Japanese Journal of Applied Physics
Vol. 44, No. 8, 2005, pp. 6101–6104

Chirp Control of Free Carrier Injection in GaAs Using Femtosecond Optical Pulses

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(Received January 11, 2005; accepted May 11, 2005; published August 5, 2005)

Control of photoinjection of free carriers in bulk GaAs at room temperature was achieved by changing the chirp of the excitation light pulses having a duration in the 10 fs regime. It was observed from pump-probe measurements that the transmittance increases for negatively chirped pump pulses, which is opposite to the trend observed with other materials. The result is explained by a combination of an intrapulse pump-dump process and band-gap renormalization, and shows the possibility of a new way to control the ultrafast dynamics of many-body systems in semiconductors.
[DOI: 10.1143/JJAP.44.6101]

KEYWORDS: chirp, GaAs, femtosecond, Urbach tail

1. Introduction

Control of dynamical properties of semiconductors using optical pulses is of great interest since it can open a new way to create electron states unachievable by other methods and to obtain new nonlinear optical properties of materials. Chirp is the most important factor that describes the coherent properties of ultrashort optical pulses. A pulse is referred to as positively chirped when the instantaneous frequency within the pulse increases temporally, and negatively chirped *vice versa*. Quantum control of the ultrafast nonlinear optical response of organic molecules has been achieved using chirped excitation pulses.[1–4] In these studies, the excited-state population of the molecules was measured by a pump-probe method while changing the chirp condition of the pump pulses, and it was observed that the excited-state population created by the pump pulses decreases when the magnitude of the negative chirp of the pump pulse is increased. This observation has been explained on the basis of the chirp dependence of the efficiency of an intrapulse pump-dump process. Molecules in the excited states having a large amount of extra energy relax to the bottom of the excited state on the femtosecond time scale. When the molecules are pumped by negatively chirped pulses, the excited-state population is expected to be dumped to the ground state by the low-frequency components of the pump pulses more effectively than when pumped by positively chirped pulses.

Kunde *et al.* observed the chirp-controlled nonlinear optical response of AlGaAs.[5] They measured pump-probe responses of a thin $Al_{0.06}Ga_{0.94}As$ sample at a carrier density of $3 \times 10^{17}\,cm^{-3}$, and observed a chirp-dependent response in the delay time region in which the pump and the probe pulses are temporally overlapped. The observed results were explained on the basis of the chirp-dependent overlap of different frequency components of the pump and probe pulses. They did not observed, on the other hand, any dependence of the response at large delay times, where there is no overlapping between the pump and probe pulses. In other words, they did not observe any chirp dependence of the total number of photoinjected carriers.

In this paper, we report our results on the chirp dependence of femtosecond pump-probe signals from bulk GaAs, in which chirp-dependent photoinjected carrier density was observed.

2. Experiments

We measured the chirp dependence of the pump-probe signal intensity of a bulk GaAs sample having a thickness of about 10 μm using 13 fs, 790 nm optical pulses. Most of the pump pulse energy is absorbed by a sample of this thickness, and only the low-energy spectral components of the probe pulse having a photon energy below the band gap pass through the sample. Thus in the measurements using samples of this thickness, the carrier-distribution dependence of the absorbance of the sample in the Urbach tail spectral region is observed. From the experiments at carrier densities of $3 \times 10^{18}\,cm^{-3}$ and higher, we observed that the transmittance of the sample at large delay times increases as the pump pulses are negatively chirped, which is in contrast to the trend observed with other materials.[1–4]

The sample used was a wedge-shaped semi-insulating GaAs crystal obtained by etching a thicker GaAs wafer. We performed transmission-type pump-probe measurements of this sample. The thickness of the probed position of the sample was about 10 μm. The pump and probe pulses were obtained from a mode-locked Ti:sapphire laser (Femtolasers, Femtosource Scientific Pro), which had a central wavelength of 790 nm and a spectral width of 100 nm. The pulses had a temporal width of 13 fs when unchirped, and the chirp of the pulses was controlled using a fused silica prism pair. The chirp was characterized by the second-order phase, ϕ_2, which is defined by the expression for the frequency dependence of the phase of the optical field:

$$\phi(\omega) = \phi_0 + \phi_1(\omega - \omega_0) + \frac{1}{2}\phi_2(\omega - \omega_0)^2. \qquad (1)$$

Experimentally, ϕ_2 of the pulses was characterized by a frequency-resolved optical gating (FROG) method.[6] Both the pump and the probe pulses had the same chirp in the experiments. The power of the probe beam was set at about a tenth of that of the pump beam. All experiments were performed at room temperature.

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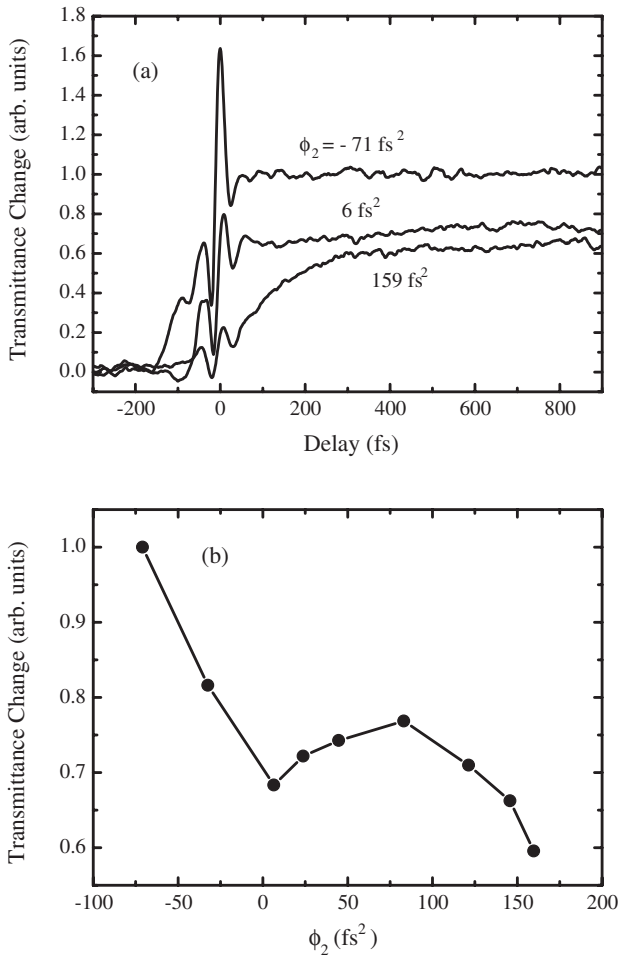

Fig. 1. (a) Delay-time dependence of transmittance change observed by pump-probe measurements of 10-μm-thick GaAs sample at excitation density of 5.9×10^{14} photons/cm^2. Results obtained using three values of the second-order phase, $\phi_2 = -71$, 6, and 159 fs^2, of the pump and probe pulses are plotted. (b) Transmittance change at large delay times as a function of second-order phase of pump pulse. The excitation density was maintained at 5.9×10^{14} photons/cm^2.

3. Results and Discussion

In Fig. 1(a) are shown the pump-probe signal intensities obtained using negatively chirped ($\phi_2 = -71$ fs^2), almost transform-limited ($\phi_2 = 6$ fs^2), and positively chirped ($\phi_2 = 159$ fs^2) pulses. Here, the excitation density was maintained at 5.9×10^{14} photons/cm^2. When the absorbance of the sample is assumed to be 10^4 cm^{-1}, this excitation density corresponds to a photoinjected carrier density of 5.9×10^{18} cm^{-3}.

In the figure, it is clearly seen that two features depend on the chirp; (i) the temporal profiles between -200 and 200 fs and (ii) the signal level at large (> 300 fs) delays. The first feature has already been observed by Kunde *et al.*,[5] and is explained by taking into account the chirp-dependent temporal overlapping between the pump and probe pulses. By excitation at 790 nm, electrons are generated with a large excess energy, and bleaching of the sample is observed only on the low-energy side of the probe spectrum. Transmittance change, therefore, is observed only when the high-energy side of the pump pulse precedes the low-energy side of the probe pulse. This leads to a faster rise of the transmittance change for negative chirp, and a slower rise for positive chirp.

The second feature described above, on the other hand, was not observed by Kunde *et al.*[5] Their results showed that the signal level at large delays was independent of the chirp. The dynamics of free carriers after photoinjection can be divided into two regimes. Photoinjected carriers thermalize within the conduction band in less than 200–300 fs, and recombine with holes on the 100 ps time scale. Thus, the pump-probe signal intensity observed at long (> 300 fs) delay times should correspond to the amount of carriers photoinjected by the pump pulse. The present experimental results, therefore, show that the control of photoinjection of carriers in bulk GaAs has been performed using chirp control of the pump pulse.

We plot in Fig. 1(b) the ϕ_2 dependence of the pump-probe signal intensity at large delays obtained at the same excitation density. The observed dependence shows a large bleaching in the negative chirp region. The signal decrease observed at around $\phi_2 = 0$ will be referred to later. Several groups[1–4] have observed the chirp dependence of the pump-probe signal of organic molecules, where a smaller bleaching in the negative chirp region was obtained. Those results were attributed to an intrapulse pump-dump process, which exists only when the pump pulse is negatively chirped. In systems such as large organic molecules, intramolecular relaxation within the excited state occurs on the femtosecond time scale after optical excitation, which leads to a stimulated gain in the spectral region below the excitation light spectrum. Thus, an intrapulse pump-dump process only occurs when the sample is excited by negatively chirped pulses.

In semiconductors, such as GaAs, the initial dynamics after optical excitation is essentially the same as that of molecular systems. An ultrafast relaxation of photoinjected electrons has been observed,[7,8] and the relaxation results in a shift of the bleaching with time towards a lower energy.[5,7] It shows that an intrapulse pump-dump process should also occur with semiconductors pumped by negatively chirped pulses. Although we have not observed the spectrally resolved pump-probe signal using the present sample, the experimental results on the chirp dependence of the pump-probe signal around the time origin is consistent with the temporal evolution of the bleaching spectrum described above.[5]

Our results, in Fig. 1(b), indicate that the amount of photoinjected carriers in bulk GaAs depends on the chirp of the pump pulse, although the chirp dependence of the transmittance change is in the opposite direction compared with that of previous reports.[1–4] This is explained by the singular excitation density dependence of the pump-probe signal intensity observed with the present sample as described below.

Since the present sample is much thicker than the absorption depth of the excitation light, almost all the energy of the pump pulse is absorbed by the sample medium irrespective of the occurrence of pump-dump processes. Only the spatial distribution of the carrier density depends on the chirp of the pump pulse. A considerable portion of the spectrum of the probe pulse, on the other hand, is also absorbed within the sample, and only the spectral portion

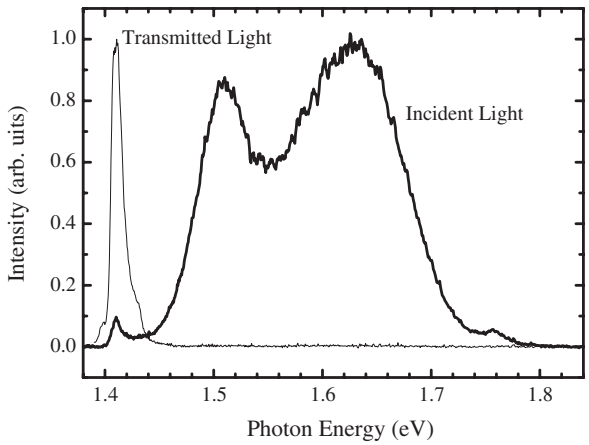

Fig. 2. Spectra of light used in pump-probe experiments. The thick line shows the spectrum of the pump and probe pulses before incidence on the sample. The thin line shows the spectrum of the probe pulse transmitted through the sample.

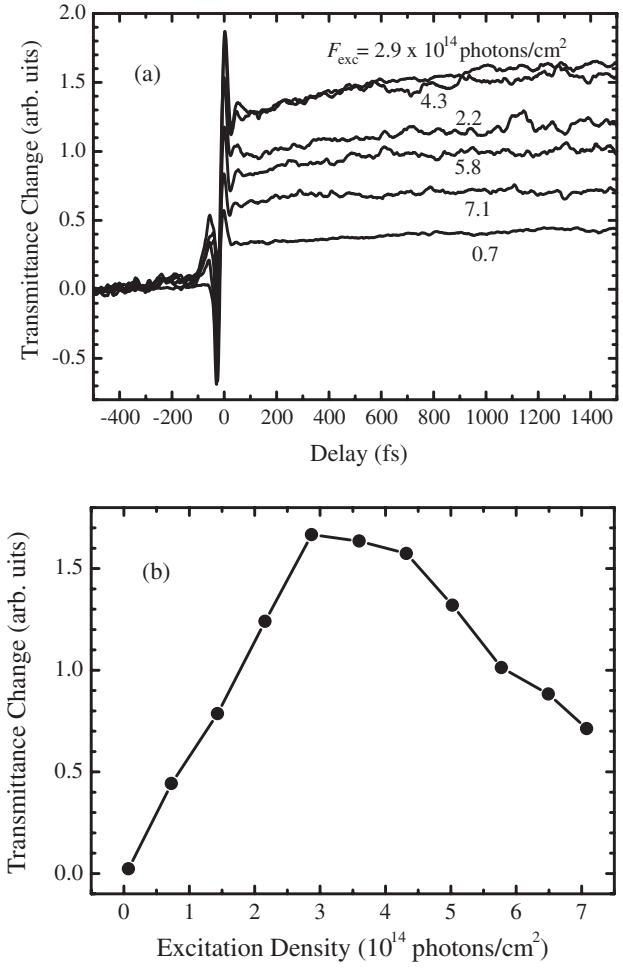

Fig. 3. Excitation density dependence of pump-probe signal intensity obtained using unchirped pump pulses. (a) Delay-time dependence of transmittance change obtained at several values of excitation densities, F_{exc}. (b) Transmittance change at large (> 1000 fs) delays are plotted as a function of excitation density.

below the band gap energy (1.428 eV), i.e., that in the Urbach tail region,[9–14] can be transmitted through the sample. The spectra of the incident and transmitted light are plotted in Fig. 2. Thus we observe the transmittance change of the sample in the Urbach tail spectral region in the experiments.

The excitation density dependence of the pump-probe signal intensity observed from the same sample using unchirped pump and probe pulses is shown in Fig. 3. The signal intensities at long delay times increase linearly with the excitation density for smaller values ($< 3 \times 10^{14}$ photons/cm^2) of excitation density and decrease for larger values. The decrease observed at large excitation densities can be attributed to band-gap renormalization,[15,16] although details of the effect of band-gap renormalization on the absorption in the Urbach tail region is to be studied further. Band-gap renormalization is a many-body effect where the band gap is reduced by the presence of free electrons. The experimental results in Fig. 3 show that the absorption of the sample in the Urbach tail region bleaches linearly with the photoinjected carrier density at excitation densities less than $<3 \times 10^{14}$ photons/cm^2, and that at excitation densities greater than this value the transmittance decreases. Since under the present experimental conditions, almost all the pump pulse energy is absorbed by the sample, the photoinjected carrier density per unit area does not depend on the chirp of the pump pulse even if a pump-dump process occurs. Thus, the pump-probe signal intensity at large delay times does not depend on the chirp at low excitation densities. At larger excitation densities, on the other hand, the absorption spectrum is affected by change in the distribution of the local carrier density per unit volume, which leads to change in the transmittance of the probe light in the Urbach tail spectral region. From a series of experiments using various excitation densities and chirps, it was found that the chirp dependent pump-probe signal intensity, as shown in Fig. 1, was observed at excitation densities above $<3 \times 10^{14}$ photons/cm^2, which corresponds to the region where the excitation density dependence of the signal intensity is inverted, as shown in Fig. 3. This shows that the singular excitation density dependence of the pump-probe signal intensity is required for the observation of the chirp-dependent signal.

On the basis of the finding on the excitation density dependence of transmittance change described above, the observed chirp dependence of the pump-probe signal intensity is explained as follows. When the sample is excited by positively chirped pulses, no pump-dump processes occur, and the spatial distribution of photoinjected carrier density is almost exponential as a function of the depth. When excited by negatively chirped pulses, an intrapulse pump-dump process occurs, and the absorption around the sample surface is reduced. The total carrier number, however, does not change even in this case, which leads to a more uniform spatial distribution of carriers. When the excitation density is below $<3 \times 10^{14}$ photons/cm^2, no chirp dependence is observed since the observed transmittance change is proportional to the spatially integrated carrier density per area. When the excitation density exceeds that value, change in the carrier density distribution affects the transmittance through band-gap renormalization, and chirp dependence is observed.

In Fig. 1(b), we observe a decrease in the signal intensity around $\phi_2 = 0$. A similar chirp dependence was also

observed by other groups.[1–3] Since the pump pulse duration is shortest at $\phi_2 = 0$, this suggests the existence of a higher-order process that depends only on the instantaneous light intensity. Ultrafast photoinduced absorption in GaAs in a spectral region higher than that of the excitation pulse has been observed,[5,7] and attributed to many-body effects.[17] Since this process occurs only during the pump pulse, it may enhance overall absorption of the pump light. Another possibility is that there is a contribution from instantaneous two-photon absorption.

4. Conclusion

We have achieved control of carrier photoinjection in a thick bulk GaAs sample using femtosecond chirp-controlled excitation pulses. The observed chirp dependence of the pump-probe signal intensities was in the direction opposite to previous observations obtained using organic molecules. On the basis of the singular excitation density dependence of the pump-probe transmittance change observed with the thick sample, we conclude that an intrapulse pump-dump process occurs when pumped by negatively chirped pulses even in many-body electron systems in semiconductors.

Acknowledgements

This work has been supported in part by the 21st Century Center of Excellence (COE) Program "Promotion of Creative Interdisciplinary Materials Science for Novel Functions" under the Ministry of Education, Culture, Sports, Science and Technology.

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