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# Chirped-pulse manipulated carrier dynamics in low-temperature molecular-beam-epitaxy grown GaAs

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Chirped pulse controlled carrier dynamics in low-temperature molecular-beam-epitaxy grown GaAs are investigated by degenerate pump-probe technique. Varying the chirped condition of excited pulse from negative to positive increases the carrier relaxation time so as to modify the dispersion and reshape current pulse in time domain. The spectral dependence of carrier dynamics is analytically derived and explained by Shockley-Read Hall model. This observation enables the new feasibility of controlling carrier dynamics in ultrafast optical devices via the chirped pulse excitations. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4875027]

Selective carrier population transfer using chirped pulses has been extensively studied based on their potential applications in various areas, such as optical quantum control,[1–8] spectroscopy,[9] and Bose-Einstein condensates.[10] The chirped pulse control of carriers in direct band-gap semiconductors was discussed[11] to enable the optimization ultrafast optoelectronic devices under chirped pulse illumination. In 1980s, Ippen *et al.* reported the chirp dependent relaxation dynamics pumped by either positively chirped or negatively chirped pulses.[12] Yet, no clear discussion on the mechanisms related to carrier dynamics excited and controlled by chirped optical pulses is addressed. Among the ultrafast optoelectronic materials to be studied, either the low-temperature molecular-beam-epitaxy (MBE) grown GaAs (LT-GaAs)[13] or the arsenic-ion-implanted GaAs (GaAs:As+)[14–16] is potentiate for ultrafast photonic devices because of its high mobility, short carrier lifetime, and high breakdown voltage. In the late 1990s, several groups have developed rate equation sets for LT-GaAs, which accurately predicted the carrier dynamics through a wide scanning on intensities and wavelengths of excitation pulse.[17–19]

In this Letter, we present a systematic investigation of the carrier dynamics controlled by pulse excitation with varying chirp. The change on lifetime of excited carriers under pulse excitation with different chirps, the simplified rate equation model based on the band diagram of LT-GaAs, and the phenomenal explanation on the spectrally dependent carrier dynamics are addressed. The modified rate equation considers the absorption due to population in valance band via single and two photon processes and the contribution of mid-gap states. We present the model in a semi-classical picture that is different from the adiabatic rapid passage and intra-pulse pump-dump mechanism.[2,4]

Figure 1 shows the band diagram of LT-GaAs used in this paper. In principle, when LT-GaAs is excited by femtosecond optical pulses with photon energies above the band gap, electrons are mostly excited to the upper state (excited state) of the conduction band with high thermal energy. The free carriers in the conduction band can either be trapped by the mid-gap state or relax to the bottom of the conduction band. The free carriers in the bottom of the conduction band also can be trapped by mid-gap states. The two-photon absorption process and mid-gap state absorption process are neglected here due to the selection of excitation photon energy.

The carrier dynamics for LT-GaAs can be described by the following equations:

$$\frac{dn}{dt} = \frac{I\alpha}{h\nu} - \frac{n}{\tau_1} - \frac{n}{\tau_2}, \tag{1}$$

$$\frac{dN}{dt} = \frac{n}{\tau_2} - \frac{N}{\tau_3}, \tag{2}$$

where n is the population of carriers at the upper state of the conduction band, N is the population of carriers at the
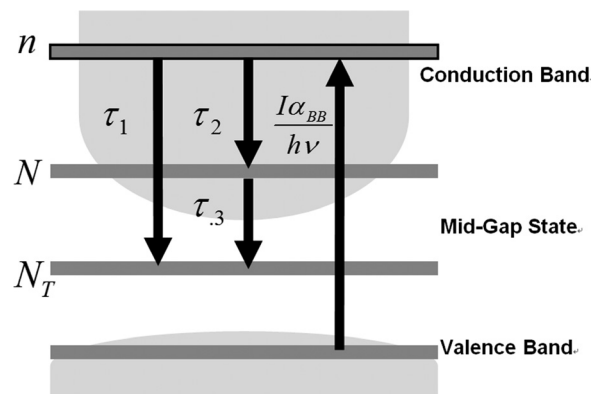


FIG. 1. The band diagram of low-temperature-grown GaAs.

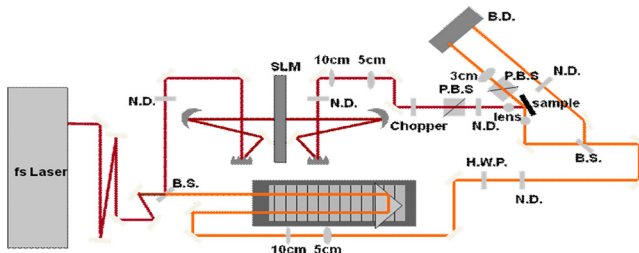a)Author to whom correspondence should be addressed. Electronic mail: chuckcklee@yahoo.com

FIG. 2. The experimental setup of excitation chirp controlled pump probe system.

bottom of the conduction band, I is the intensity of the excitation pulse, $\alpha$ is the band to band absorption coefficient, $h\nu$ is the photon energies, $\tau_1$ is the carrier trapping time from the upper state of the conduction band to the mid-gap state, $\tau_2$ is the carrier cooling time in the conduction band, and $\tau_3$ is the carrier trapping time from the bottom of the conduction band to the mid-gap state.

Since the refractive index change in the semiconductor material is proportional to the carrier density in the conduction band, the carrier dynamics in LT-GaAs can be recorded by a time resolved pump-probe reflectivity measurement. A Ti:sapphire oscillator operating at 800 nm with central photon energy of 1.55 eV. The FWHM bandwidth of the laser pulse is 10 nm and the associated transform-limited pulse duration is around 90 fs in FWHM. The pulse duration of the pump beam was tailored by a zero dispersion pulse compressor consisting of a pair of gratings 600 g/mm, two parabolic reflectors with a focal length 37.5 cm, and a liquid crystal spatial light modulator (CRI, SLM-128 phase mode). The pump beam was focused down to 100 $\mu$m or 20 $\mu$m with two different objective lenses and the incident angle was 25° off normal. The probe beam had 1 mW average power and the incident angle was 55°. The transient reflectivity, indicating

the population of free carriers in the conduction band, was recorded as a function of the time delay between two pulses and the carrier relaxation times were retrieved by fitting the signal with Eqs. (1) and (2). The experimental setup of the femtosecond pump-probe system with excitation chirp controlled pump beam is schematically shown in Fig. 2.

Results for the LT-GaAs sample at low pump fluence of $5 \times 10^{-3}$ MW/cm$^2$ are presented in Fig. 3. The two chirped excitation conditions, $\pm 8000$ fs$^2$ at rising regions exhibit equivalent pulsewidth; however, the falling region under positive chirped excitation is larger than the negative one. The carrier relaxation times $\tau_1$, $\tau_2$, and $\tau_3$, are retrieved from experimental data with a de-convolution procedure via Eqs. (1) and (2). Apparently, the trend of experimental data (see green lines in Fig. 3) reveals that all three relaxation times increase as the quantity of excitation chirp increase from negative to positive, as shown in Figs. 3(b)–3(d). We attribute this behavior to the spectrally dependent carrier dynamics in LT-GaAs, and similar discussions were mentioned by Ippen *et al.* previously.[12,22] The bandgap dependent carrier trapping time was correlated with the gross effect of possible scattering channels depending on the photon energy. Horng *et al.* also observed shorter carrier cooling time in GaAs under excitations at low central wavelength.[21–23] The consistent observations from two different groups result from the fact that the photo-excited carriers with sufficient excess kinetic energy is able to induce the optical phonon vibration. Especially, when the hot carriers are excited to upper state of conduction band by absorbing high-energy photons, jumping to above bandgap with an excess kinetic energy of smaller than 300 meV is mandatory to screen out the split-off band transition and also to reduce the probability of inter-valley scattering.

Despite the chirp dependent absorption due to intrapulse pump-dump process and bandgap renormalization,[24] we propose a model to explain the carrier relaxation time
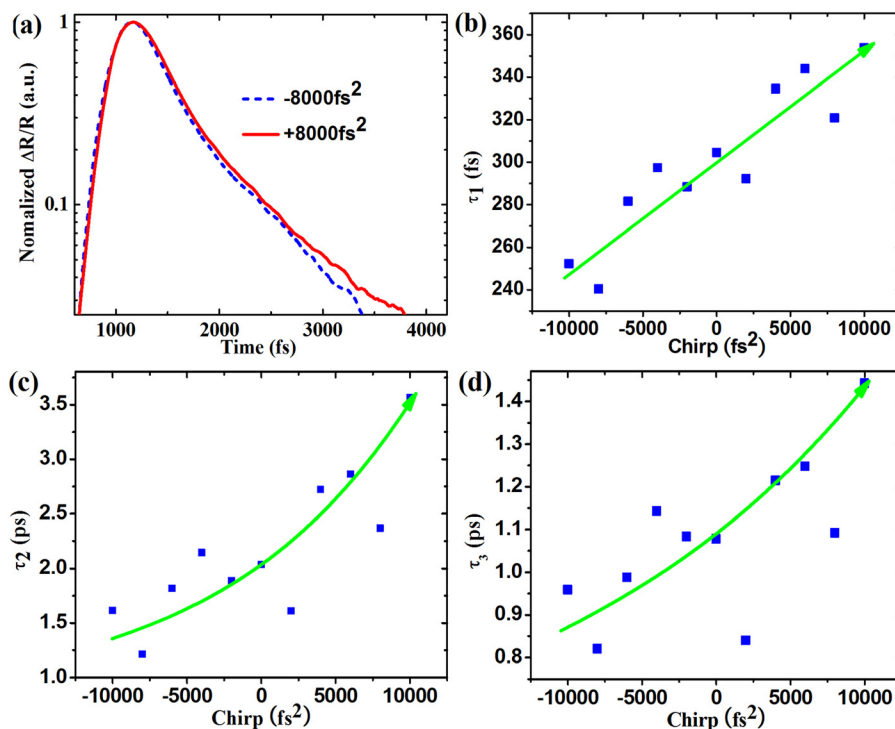


FIG. 3. (a) Normalized transient reflectivity signals with pump fluence $5 \times 10^{-3}$ MW/cm$^2$ and chirped conditions $\pm 8000$ fs$^2$. (b)–(d) Carrier lifetimes $\tau_1$, $\tau_2$, and $\tau_3$ as a function of excitation chirp with green lines indicating the increasing trend.
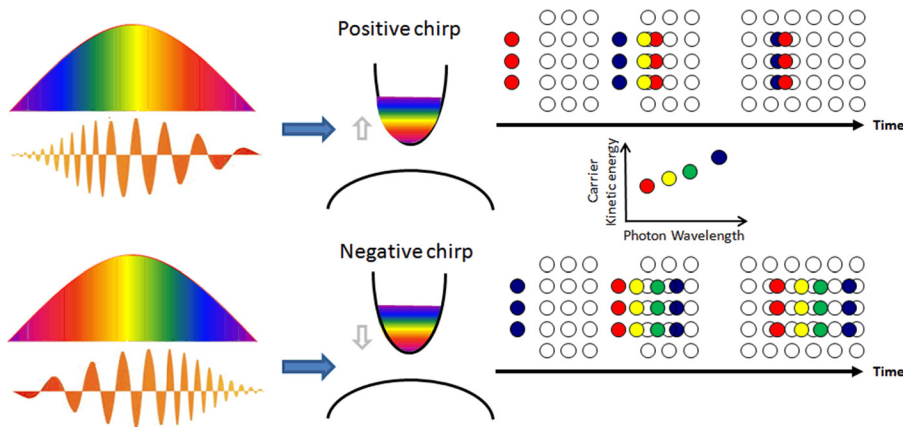
FIG. 4. The schematic illustration of the carrier dynamics in LT-GaAs under the excitation by femtosecond laser pulses with varying sign of chirp.

increment as a function of the excitation chirp (when increasing from negative to positive). Under a positively chirped excitation, the photo-excited carriers with low excess kinetic energy are generated first, which exhibit long carrier cooling time corresponding to low thermal velocities. These electrons eventually leads to a weak carrier diffusion based on the Shockley-Read-Hall (SRH) model.[25] The subsequent carriers with higher thermal velocities will diffuse faster to spatially overlap the former excited carriers. This results in a local increase of photo-carrier density in LT-GaAs and long carrier trapping time $\tau_1$, as a consequence of band filling and the reduction of the rate of trapping occupancy inferred by SRH model.[26] With a negatively chirped excitation, the photo-excited carriers lagged behind the peak excitation have lower thermal velocities than the former and free carries spreading out in space. At last stage, these interactions decrease photo-excited carrier density and cause shorter carrier trapping time $\tau_1$. Therefore, the carrier trapping times $\tau_1$ increase with enlarging excitation chirp from negative to positive. Following the same chirping dependent local density variation, the carrier trapping time $\tau_3$ for the population in the bottom of conduction band also lengthens with enlarging chirp of excitation.

To reinforce, the illustration on the carrier dynamics in the LT-GaAs under the transient excitation by femtosecond laser pulses with varying signs of chirp is shown in Fig. 4. In principle, the hot carrier cooling time can be reduced when increasing the pumping photon energy to slightly above band-gap. For a positive chirped excitation, carriers with long cooling time are generated first. At this moment, only few optical phonons are excited. Although the subsequently excited carriers exhibit short cooling time with a stronger carrier-

phonon interaction,[20] carriers are obstructed during period of relaxation. This eventually lengthens the carrier cooling time $\tau_2$. On the contrary, when excited by negatively chirped optical pulse, the leading carriers induce large optical phonon population that is able to scatter the subsequent carriers and results in short cooling time $\tau_2$. In our experiment, even the inter-valley scatterings from $\Gamma$ to L still occurs even with an excessive photon energy of only 0.12 eV, but these scatterings fail to favor lengthen hot carrier cooling time. The result clearly elucidates that the carrier dynamics can be manipulated with chirped excitation, indicating the feasibility of controlling carriers in samples by chirped pulses.

In addition, the dynamic responses under various excitation intensities are also investigated, since the chirp dependence can be attributed to spatial photo-carrier density variation. By varying pump power and focus length, the chirp dependent carrier trapping time ($\tau_1$) at various pump fluence is obtained, as shown in Fig. 5. The carrier trapping time boosts as the excitation fluence increases, and the chirped dependency become significant at high pump fluence. From the modified SRH model, the trapping ability of defects goes down such that carrier trapping time increases at high excitation density. Therefore, the carrier trapping times $\tau_1$ boost as the excitation fluence increases. This eventually results in the coupling strength between carriers enhanced at high excitation density. Therefore, the rising chirped dependency is straightforward.

In conclusion, the excitation chirp dependent carrier relaxation time variation is observed, which lengthens the carrier relaxation process by detuning the chirp of excitation from negative to positive. The chirped pulse excitation pump-probe analysis elucidates the spectral dependence of carrier dynamics in LT-GaAs, which discloses the feasibility of controlling carrier dynamics in ultrafast semiconductor with chirped pulses. The introduction of chirping excitation directly re-arranges the photon energy distribution in time domain and creates an environment to be in favor of some specific interactions. This approach should be applicable in various physical systems. For example, more efficient broadband THz generation from a photoconductive antenna with its lifetime shortened by manipulating the excitation chirp should be expectable.[27]
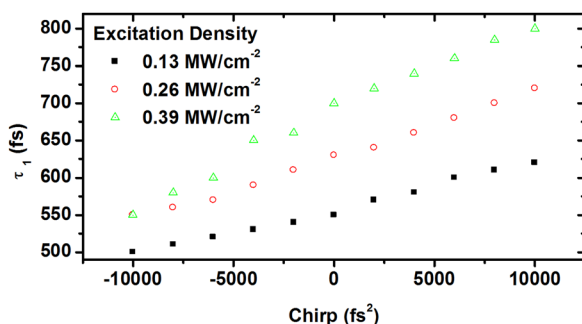
FIG. 5. Carrier lifetimes $\tau_1$ as a function of excitation chirped conditions.

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