S, Heesel H and Kurz H 1993 *Phys. Rev.* B **48** 17818
[24] Zhang H C *et al* 2000 *Acta Phys. Sin.* **49** 1171 (in Chinese)
[25] Hase M *et al* 1998 *J. Lumine.* **76** 68