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 new way to control nonlinear optical response of semiconductors.

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Coherent control of dynamical properties of semiconductor materials is of great interest since it can open a new way to creat 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 moleclues 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. Since excited-states having larger energy lose the extra energy on the femotosecond time scale, when 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 chirp-controlled nonlinear optical response of AlGaAs [3]. They measured pump-probe responses of a thin $Al_{0.06}Ga_{0.94}As$ sample at a carrier density of 3×10^{17} cm^{-3}, and observed chirp-dependent response in the delay time region where the pump and the probe pulses are overlapped. 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.

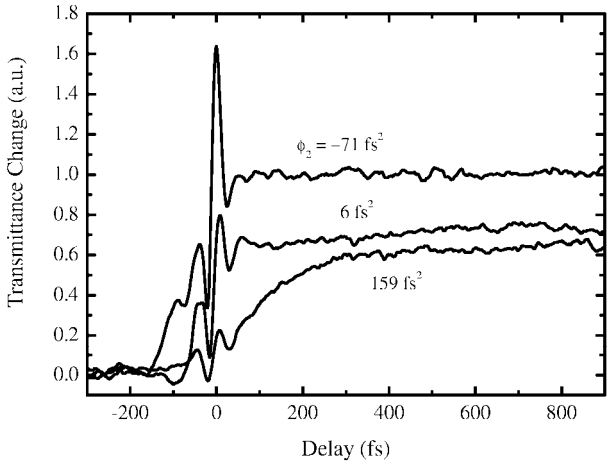

Fig. 1. Delay-time dependence of transmittance change observed by pump-probe measurements of a 10-μm-thick GaAs sample at a carrier density of 5.9 × 10^{18} cm^{-3}. Results obtained using three values of second-order phase, ϕ_2, of the pump and probe pulses are plotted.

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 pulses. 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 contrast to the trend observed with other materials [1, 2].

The sample used was a wedge-shaped semi-insulating GaAs crystal obtained by etching a 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, and 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 was measured using the frequency-resolved optical gating (FROG) method. Both the pump and the probe pulses had the same chirp in the experiments. All experiments were performed at room temperature. In Fig. 1 are shown the pump-probe signal intensities obtained using positively chirped ($\phi_2 = -71$ fs^2), almost transform-limited ($\phi_2 = 6$ fs^2), and negatively chirped ($\phi_2 = 159$ fs^2) pulses. The pump pulse energy was kept constant, and the density of the created carriers was 5.9×10^{18} cm^{-3}, which was estimated from the pump pulse energy and the excited area of the sample.

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 already observed by Kunde et al. [3], 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 large excess energy, and bleaching of the sample is observed only on the low-energy side of the probe spectrum. Thus transmittance change 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. [3], where the signal level at large delays was independent of the 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. Cerullo et al. [1] and Misawa et al. [2] have observed chirp dependence of pump-probe signal of organic molecules, where smaller bleaching at negative chirp was observed. Those results were attributed to an intrapulse pump-dump process which exists only when the pump pulse is negatively chirped. The present results, however, show the opposite dependence of the pump-probe signal on the chirp of the pump pulse. 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

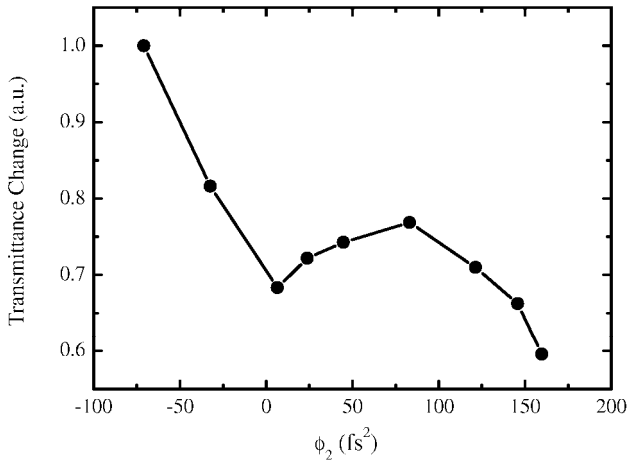

Fig. 2. Chirp dependence of the transmittance change of the pump-probe measurements at large delays, obtained at a carrier density of 5.9×10^{18} cm^{-3}.

pump-dump process is also expected to occur.

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. In Fig. 3, we plot the carrier-density dependence of the pump-probe signal intensity at large delays, which was measured using unchirped pump pulses. The signal intensities increase linearly to the carrier density for smaller values ($< 3\times10^{18}$ cm^{-3}) of carrier density, and decrease for larger values. Since the present sample had a thickness of about 10 μm, which is much longer than the penetration length of resonant light, only the spectral portion of the probe pulse below the bandgap (& 868 nm) 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 measurememt 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 pump-probe signal shows negative carrier-density dependence, as seen in Fig. 3. In this carrier density region, a pump-dump process occurs for negatively-chirped pump, which leads to more uniform distribution of carriers. When pumped by positively-chirped pump pulses, on the other hand, the effect of the pump-dump process is smaller, and the carrier density at the front surface becomes larger, resulting in increase in absorption of probe light due to bandgap renormalization. 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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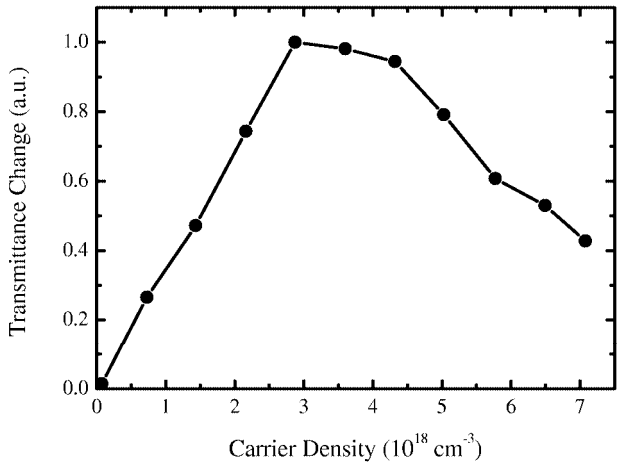

Fig. 3. Carrier density dependence of the pump-probe signal intensity at large delays. The data were obtained using unchirped pump pulses.