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Ultrafast intraband electron dynamics of GaAs and InP observed by terahertz emission

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Abstract

We have studied the ultrafast dynamics of electrons generated by femtosecond optical pulses with positive and negative excess energies in GaAs and InP by observing the waveform of the emitted terahertz radiation. Subpicosecond intraband relaxation was observed with positive excess energies. With negative excess energies, a picosecond transition from the Urbach state to free carrier states was observed. This dynamical behaviour was found to be very sensitive to the applied field magnitude in the range of several kV/cm.

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Keywords: THz radiation; Gallium arsenide; Indium phosphide; Urbach tail

1. Introduction

Ultrafast dynamics of carriers in semiconductors is of great importance for understanding the basic physics and for applications of semiconductors in high-speed electronic devices, and has been studied extensively with various ultrafast nonlinear spectroscopic techniques, such as pump–probe and four-wave mixing measurements [1]. Recently, techniques for generation and detection of terahertz (THz) electromagnetic radiation using femtosecond optical pulses have been developed [2–5], and are widely used in spectroscopy and imaging. In these applications, biased or unbiased semiconductors are most often used as the emitter material. In these emitter materials, transient current generated by ultrashort optical pulses serves as the source of the THz radiation. This implies that the magnitude and the temporal waveform of the emitted THz pulses carry rich information on the ultrafast dynamics of the coherent motions of the photogenerated carriers in the emitter materials.

So far, several studies on the ultrafast electron dynamics using the THz emission technique have been reported [6–9]. In these studies, electron velocity dynamics in GaAs under relatively high bias field (typically several tens of kV/cm) were observed by applying a voltage to the sample with very narrow electrode spacing (0.5–4 μm). In the present study, we adopted a different structure, a large-aperture photoconductive antenna, as the THz emitter, and observed the ultrafast dynamics of the mobility of photogenerated electrons in GaAs and InP under moderate electric field.

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2. Experiments

A schematic for the experimental set-up is shown in Fig. 1. Briefly, a GaAs or InP large-aperture photoconductive antenna [2,3] with a 3-cm gap between the electrodes was pumped at a repetition rate of 1 kHz by 150-fs tunable optical pulses obtained from an optical parametric amplifier (OPA). The temporal waveform of the electric field of the emitted THz radiation was measured using a standard setup [10] of the electro-optic (EO) sampling method. The pump photon energy was tuned across the band-gap energy of each material; 1.428 eV for GaAs and 1.351 eV for InP. The spectral width of the pump pulse was narrowed to 10 nm using a band-pass filter. The pulse energy of the pump pulse was about 3 μJ. The photogenerated electron density is estimated to be $10^{15}-10^{17}\,\mathrm{cm}^{-3}$, depending on the absorption coefficient at the pump wavelength. The probe pulse of the EO sampling measurement was obtained by splitting off a small part of the output of the regenerative amplifier, which was the seed of the OPA. The pulse width and the wavelength of the probe pulse were 130 fs and 800 nm, respectively. A Si wafer was placed in the path of the THz radiation in order to block light and pass only THz waves. The pump pulse train was chopped at 500 Hz and the EO sampling signal was lock-in detected at this frequency.

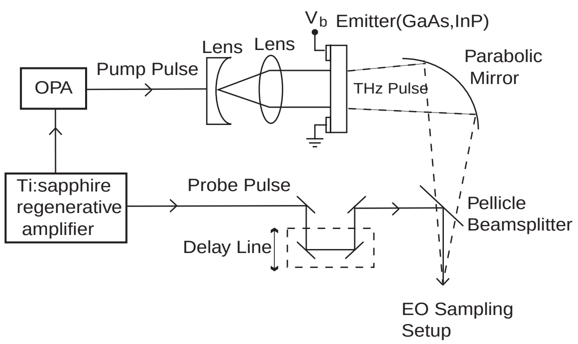

Fig. 1. Schematic of the experimental set-up for the THz waveform measurement. OPA is for an optical parametric amplifier.

3. Results and discussion

Pump photon energy dependence of the THz waveforms is shown in Fig. 2. The observed tendency is summarized in Fig. 3, where the peak electric field magnitude and the pulse width of the THz pulse are plotted as functions of the excess energy (pump photon energy minus band-gap energy). When keeping in mind that the pump pulse has a spectral width of about 15 meV, it is seen that the largest field and shortest pulse width are obtained in both GaAs and InP when pumped by optical pulses with photon energy just above the band-gap energy. The excess energy

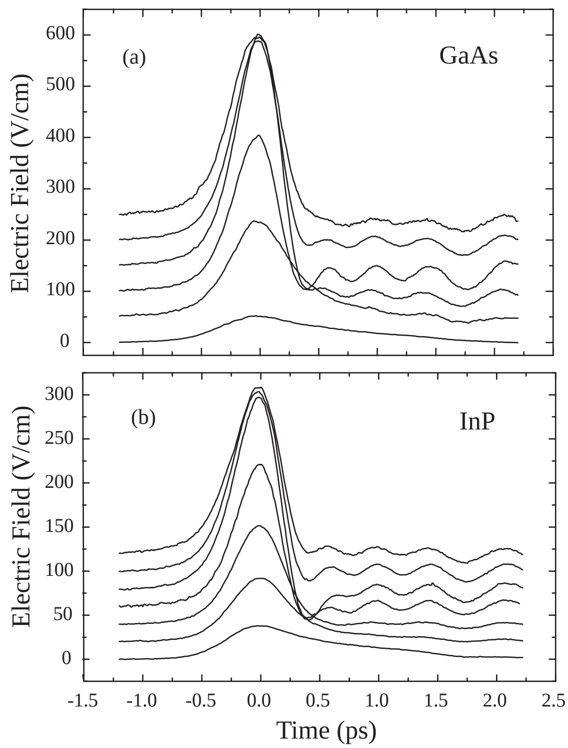

Fig. 2. Temporal waveforms of the THz pulses emitted from (a) GaAs and (b) InP for several values of the excess pump photon energy with respect to the band-gap energy. The bias electric field applied to the emitter material was kept at 6.7 kV/cm. In (a), waveforms are shown for excess energies of $122, 66, 14, -3, -19$, and -35 meV from the top to the bottom. Each plot is shifted upward by 50 kV/cm. In (b), waveforms are shown for excess energies of $198, 108, 11, -3, -18, -32$, and -46 meV from the top to the bottom. Each plot is shifted upward by 20 kV/cm.

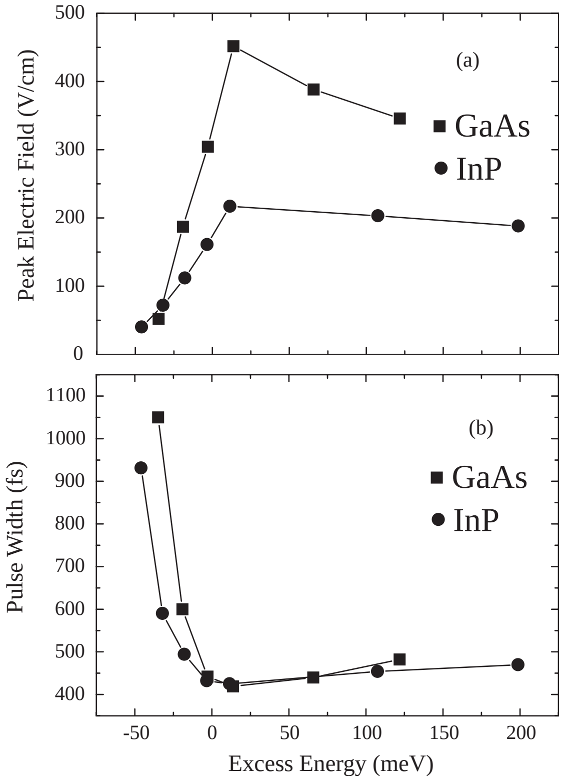

Fig. 3. The peak electric field and the temporal width of the emitted THz pulses are plotted as a function of the excess energy in (a) and (b), respectively, for GaAs (square) and InP (circle).

dependence of the peak field and the pulse width show a dramatic change when the sign of the excess energy is changed.

For positive excess energies, the peak field is slightly decreased and the pulse width is slightly increased for larger excess energies. The observed THz waveform can be regarded as the time derivative of the time dependence of the transient current induced by the photogeneration of electrons in the emitter materials [3]. Any contribution of photogenerated holes to the current can be neglected since the mobility of holes is much smaller. Electrons generated in the conduction band with large excess energy have smaller mobility than at the bottom of the band, due to a larger scattering rate, and relax to the bottom on a subpicosecond time scale [1]. Correspondingly, the electron mobility is expected to increase on the same time scale [11], resulting in a slower rise of the current, or a broader THz pulse width. In contrast, when generated by pump light with photon energy slightly larger than the band-gap energy, electrons are created directly at the bottom of the conduction band, leading to an almost pulse width limited rise time of the mobility. Thus, we obtained the shortest and highest THz pulse at this pump photon energy.

With negative excess energies, on the other hand, we observed an exponentially decaying tail in the THz waveforms with a time constant of 1–2 ps, which corresponds to a slower rise in the mobility of electrons photogenerated in this energy region. This energy region corresponds to the Urbach tail [12], where the absorption coefficient decreases exponentially with photon energy. The Urbach tail is generally attributed to disorder in the electron potential due to electron–phonon interaction and/or defects and impurities in the crystal. The present experimental results show that the electrons excited in the Urbach tail region (Urbach electrons) do not contribute to the macroscopic current just after the excitation, and that a transition to a mobile state occurs in 1–2 ps. We believe that the present results are the first experimental observation of the ultrafast dynamics of Urbach electrons.

Furthermore, we have found that this dynamical behaviour depends sensitively on the applied bias field in the moderate field magnitude region of several kV/cm. The bias field dependence of the THz waveform emitted from GaAs with excess energy of -35 meV is shown in Fig. 4. Similar results have been obtained with InP. It is seen that the decay of the THz field becomes faster for larger bias fields. The decay times obtained by exponential fitting were 1.4 and 0.7 ps for bias fields of 1.3 and 9.3 kV/cm, respectively. Dow and Redfield [13] explained the Urbach tail formation by electric-field ionization of excitons and estimated the average effective field magnitude due to electron–LO–phonon interaction at 6.9 kV/cm in GaAs, which lies in the observation range of the present study. The fact that a large field dependence of the THz decay time has been observed in this field region in the present study shows that the observed current dynamics corresponds to thermal

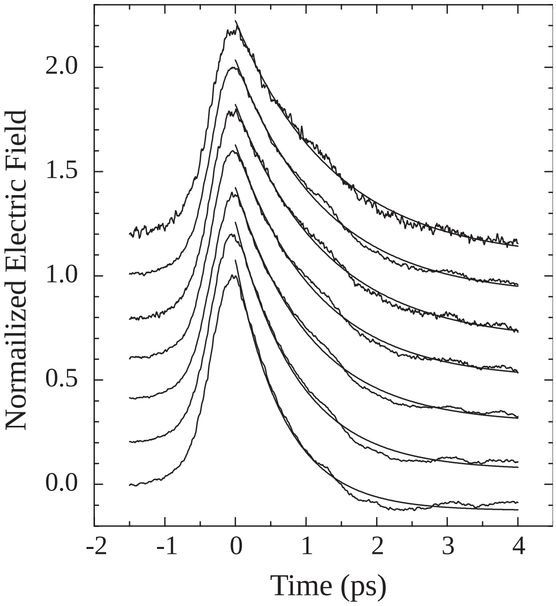

Fig. 4. Thick lines show the bias field dependence of the temporal waveforms of the THz pulses emitted from GaAs pumped with an excess energy of -35 meV. The values of the bias field are $1.3, 2.7, 4.0, 5.3, 6.7, 8.0$, and 9.3 kV/cm from the top to the bottom. Thin lines show the fit of the decaying part to an exponential function.

excitation of electrons trapped in a random potential well made by interaction with LO phonons to free carrier states.

4. Conclusion

We have studied the ultrafast dynamics of electrons photogenerated with positive and negative excess energies in GaAs and InP by observing the waveform of the emitted THz radiation. Subpicosecond intraband relaxation was observed with positive excess energies. With negative excess energies, picosecond transition from the Urbach state to free carrier states was observed. This dynamical behaviour was found to be very sensitive to the field magnitude in the range of several kV/cm.

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