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Coupled Bloch-Phonon Oscillations in Semiconductor Superlattices

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We investigate coherent Bloch oscillations in $GaAs/Al_xGa_{1-x}As$ superlattices with electronic miniband widths larger than the optical phonon energy. In these superlattices the Bloch frequency can be tuned into resonance with the optical phonon. Close to resonance a direct coupling of Bloch oscillations to LO phonons is observed which gives rise to the coherent excitation of LO phonons. The density necessary for driving coherent LO phonons via Bloch oscillations is about 2 orders of magnitude smaller than the density necessary to drive coherent LO phonons in bulk GaAs. The experimental observations are confirmed by the theoretical description of this phenomenon

The coupling of fundamental excitations is one of the most intriguing subjects in solid state physics. Such coupling phenomena involving light, phonons, charge carriers, and electric fields give rise to new physical phenomena such as phonon-polaritons [1], exciton-polaritons [2], and plasmon-phonon modes [3]. In compound semiconductors relevant for optoelectronic and electronic applications, Fröhlich coupling, i.e., the interaction of electrons with polar lattice vibrations (LO phonons) mediated by an electric field, is well known to determine some of the key physical properties [4], e.g., energy relaxation times of electrons as short as 100 fs [5]. A new coherent excitation has been realized in semiconductor heterostructures with an artificial periodicity: coherent, tunable electronic Bloch oscillations which are excited by ultrashort laser pulses [6,7] via the coherent superposition of several Wannier-Stark states in a biased semiconductor superlattice (SL) [8,9]. These electronic wave packets perform spatial oscillations along the growth direction of the SL [7,10]. Bloch oscillations—although not an elementary excitation—are an elementary dynamical phenomenon of a particle in a periodic potential under the influence of a constant force—as predicted in general for solids early by Zener and Bloch [11] and for SLs by Esaki and Tsu [12]. The Bloch frequency ν_B can be tuned by an applied electric field according to $\nu_B = eFd/h$, where d is the SL period, F an externally applied electric field, e the electron charge, and h Planck's constant. The frequency range in which Bloch oscillations can be observed is given by a lower limit determined by the dephasing rate and an upper limit due to the finite miniband width of the SL. Here, we report on a strong coupling between Bloch oscillations and LO phonons in wide miniband SLs when both are brought into resonance. Close to resonance this coupling leads to a transfer of electronic coherence to the lattice.

Theoretically the influence of LO phonons on coherent Bloch oscillations has been addressed with emphasis on the influence of *scattering* on the electronic coherence [13,14]. The influence of the Wannier-Stark-phonon resonance in SLs has been treated theoretically by Govorov [15] with a prediction on changes of the absorption line shape of the interband transitions. Anomalies in the transport properties have been proposed to occur under electron-phonon resonance conditions [16,17]. Recently, Ghosh *et al.* theoretically treated the coupling of plasmons, Bloch oscillations, and optical phonons in a SL [18].

The experimental techniques applied are femtosecond time-resolved optical pump and probe techniques [5]. The laser source is a Kerr-lens mode-locked Ti:sapphire laser delivering optical pulses of 50 fs duration corresponding to a spectral width of 40 meV. Two different SLs are investigated: Sample A (sample B) consists of 35 periods GaAs wells of 5.1 nm (6.7 nm) width and $Al_{0.3}Ga_{0.7}As$ barriers of 1.7 nm (1.7 nm) width. The coupling of the lowest quantized electronic levels in the quantum wells forms an electronic miniband of 60 meV width (sample A) and 36 meV width (sample B) which is for sample A significantly larger than the LO phonon energy of 36 meV in GaAs [19]. The large miniband width of sample A allows us to tilt the potential so strong that the spacing between Wannier-Stark states becomes larger than the phonon energy before these states become fully localized in one well, in other words, that the Bloch frequency can be tuned above the LO phonon resonance. For applying a static electric field, the SLs are embedded in a *p-i-n* diode (sample A) or a Schottky diode (sample B). The coupled modes of Bloch oscillations and phonons give rise to a coherent longitudinal polarization along the growth direction of the SL. Associated periodic changes in the reflectivity and transmission are traced in the time domain by a time-delayed probe pulse. These techniques are known as reflective/transmitive electro-optic sampling (REOS/TEOS) based on a polarization analysis of the probe pulse [20].

Figure 1(a) shows the time-resolved oscillatory traces of the REOS data recorded at 10 K for sample A. The

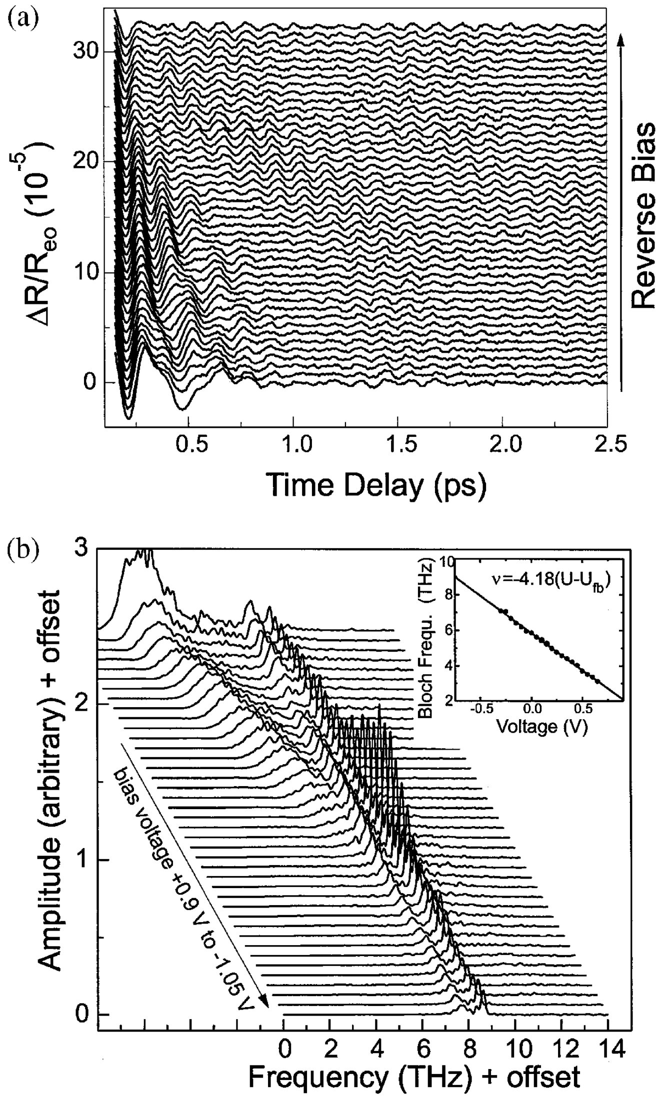

FIG. 1. (a) Extracted oscillatory traces of REOS signals recorded in sample A at 10 K for reverse bias voltages of 0.9 V (bottom) to -1.05 V (top) in increments of -0.05 V. (b) Fourier transforms of the data in (a), not normalized. The inset shows the Bloch frequency versus the applied bias voltage extracted from (b).

laser spectrum is centered between the 0 and -1 Wannier-Stark transition and the estimated excitation density corresponds to 44×10^{14} electron-hole pairs per cm^{-3}. The range of the bias voltage applied to the *p-i-n* diode is $+0.9$ to -1.05 V. The laser wavelength is not corrected for the quantum confined Stark effect of the Wannier-Stark transitions for higher electric fields. For positive voltages the signals exhibit a rapid dephasing with a frequency of 3 THz and a dephasing time in the sub-ps range. For decreasing voltage, i.e., increasing electric field in the diode, the frequency rises and the associated amplitude rapidly drops. At higher electric fields the signal is dominated by a long-lived oscillation with a frequency of 8.8 THz and a dephasing time of 10 ps. In Fig. 1(b) the corresponding numerical Fourier transforms are shown. The spectra show several features.

(i) For all applied voltages a small contribution at the TO and LO phonons of 8.0 and 8.8 THz, respectively, is observed. This feature stems from the excitation of coherent plasmon-phonon coupled modes in the highly doped cladding layers of the *p-i-n* diode [21,22]. This contribution is the same at low and high fields thus strongly suggesting that it is independent of the bias applied.

(ii) A broad spectral peak appears at low frequencies (3 THz) at the lowest applied voltage and shifts proportional to the increasing internal electric field. The center of the peak is plotted versus the applied voltage in the inset of Fig. 1(b). The frequency is linear proportional to the applied voltage and can thus be clearly identified with Bloch oscillations. The observed slope of -4.18 THz/V is in accordance with the theoretical slope of $ed/hl = -4.2$ THz/V for this sample, where l is the width of the intrinsic layer of the *p-i-n* diode. The central Bloch peak can be identified unambiguously between 3 to 7 THz with an error bar of 0.5 THz. For higher frequencies the Bloch peak overlaps with the phonon resonance so that no frequency can be assigned. This does not imply that no Bloch oscillations are excited, but rather that due to their rapid dephasing and the frequency overlap with the phonons they cannot be extracted from the data. The miniband width of this sample should allow one to excite Bloch oscillations above the phonon resonance up to 60 meV. Another limit, however, is given by the spectral width of the laser, which should allow the excitation and detection up to energies of approximately 40 meV (10 THz). The complete absence of Bloch oscillations above the phonon frequency is attributed to the faster dephasing and the dispersion of the electro-optic coefficient, which strongly decreases above the phonon frequency. The slope shown in the inset of Fig. 1(b) exhibits no deviation from the expected Bloch slope even close to the phonon resonance, which is in contrast to the dispersion obtained for plasmon-phonon coupling.

(iii) A contribution is observed at the LO phonon frequency of 8.8 THz which strongly depends on the applied bias. This contribution is associated with the long-lived oscillation observed in the time domain data. The latter feature is attributed to LO phonons which are coherently excited by the coupling to Bloch oscillations. In order to support this interpretation we plot the amplitude of the Bloch component and of the coherent LO phonons versus the Bloch frequency in Fig. 2. The amplitude of the Bloch oscillations shows a sharp drop at the lowest frequencies (<4 THz) and a slower decrease with rising frequency. The LO phonon amplitude shows a constant background from the coherent plasmon-phonon contribution from the cladding layers [see (i)], a pronounced resonance close to 7.5 THz, and a rapid drop towards a minimum close to the LO phonon. Above the LO phonon a second smaller maximum is observed. The observed resonances are attributed to the interaction of the LO phonons with the Bloch oscillations. Under the assumption of field-independent dephasing times of the Bloch oscillations the theory predicts

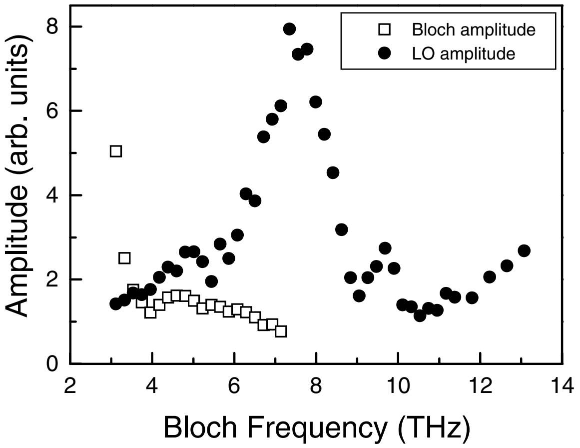

FIG. 2. Amplitude of the Bloch oscillations (open squares) and of the LO phonons (solid circles) versus the Bloch frequency.

a maximum excitation of LO phonons under resonance conditions ($\nu_{\mathrm{LO}} = \nu_{\mathrm{B}}$) due to parametric excitation [18]. However, the electronic dephasing time strongly increases under resonance conditions due to LO phonon assisted relaxation between the Wannier-Stark levels [14]. This effect will lead to a strongly reduced electronic driving force for the lattice polarization in resonance and thus to a drop of the coherent LO phonon amplitude. This effect is strongest in resonance. Theoretically this effect is obtained by incorporating a field-dependent dephasing time for the Bloch oscillations [18]. Above the phonon resonance, the dephasing time is expected to rise again rapidly since the resonance condition is not strictly fulfilled anymore [14]. We thus attribute the second smaller maximum of the phonon amplitude above the resonance at 9.5 THz to this effect.

The observed coupling exhibits an analogy to plasmon-phonon coupling which leads to coupled modes below and above the phonon resonance [3]. The resonance condition, i.e., the electron density at which the plasma frequency equals the LO phonon frequency, is given in bulk GaAs when the electron densities are 6×10^{17} carriers per cm^3. The excitation density in the presented experiments corresponds to $4 \times 10^{14}\ \mathrm{cm}^{-3}$. Thus resonant Bloch-phonon coupling is observed at more than 2 orders of magnitude lower densities indicating that the observed coupling relies fully on the coherent polarization associated with Bloch oscillations. At this density no screening of the LO phonon can occur which—in the case of plasmon-phonon coupling—leads to the appearance of the screened LO phonon at TO frequency above resonance. The bias-independent contribution, which exhibits LO and TO components, stems from the excitation of a coherent plasmon-phonon coupled mode in the highly doped GaAs cladding layers of the *p-i-n* diode [22] which are excited since the laser energy is approximately 100 meV above the GaAs band gap. The absence of a bending of the slope of the frequency obtained for the Bloch oscillations suggests that there is no anticrossing of the Bloch oscillations with the optical phonon resonance in agreement with the theory [18].

The main findings derived from sample A are confirmed by experiments on sample B. Although this sample has a miniband width equal to the LO phonon energy, i.e., the wave packets become strongly localized in resonance, the higher sample quality associated with larger wells allows the observation of Bloch oscillations with longer dephasing times than in sample A. Even at room temperature dephasing times in the range of 200–300 fs are observed. Figure 3(a) shows the Fourier transforms obtained from the time-resolved data from sample B at 300 K. The broad Bloch peak reveals the fast dephasing of the electronic coherence at room temperature due to the scattering with thermally activated LO phonons. When the Bloch peak approaches the optical phonon resonance a pronounced and sharp peak appears at the LO phonon. No bias-independent contribution at the TO or LO phonon is observed as in sample A, since the sample is a Schottky diode with the SL embedded in undoped $\mathrm{Al_{0.3}Ga_{0.7}As}$ cladding layers. Figure 3(b) shows the plot of the amplitude of the LO phonons and of the Bloch oscillations versus the Bloch frequency. The Bloch amplitude shows the expected drop due to the strong localization of the wave functions, while the phonon amplitude rises strongly when the Bloch frequency comes

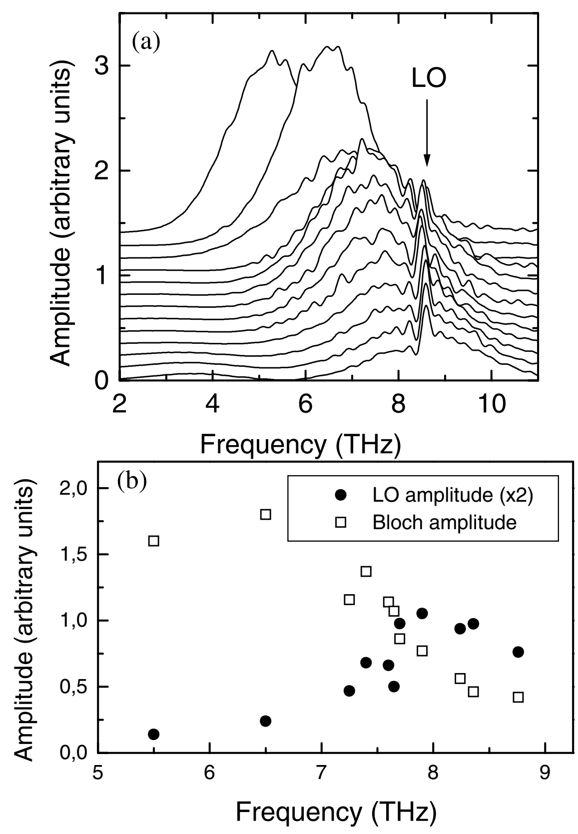

FIG. 3. (a) Fourier transforms of coupled Bloch-phonon spectra recorded at room-temperature for sample B for bias voltages of −2.5 V, −3 V until −3.9 V in increments of −0.1 V. (b) Phonon (solid circles) and Bloch amplitudes (open squares) versus the Bloch frequency.

close to resonance. A full resonance behavior cannot be resolved due to the localization of the wave packets at the miniband width of 36 meV. However, again the coherent excitation of LO phonons at very low excitation densities due to the Bloch-phonon coupling is confirmed. The onset of the maximum of the phonon amplitude is not as much shifted to lower frequencies below the phonon resonance as in sample A. This difference is due to the strong localization of the wave functions in the 36 meV sample close to resonance thus strongly reducing the effect of LO phonon assisted transitions which contribute to a rapid dephasing of the Bloch oscillations in sample A [23].

Another analogy to the experiments presented here may be found in doubly resonant Raman scattering in SLs [24] or in resonant polaron coupling between Landau levels with a splitting of one LO phonon energy [25]. However, the observations reported here are based on the fact that the electronic system is excited coherently, which is not the case in continuous wave experiments.

With regard to the observation of THz electromagnetic emission from Bloch oscillations [7] and coherent phonons [26], we expect that the SL biased into the Bloch-phonon resonance is a strong emitter of electromagnetic radiation at the LO phonon frequency. Because of the longer dephasing time of the lattice mode, room temperature emission can be obtained for times exceeding the lifetime of pure electronic Bloch oscillations by an order of magnitude [27]. This effect might provide a way to obtain amplification of the emission from this system. We further propose that the observation of strongly damped Bloch oscillation via the coupling to LO phonons is a feasible way to prove the existence of Bloch oscillations in bulk semiconductors.

In conclusion we presented the observation of the coupling of coherent electronic wave packets with LO phonons in semiconductor SLs of different miniband widths. We found that Bloch oscillations tuned into resonance with LO phonons provide an effective way for the coherent excitation of optical phonons. The exact position of the maximum phonon amplitude strongly depends on the dephasing time of the electronic coherence.

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