Phase shift of terahertz Bloch oscillations induced by interminiband mixing in a biased semiconductor superlattice

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We investigate terahertz waveforms emitted from a GaAs-based superlattice with interminiband mixing under various dc bias electric fields, injecting electrons into the conduction first miniband with femtosecond optical pulses. Bloch oscillations are observed under relatively low bias fields; surprisingly, the oscillation phase is found to shift by $\pi/2$ from those reported previously for Wannier–Stark ladder states in nearly isolated minibands. The oscillatory feature of terahertz emission is gradually destroyed under higher bias fields. We show that the observed $\pi/2$ phase shift can be ascribed to significant delocalization of electron wavefunctions driven by interminiband mixing.

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Semiconductor superlattices (SLs) are known to have a series of voltage-tunable equidistant energy levels, called a Wannier–Stark ladder,1–5 when a relevant SL miniband in the energy band structures can be regarded as isolated under dc bias voltage.6–7 Recent phase-sensitive measurements of Bloch oscillations (quantum beats) in GaAs-based SLs have revealed that electrons distributed onto a Wannier–Stark ladder without population inversion possess a capacitive nature and exhibit a unique oscillation phase,8–10 which is equivalent to the existence of a voltage-tunable terahertz gain (called the Bloch gain) in the steady state,11–14 up to above room temperature. On the other hand, Wannier–Stark ladder states are not well defined theoretically as energy eigenstates in the situation where interminiband mixing is substantial;6,7,15 electrons exhibit Zener tunneling16 into higher minibands under high-bias conditions depending on SL structures, as confirmed by several types of experiments.7,17–22 However, it is not well understood how interminiband mixing affects the phase of Bloch oscillations in biased SLs.

In this letter, we report terahertz waveforms emitted from a GaAs-based SL whose conduction first miniband is expected to significantly mix with higher minibands under dc bias electric fields. Electrons injected into the conduction first miniband by femtosecond optical pulses exhibited Bloch oscillations under relatively low bias fields. Surprisingly, the oscillation phase was found to shift by $\pi/2$ from those reported previously for Wannier–Stark ladder states in nearly isolated minibands.8–10 The oscillatory feature of terahertz waveforms was gradually destroyed and varied to a monotonic feature as the bias field was increased to much higher values. By computing the energy eigenstates of electrons under bias field, we show that the observed $\pi/2$ shift in the oscillation phase can be ascribed to significant delocalization of electron wavefunctions driven by interminiband mixing.

The sample in our experiment was an undoped GaAs (16.3 nm)/Al0.1Ga0.9As (1.0 nm) SL grown with 59 periods by molecular beam epitaxy on a Si-doped GaAs (001) substrate. By using the Kronig–Penney model, we estimated that the first miniband in the conduction band had a width of 14.8 meV, the second miniband had a width of 47.9 meV, and they were separated by a minigap as narrow as 8.4 meV.23–28 Under dc bias voltages applied between the front and back surfaces of the sample at 80 K, electrons were excited near the bottom of the conduction first miniband by optical pulses (with a duration of $\sim100$ fs) delivered from a mode-locked Ti:sapphire oscillator. The electron density was kept as low as $\sim2 \times 10^{14}$ cm$^{-2}$ to minimize the field screening effect and the dynamic depolarization effect.29 The transient terahertz electric field emitted from the sample was detected in the time domain using a ZnTe electro-optic sensor with flat sensitivity up to $\sim3$ THz. The time origin ($t = 0$) of terahertz emission was determined by the maximum entropy method$^{8,30}$ with an...
accuracy of ±15 fs. More details of our experimental method can be seen in Ref. 9.

Figure 1 shows the terahertz waveforms emitted from the sample under different bias fields \( F \) of up to 11 kV cm\(^{-1}\). We find three pronounced features in these waveforms. First, damped oscillations within a few cycles appear for relatively low bias fields (\( F = 0.88\)–4.8 kV cm\(^{-1}\)). The terahertz field has the largest peak at exactly \( t = 0 \), and exhibits oscillation period shortening with increasing bias field. This suggests that, as compared with previous reports on Bloch oscillations occurring in nearly isolated minibands,\(^8,9\) damped oscillations observed here have a similar behavior of resonance frequency but a significantly different initial phase. A quantitative analysis of the terahertz waveforms will be performed later to confirm this feature. Second, the terahertz field has a monocyclic shape for a high bias field (\( F = 11 \) kV cm\(^{-1}\)), changing from positive to negative values and then fading away. This indicates that electrons are accelerated initially in the conduction first miniband and subsequently undergo Zener tunneling into higher minibands.\(^7\) Finally, the crossover from the first feature to the second feature shows up for medium bias fields (\( F = 6.8\)–8.7 kV cm\(^{-1}\)), meaning that part of the electrons perform damped oscillations in the presence of interminiband Zener tunneling. These three features are consistent with those of photocurrent spectra, as shown in Fig. S1 in the online supplementary data (stacks.iop.org/APEX/12/041003/mmedia).

Below, we examine the first feature described above for low bias fields. The terahertz waveforms observed with apparently pure damped oscillations under \( F = 2.8, 3.3, \) and 3.8 kV cm\(^{-1}\) are shown by circles in Figs. 2(a)–2(c). To reproduce these damped oscillations, we assume that transient current has the form of

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

where \( J_0 \) is the magnitude of current, \( \Theta(t) \) is the unit step function, and \( \omega_B, \gamma, \) and \( \alpha \) are to be determined as fitting parameters. We assume the terahertz waveform in each bias field is reproduced by damped sinusoidal currents (with a temporal resolution of \( \tau_{\text{res}} = 0.29 \) ps). The oscillation phase can be interpreted in terms of (d) electron motion on the miniband energy-momentum dispersion curve, which is expected to be suitable for electrons with spatially extended envelope wavefunctions.

![Fig. 2.](Color online) Terahertz waveforms for bias fields \( F \) of (a) 2.8 kV cm\(^{-1}\), (b) 3.3 kV cm\(^{-1}\), and (c) 3.8 kV cm\(^{-1}\), reproduced by damped sinusoidal currents (with a temporal resolution of \( \tau_{\text{res}} = 0.29 \) ps). The oscillation phase can be interpreted in terms of (d) electron motion on the miniband energy-momentum dispersion curve, which is expected to be suitable for electrons with spatially extended envelope wavefunctions.

![Fig. 3.](Bias-field dependence of fitting parameters obtained from the waveforms in Fig. 2. (a) Resonance frequency \( \omega_B/2\pi \) (circles). (b) Initial phase \( \alpha \) (squares) and relaxation time \( 1/\gamma \) (triangles). The dashed line in (a) shows the expected Bloch frequency \( eFd\hbar \) versus bias field \( F \). Indicated by the dotted line in (b) for comparison is an initial phase reported in Ref. 9 with \( F = 11 \) kV cm\(^{-1}\).)
step function, $1/\gamma$ is the relaxation time, $\omega_B/2\pi$ is the resonance frequency, and $\alpha$ is the initial phase. The simulated $dJ/dt$ can be compared with the measured terahertz fields after $dJ/dt$ is convolved with a system response function that characterizes the temporal resolution of $\tau_{\text{res}} = 0.29$ ps due to the finite bandwidth ($\sim 3.5$ THz) of our experimental setup mentioned earlier.

The simulation results for the terahertz field are shown in Figs. 2(a)–2(c) by curves, and the corresponding sets of fitting parameters $\omega_B/2\pi$, $\alpha$, and $1/\gamma$ are plotted in Fig. 3 by symbols.\(^{31}\) The agreement between the simulated and observed terahertz waveforms in Fig. 2 is excellent for the three low bias fields $F$ of 2.8, 3.3, and 3.8 kV cm\(^{-1}\). As seen in Fig. 3, the resonance frequency $\omega_B/2\pi$ matches well with the expected Bloch frequency $ef/dh$ (dashed line) for the present SL period of $d = 17.3$ nm, and the initial phase $\alpha$ nearly equals $-\pi/2$. Thus, we find that Bloch oscillations accompanied by the damped $\sin \omega_B t$-like current indeed occurred in the present narrow-minigap SL. Note that the oscillation phase observed here has a peculiar shift by $\pi/2$ from those reported previously for Wannier–Stark ladder states in nearly isolated minibands, where damped $\cos \omega_B t$-like currents with $\alpha \sim 0$ [e.g., $0.12\pi$ as indicated by the dotted line in Fig. 3(b) for the case of a GaAs (7.5 nm)/AlAs (0.5 nm) SL at 80 K] are expected to flow.\(^{8,9,30}\) The relaxation time $1/\gamma$ shortens monotonically with increasing bias field, reflecting the suppression of Bloch oscillations due to interminiband Zener tunneling.

To discuss the physical reason for the observed $\pi/2$ phase shift, we computed the energy eigenstates of electrons under bias fields by using the finite element method. Envelope wavefunctions with large amplitudes in several neighboring quantum wells under $F = 2.8$ kV cm\(^{-1}\) are shown in Fig. 4(a); they are compared with ones reported previously for Wannier–Stark ladder states in nearly isolated minibands [see, e.g., Fig. 4(b) for the case of a GaAs (7.5 nm)/AlAs (0.5 nm) SL].\(^{10}\) We find that the envelope wavefunctions in Fig. 4(a) are significantly delocalized into downstream quantum wells, and are slightly different in shape from one another, whereas those in Fig. 4(b) exhibit clear localization and exactly the same shapes with the mutual spatial shift by $d = 8.0$ nm (i.e., Wannier–Stark localization). This suggests that the oscillation phase seen in Figs. 2(a)–2(c) is governed by a physical mechanism other than the capacitive mechanism\(^{8}\) based on the translationally symmetric distribution of electrons created by an optical pulse under Wannier–Stark localization; the damped $\sin \omega_B t$-like current can be regarded as a conductive response of delocalized electrons (i.e., a true current rather than displacement current) to the $\sin \omega_B t$ component of an optically
switched step-function-like effective bias input in our measurement scheme.8)

Taking the electron delocalization into account, let us interpret the observed oscillation phase by adopting the miniband transport model, shown schematically in Fig. 2(d). Here, electrons are created near the bottom of the conduction first miniband at \( t = 0 \) with spatially extended envelope wavefunctions, and they gain momentum, \( \hbar k_z \), owing to their ballistic acceleration along the SL axis (\( z \) axis) under bias field.8) Electrons then experience either Bragg reflection or interminiband Zener tunneling at the wavenumber of \( k_z = n\pi/d \). Thus, part of the electrons are expected to be reflected with \( k_z = -n\pi/d \) and to exhibit Bloch oscillations, producing a damped \( \sin \omega_d t \)-like true current (because they are created initially at \( k_z \approx 0 \)). This interpretation allows us to conclude that the significant delocalization of electron wavefunctions in Fig. 4(a) driven by interminiband mixing is essential for the observed \( \pi/2 \) shift in the oscillation phase.

Finally, we would like to comment on the historical background of the miniband transport model in Fig. 2(d). This model was originally provided by Zener16) in 1934 and has often been used to discuss oscillation phenomena observed with resonance frequencies nearly equal to \( eF/dh \).7) However, the oscillation phase predicted by the miniband transport model has never been realized by experiments, where electrons are usually localized even at the moment of optical pulse excitation.8) Our experiment has demonstrated that the oscillation phase expected by Zener indeed appears in the absence of Wannier–Stark localization and has suggested that the miniband transport model is suitable for treating the crossover between Bloch oscillations and interminiband Zener tunneling. The tunneling probability is given by

\[
P_t = \exp[-m^*\varepsilon_g^2/(4\hbar^2 eF)],
\]

where \( m^* \) is the electron effective mass and \( \varepsilon_g \) is the relevant miniband width.16) The uniqueness of the present narrow-minigap SL lies in its relatively high tunneling probability of \( P_t \gtrsim 0.1 \) throughout the bias field range studied here; ordinary SLs with nearly isolated minibands satisfy \( P_t \ll 0.1 \).

In summary, we measured and analyzed terahertz waveforms emitted from a GaAs-based SL with interminiband mixing under dc bias electric fields. Electrons created near the bottom of the conduction first miniband by femtosecond optical pulses exhibited Bloch oscillations under relatively low bias fields as well as the crossover from Bloch oscillations to interminiband Zener tunneling under higher bias fields. The key result is that the oscillation phase had a peculiar shift by \( \pi/2 \) from those reported previously for Wannier–Stark ladder states in nearly isolated minibands. By computing the energy eigenstates of electrons under bias field, we showed that the observed \( \pi/2 \) phase shift can be ascribed to significant delocalization of electron wavefunctions driven by interminiband mixing. Thus, our findings offer a deep insight into the fundamental aspect of electron transport under bias field and also wider possibilities of tailoring ultrafast true and displacement currents for optoelectronic applications on the basis of electron wavefunctions.

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