Capacitive response and room-temperature terahertz gain of a Wannier–Stark ladder system in GaAs-based superlattices

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We investigate the phase and terahertz (THz) gain of Bloch oscillations in GaAs-based superlattices at various temperatures of $T = 80$–298 K by using THz emission spectroscopy under bias electric fields. The transient current is found to start from its maximum nearly as damped cos $\omega_B t$ ($\omega_B/2\pi$: Bloch frequency) throughout this temperature range, having only a small initial phase even for $kT > \hbar\omega_B$ ($\hbar$: Boltzmann constant) and dephasing time shortening with increasing temperature. A spectral analysis indicates inversionless THz gain that originates from the capacitive nature of a Wannier–Stark ladder system with broadened energy levels at room temperature. © 2016 The Japan Society of Applied Physics

In the terahertz (THz) region, there has been a great need for gain media that allow for wide tunability and room-temperature operation of compact solid-state sources.1) It is well known that $kT_c \approx \hbar \omega_B$ (kB: Boltzmann constant) provides the temperature $T_c$ above which the population inversion relevant to THz gain at photon energy $\hbar \omega_B$ cannot be easily achieved in nanostructures such as quantum cascade lasers.2,3) In a typical case where $\omega_B/2\pi \sim 2$ THz, $T_c$ is estimated to be as low as $\sim 100 \text{K}$. Several theoretical studies have predicted that a Wannier–Stark ladder system in semiconductor superlattices (SLs) will have voltage-tunable THz gain without population inversion at not only low temperatures ($kT < eFd$) but also high temperatures ($kT > eFd$) up to room temperature, when carriers have moderate scattering rates under dc bias electric field $F$ and SL period $d$.4–7) Here, $eFd$ is the energy separation between neighboring Wannier–Stark levels. This type of THz gain was also supported experimentally,8,9) and has since been demonstrated more directly by measuring the complex conductivity spectra of GaAs-based SLs at low temperatures.10–12)

In a previous study,11) we revealed that the unique phase of transient Bloch oscillations (coherent quantum beats) observed at 10 K reflects the translationally symmetric distribution of electrons onto a Wannier–Stark ladder and their capacitive response to an optically switched step-function-like bias electric field; these features are equivalent to the existence of inversionless THz gain in the steady state, where Bloch oscillations are fully damped. The capacitive response reported11) produces a cos $\omega_B t$-like current for $t > 0$, with $\omega_B/2\pi$ equal to $eFd/\hbar$, under the step-function-like bias electric field. Although Bloch oscillations in themselves can be observed at higher temperatures as well,13–15) it has never been examined how the oscillation phase and hence the THz gain in semiconductor SLs behave when $kT$ goes beyond $eFd$ with increasing temperature up to room temperature.

In this paper, we report phase-sensitive THz measurements of Bloch oscillations in GaAs-based SLs at various temperatures ranging from 80 to 298 K. The transient current induced by the femtosecond optical excitation of electrons under bias electric field $F$ was found to start from its maximum nearly as damped cos $\omega_B t$ throughout this temperature range, with $\omega_B/2\pi$ almost equal to the expected Bloch frequency $eFd/\hbar$. We obtained the initial phase remaining very small even for $kT > eFd$ as well as the dephasing time shortening gradually with increasing temperature. These features led to complex conductivity spectra that provide direct evidence for the existence of inversionless THz gain in SLs at room temperature.

The sample used here was an undoped GaAs (7.5 nm)/AlAs (0.5 nm) SL (with 59 periods) grown by molecular beam epitaxy on a Si-doped (001) GaAs substrate. In the conduction band of the sample, the first miniband had a width of 45 meV and was separated from the second miniband by a 93-meV-wide minigap, which we estimated using the Kronig–Penney model.16) We fabricated a semi-transparent NiCr film (Schottky contact) and a AuGeNi film (Ohmic contact) onto the front and back surfaces of the sample, respectively. Under dc bias voltages applied between these film electrodes, electrons were excited from the valence band to the conduction band by optical pump pulses with a duration of $\sim 100 \text{fs}$. The electron concentration was kept as low as $2 \times 10^{14} \text{cm}^{-3}$.

The transient current then flowed through the sample and emitted THz electric field into the free space. Here, THz signals were governed by electron motion because the miniband width for heavy holes is very small and the optical absorption involving light holes is relatively weak.5,10,11) The THz electric field was detected in the time domain through the Pockels effect of a 0.1-mm-thick (110) ZnTe crystal, which provided flat sensitivity up to $\sim 3.5$ THz. We performed this measurement at various sample temperatures ranging from 80 to 298 K. Considering the known temperature-dependent band gaps of the host materials,17) we adjusted the central photon energy of optical pump pulses so that electrons would be injected into nearly the same energy positions of the conduction first miniband at all temperatures.

Figure 1 shows the temporal waveforms of the THz electric field emitted from the sample at $T = 150 \text{K}$, which satisfies $kT > eFd$ for $F = 11$–13 kV/cm and $d = 8.0 \text{nm}$, with pump photon energies of $E_{pm} = 1.512$–1.538 eV. Here, both the THz emission and dc photocurrent were quenched for
The THz waveform was determined by the maximum entropy method\textsuperscript{11,12} with an accuracy of ±15 fs (vertical dashed lines). As seen in Fig. 1(a) for $E_{\text{pm}} = 1.518$ eV, the THz waveforms have an oscillatory signal whose frequency $\omega_B/2\pi$ increases with increasing bias electric field $F$ and almost equals the expected Bloch frequency $eF\tau_c$. The details of this behavior are shown in Figs. S1 (the plot of oscillation frequency versus bias electric field) and S2 (amplitude spectra) in the online supplementary data at http://stacks.iop.org/APEX/9/112101/mmedia. The THz signal can thus be ascribed to Bloch oscillations. Furthermore, as seen in Fig. 1(b) for $F = 13$ kV/cm, the THz signal exhibits no essential variation in its shape when $E_{\text{pm}}$ varies considerably; it always evolves nearly as damped $-\sin \omega_B t$ for $t > 0$. Note here that $eF\tau_c/h = 2.5$ THz and hence $T_c = 120$ K for $F = 13$ kV/cm. The features of Bloch oscillations described above are very similar to those observed previously at a much lower temperature of 10 K,\textsuperscript{11} suggesting the capacitive response of electrons distributed onto a Wannier–Stark ladder even for $kT > eF\tau_c$.

The THz waveforms measured for $\omega_B/2\pi = 2.3$ THz at (a) 80 K, (b) 150 K, and (c) 298 K are shown in Fig. 2 by filled circles with the same vertical scale. Here, electrons were injected near the bottom of the conduction first miniband by setting $E_{\text{pm}}$ to 1.540 eV at 80 K, 1.518 eV at 150 K, and 1.461 eV at 298 K. As seen in the figures, the damped oscillation continues for the longest time (~1.5 ps) at 80 K. When the temperature increases and goes beyond $T_c (= 110$ K), we find that the THz signal does not change drastically; simply, the damping is slightly faster at 150 K than at 80 K. The THz signal appears also at 298 K with even faster damping, where its oscillatory character can furthermore be supported by Fig. S3 in the online supplementary data at http://stacks.iop.org/APEX/9/112101/mmedia. Note that the magnitude of THz emission decreased monotonically with increasing temperature; this behavior is discussed later. The THz waveforms obtained with the time origin in Fig. 2 allow us to examine the oscillation phase in more detail and the existence of room-temperature THz gain.

Now, we attempt to reproduce the observed THz waveforms by assuming that the transient current has a damped cosine-like form

$$J(t) = J_0 \Theta(t) e^{-\gamma t} \cos(\omega_B t + \alpha)$$

and computing $dJ/dt$ in a manner similar to those described in Refs. 11 and 12. Here, $J_0$ is the amplitude of the current,
that phonon scattering processes were thermally the temperature increased from 80 to 298 K. This indicates revealing that the capacitive nature of a Wannier–Stark ladder system with rather broadened energy levels at temperatures up to 298 K.

In summary, we performed phase-sensitive THz measurements of Bloch oscillations in GaAs-based SLs at various temperatures of $T = 80–298$ K. The transient current was found to start from its maximum nearly as damped $\cos \omega_B t$ throughout this temperature range. Here, the dephasing time shortened gradually with increasing temperature and the initial phase remained much smaller than $\pi/2$ even for $kT > \hbar \omega_B$, revealing that the capacitive nature of a Wannier–Stark ladder system without population inversion is favorably preserved for broadened energy levels at high temperatures. The obtained complex conductivity spectra provided direct evidence for the existence of inversionless THz gain that originates from the capacitive nature of the Wannier–Stark ladder system.

Table I. Dephasing time $1/\gamma$ and initial phase $\alpha$ in Eq. (1) as the adjustable parameters for reproducing the THz waveforms in Fig. 2 observed at three different temperatures.

| Temperature $T$ (K) | Dephasing time $1/\gamma$ (ps) | Initial phase $\alpha$ (rad) |
|---------------------|-------------------------------|-----------------------------|
| 80                  | 0.31                          | 0.38                        |
| 150                 | 0.27                          | 0.38                        |
| 298                 | 0.19                          | 0.28                        |

$\Theta(t)$ is the unit step function, $1/\gamma$ is the dephasing time, and $\alpha$ is the initial phase. The simulation results for the THz electric field are shown in Fig. 2 by solid curves, and the sets of fitting parameters $1/\gamma$ and $\alpha$ are listed in Table I. The agreement between the simulated and observed THz waveforms is satisfactory at each temperature. This provides an understanding of why the magnitude of THz emission varied with temperature: the initial small positive peak appears with similar vertical values in Figs. 2(a)–2(c) and the magnitude for $t > 0$ is governed by temperature-dependent dephasing time.

The dephasing time $1/\gamma$ changed from 0.31 to 0.19 ps as the temperature increased from 80 to 298 K. This indicates that phonon scattering processes were thermally activated with temperature and they significantly broadened the Wannier–Stark levels. On the other hand, the initial phase $\alpha$ was found to be much smaller than $\pi/2 (\approx 1.57$ rad) at each temperature. This means that, even though the Wannier–Stark levels become rather broadened with increasing temperature, the transient current starts from its maximum at $t = 0$ [see Eq. (1)] in contrast to the expectation from semiclassical miniband transport. As mentioned earlier, the $\omega_B$-resonant capacitive response of electrons distributed onto the Wannier–Stark ladder without population inversion has been observed at 10 K with an initial phase of $\alpha = 0.11$. In the present case, this nature of the Wannier–Stark ladder system turned out to be preserved up to room temperature with sufficiently small initial phase, which is comparable to the value of $\omega_B \delta t = \pm 0.22$ rad given by the temporal uncertainty $\delta t = \pm 15$ fs in determining the time origin.

Because electrons were subjected to an optically switched step-function-like bias electric field in our measurement scheme,8,11) we can obtain the spectral shape of complex conductivity by performing the Fourier transform of the observed THz waveforms (Fig. 2). Figure 3 shows the complex conductivity spectra at (a) 80 K, (b) 150 K, and (c) 298 K with the same vertical scale. The monotonic decrease in the magnitude of conductivity with increasing temperature comes from that of THz emission shown in Fig. 2 and can therefore be ascribed to phonon scattering. The real part of complex conductivity changes from negative to positive values (i.e., from the gain region to the loss region) across the Bloch frequency $\omega_B/2\pi$ at each temperature owing to very small $\alpha$. The negative and positive peaks of the real part are separated by $\sim \gamma/\pi$, which also gives the full width at half maximum of the imaginary part. Thus, we have demonstrated the existence of inversionless THz gain that originates from the capacitive nature of the Wannier–Stark ladder system.

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