Chirp Control of Free Carrier Dynamics in GaAs

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Abstract. The dynamics of free carriers in bulk GaAs was controlled by changing the chirp of the excitation light pulses having a duration in the 10 fs regime. Pump-probe measurements showed that the transmittance increases for negatively chirped pump pulses, which is in contrast to the trend observed with other materials. The result is explained by a combination of a pump-dump process and bandgap renormalization, and shows the possibility of a new way to control nonlinear optical response of semiconductors.

Coherent control of dynamical properties of semiconductor materials 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, and chirp dependences of ultrafast nonlinear optical response of organic molecules have been observed [1,2]. In these studies, the excited-state population of the molecules was measured using a pump-probe method while changing the chirp of the pump pulses, and it was observed that the excited-state population created by the pump pulses decreases for negatively chirped pump pulses. This observation has been explained based on the chirp dependence of the efficiency of an intrapulse pump-dump process. Kunde *et al.* observed chirp-controlled nonlinear optical response of AlGaAs at a carrier density of 3×10^{17} cm^{-3}, and observed chirp-dependent pump-probe response in the delay time region where the pump and the probe pulses are overlapped [3]. The observed results were explained based on the chirp-dependence 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 the probe pulses.

We measured the chirp dependence of the transmission-type pump-probe signal intensity of a bulk GaAs sample having a thickness of about 10 μm at room temperature. At carrier densities 3×10^{18} cm^{-3} and higher, we observed that the transmittance of the sample at large delay times increases for negatively chirped pump pulses, which is in the opposite direction to the trend observed with other materials [1,2]. The pump and probe pulses 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, which is characterized by the second-order phase, ϕ_2, was controlled using a fused silica prism pair. Both the pump and the probe pulses had the same chirp in the experiments. In Fig. 1 are shown the pump-probe signal intensities obtained using negatively chirped ($\phi_2 = -71$ fs^2), almost

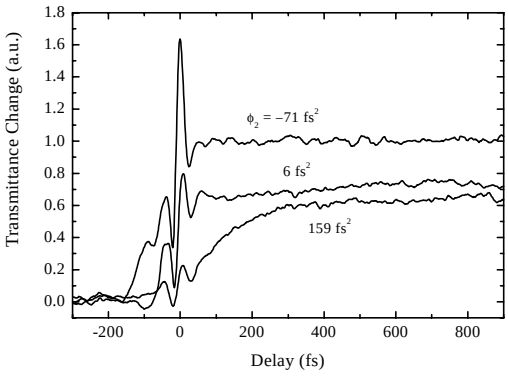

Fig. 1. Delay-time dependence of transmittance change observed by pump-probe measurements of a 10-μm-thick GaAs sample at a carrier density of 5.9 x 10^{18} cm^{-3}. Results obtained using three values of second-order phase, ϕ_2, of the pump and probe pulses are plotted.

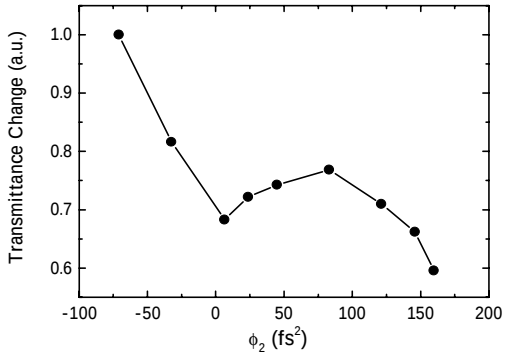

Fig. 2. Chirp dependence of the transmittance change of the pump-probe measurements at large delays, obtained at a carrier density of 5.9 x 10^{18} cm^{-3}.

transform-limited (ϕ_2 = 6 fs^2), and positively chirped (ϕ_2 = 159 fs^2) pulses. The pump pulse energy was kept constant, and the density of the created carriers was estimated as 5.9 x 10^{18} cm^{-3}.

In the figure, it is clearly seen that two features depend on the chirp; (i) the temporal profiles between −200 to 200 fs, and (ii) the signal level at large (> 300 fs) delays. The first feature was also observed by Kunde *et al.* [3], and is explained by taking into account the chirp-dependent temporal overlapping between the pump and probe pulses. The second feature, on the other hand, was not observed by Kunde *et al.* [3], where the signal level at large delays was independent of chirp. Since the carrier density is higher in the present study, we can attribute the difference to higher-order effects. We plot in Fig. 2 the chirp dependence of the

pump-probe signal intensity at large delays obtained at the same carrier density. The observed dependence shows larger bleaching at negative chirp. In experiments using organic molecules, opposite chirp dependence of pump-probe signal was observed [1,2]. Those results were attributed to an intrapulse pump-dump process which exists only when the pump pulse is negatively chirped. Since the chirp dependence of the temporal signal profile around zero delay, as seen in Fig. 1, indicates that a pump-dump process surely exists between the pump and the probe pulses, an intrapulse pump-dump process is expected to occur also in the present measurements.

We have found that the cause of the observed non-intuitive chirp dependence of the pump-probe signal is a peculiar carrier-density dependence of the pump-probe signal intensity obtained with the present experimental configuration. From the measurement of the carrier-density dependence of the pump-probe signal intensity at large delays using unchirped pump pulses, we observed that the signal intensities increase linearly to the carrier density for smaller values ($< 3 \times 10^{18}$ cm^{-3}) of carrier density, and decrease for larger values. Since the present sample had a thickness of about 10 μm, which is much larger than the penetration length of resonant light, only the spectral portion of the probe pulse below the bandgap was detected. Pump-probe measurements conducted under this condition probe the absorbance change in the Urbach tail spectral region of the sample. Most of the pump pulse energy is also absorbed by the sample of this thickness regardless of the chirp, and only the spatial distribution of carrier density depends on the chirp. When the carrier density is small, the measurement can detect the bleaching that is proportional to the carrier density. When the carrier density becomes larger, however, bandgap renormalization occurs, which will raise absorbance in the Urbach tail, leading to decrease in transmitted light intensity. The carrier density region where chirp dependence similar to that shown in Fig. 2 was observed almost agrees with that where the pump-probe signal shows negative carrier-density dependence. In this carrier density region, a pump-dump process occurs for negatively chirped pump, which leads to more uniform distribution of carriers. Decrease of signal intensity at $\phi_2 = 0$ suggests that there is a contribution from instantaneous two-photon absorption, which is dependent only on the pump pulse width.

In conclusion, we have observed chirp-controlled carrier creation in GaAs. We observed a chirp dependence of pump-probe signal intensities of a thick GaAs sample which is opposite to that observed previously with organic molecules. This result can be explained by considering a pump-dump process and bandgap renormalization in a thick sample.

References

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