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Optical investigation of Bloch oscillations in a semiconductor superlattice

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We report the study of optical dephasing of Wannier-Stark ladder excitations in a semiconductor superlattice by means of transient degenerate four-wave mixing. We observe pronounced modulations of the signal with a time period varying linearly with the electric field. The time period is found to equal the temporal periodicity of Bloch oscillations, in agreement with theory. In addition, we find that the dephasing time decreases with increasing localization of the Wannier-Stark states, which is attributed to carrier escape out of the lowest miniband.

In solid-state physics, the temporal dynamics of a Bloch electron in the presence of an electric field is a fundamental quantum-mechanical problem. The so-called acceleration theorem

$$\hbar\dot{\mathbf{k}}=e\mathbf{F} \tag{1}$$

states that the quasimomentum of an individual Bloch state varies linearly in time at a rate proportional to the electric field.[1-3] As a consequence of the crystal symmetry, however, the Bloch electron is Bragg reflected as soon as it reaches a boundary of the Brillouin zone. Thus, the constant electric field induces an oscillatory electronic motion in k space. The time period for these so-called "Bloch oscillations" is given by

$$\tau_b=\frac{h}{eFd}, \tag{2}$$

where d is the length of a unit cell.

The treatment of the problem in terms of Wannier-Stark (WS) states[4,5] is the stationary counterpart of the time-dependent description above. The application of an electric field causes a gradual localization of the initially delocalized wave functions (Stark localization). The continuous spectrum of band states is then replaced by a series of equally spaced levels called the Wannier-Stark ladder:

$$E_\nu=E_0+\nu eFd,\quad \nu=0,\pm 1,\pm 2,\ldots, \tag{3}$$

where, to a first approximation, E_0 is the quantized energy of an isolated atomic site. Note that the ladder spacing, eFd, is related to the Bloch oscillation period τ_b through Eq. (2).

Up to now, neither WS states nor Bloch oscillations have been observed in conventional bulk solids. Their occurrence in realistic bulk samples is prevented by the fact that the coherence lifetime of an electron is shorter than the period of the oscillatory motion, τ_b, for all reasonable values of the electric field. The limiting factors for the electron dephasing are given by (i) scattering due to phonons, impurities, etc., and (ii) interband tunneling due to the virtual binding character of the WS states.[5]

Bleuse, Bastard, and Voisin[6] predicted the observation of a WS ladder structure for a strongly coupled semiconductor multiple quantum well structure. In such a superlattice, so-called minibands are formed in the growth direction. The effects of an applied constant electric field on the electronic superlattice states are described in the same way as for electrons in bulk solids, i.e., by Eqs. (1)-(3). In this case, however, d is the superlattice period and E_0 is the quantized energy of an isolated quantum well state. In fact, a number of groups[7-10] have observed the field-induced transition from the miniband to the evenly spaced WS ladder in superlattice structures using steady-state optical experiments.

In order to observe the coherent quantum dynamics of a nonstationary state, excitation with a short laser pulse whose spectrum encompasses two or more resonances is required.[11] Thus, generation of Bloch oscillations in a WS ladder can be achieved by using short pulses exciting more than one WS resonance. Once the Bloch oscillations are generated, they may be detected in the time domain by transient degenerate four-wave mixing (DFWM) in the spontaneous photon echo configuration, as suggested recently.[12] However, experimental generation and detection of Bloch oscillations is still lacking in bulk solids as well as superlattices.

In this paper, we report the first transient DFWM experiments on a superlattice with an applied electric field. In the WS regime, the temporal evolution of the DFWM signal exhibits modulations with peak to peak intervals equal to τ_b. A calculation of the DFWM signal using the given experimental parameters leads to the conclusion that we observe Bloch oscillations. In addition, we find that the dephasing rate $\Gamma=1/T_2$ increases for high electric fields. Carrier escape out of the lowest miniband is probably responsible for this lifetime shortening.

The sample used in the experiments was grown by molecular-beam epitaxy on an n-doped GaAs substrate. The superlattice structure is located in the intrinsic region of a p-i-n diode. It consists of 91 periods of 95-Å GaAs and 15-Å $Al_{0.3}Ga_{0.7}As$. The sample was then processed into $200\times 200\ \mu m^2$ mesas and mounted on sapphire. The GaAs substrate was removed by wet etching to allow transmission experiments. We use a tandem synchronously pumped LDS-751 dye laser system emitting-pulses of about 500 fs duration and, alternatively, a mode-locked Ti-sapphire laser emitting pulses of 110 fs (spectral width 20 meV). All experiments are performed at a crystal temperature of about 5 K.

We use a two-pulse self-diffraction technique,[13] where the sample is excited by two subsequent laser pulses with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ at times $t=0$ and $t=\tau$, respectively. Light is then emitted into the direction $\mathbf{k}_d=2\mathbf{k}_2-\mathbf{k}_1$, due to the nonlinear optical interaction in the sample. We detect the time-integrated light signal by using a slow photodetector. The decay of this DFWM signal with increasing time delay τ is determined by the loss of coherence of the excited transition, i.e., it reflects the dephasing time T_2.

Photocurrent spectra of the superlattice sample (not shown here) exhibit, at intermediate electric fields, peaks corresponding to optical transitions of the WS ladder.[14] In this regime, the heavy-hole (hh) valence-band states are already fully localized, whereas the electron states in the conduction band are still delocalized over several superlattice periods. Accordingly, "oblique" transitions S_ν are possible between a particular localized hh valence-band state and a partially delocalized electron conduction-band state, centered in a quantum well which is ν periods away.[7–10] The observed peak positions of the "interwell" transitions S_{-1}, S_{-2}, and also S_{-3} are drawn as circles in the fan chart of Fig. 1(b). According to Eq. (3),

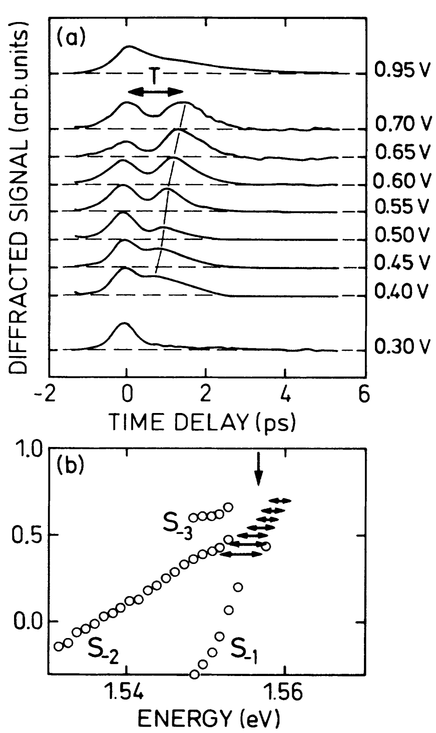

FIG. 1. (a) DFWM signal vs time delay between the two exciting laser pulses for several voltages in the WS ladder regime, showing modulations of period T. (b) The peak positions of "oblique" transitions are plotted as circles vs the applied voltage. The energy intervals h/T, with T from (a), are shown as horizontal arrows.

the energetic spacing between adjacent WS transitions amounts to eFd. It is important to note that the photocurrent measurements of peak positions in Fig. 1(b) were made under the same conditions as the DFWM experiments (i.e., with pulsed laser excitation), where carrier densities of about 5×10^9 cm^{-2} are created. This is a necessary prerequisite for a direct comparison of the transient DFWM curves shown in Fig. 1(a) and the spectrally resolved fan chart of Fig. 1(b), since space charge effects lead to a partial screening of the applied electric field.[15]

In Fig. 1(a), the temporal evolution of the DFWM signal is shown for several applied voltages at forward bias. As indicated by the vertical arrow in the fan chart of Fig. 1(b), the laser frequency is centered in the regime of oblique optical transitions of the WS ladder. For an applied voltage of 0.95 V, the decay of the DFWM signal is approximately exponential and does not show any extra features. The corresponding time constant has about the same value as in the case of an isolated quantum well.[16] In the voltage range from 0.7 to 0.4 V, however, a pronounced modulation is observed in each DFWM curve. The time duration T between the observed peaks decreases with increasing electric field and ranges from 1.4 ps for 0.7 V to about 0.7 ps for 0.4 V. In order to identify the origin of these modulations, the energetic intervals h/T are drawn as horizontal arrows in the WS fan chart of Fig. 1(b). For applied voltages less than 0.5 V, we can directly compare the calculated h/T values (obtained from the transient DFWM experiment) with the energetic spacing eFd between the S_{-1} and S_{-2} transitions (obtained from the photocurrent spectrum) and find good agreement. For applied voltages larger than 0.5 V, the h/T values decrease as expected from the tendency of the WS ladder spacings.[17] The agreement of h/T with eFd means that the observed modulation times T are equal to the time periods τ_b expected for Bloch oscillations [Eq. (2)].

It should be possible to observe more than one oscillation period by using shorter pulses. We thus also perform transient DFWM experiments on the biased superlattice by using the mode-locked Ti-sapphire laser. In Fig. 2, the temporal evolution of the DFWM signal is shown for an experimentally determined WS spacing of about 6.2 meV, which is comparable to the situation at 0.4 V in Fig. 1. As indicated by the arrows in the upper part of Fig. 2, the spectrum of the laser pulse now encompasses about five WS transitions. Indeed, the DFWM curve now exhibits three peaks for time delays of 0, 0.7, and 1.4 ps. With $eFd=6.2$ meV, the time period τ_b expected for Bloch oscillations amounts to 0.67 ps [Eq. (2)], again in good agreement with the observed oscillation period of $T=0.7$ ps. As we will show in the following discussion, we can indeed associate the observed modulations in the DFWM signal with Bloch oscillations of electrons in the superlattice.

Zakharov and Manykin[18] have theoretically investigated the effect of electric fields on the spontaneous photon echo signal measured in a transient DFWM experiment in a bulk semiconductor. More specifically, von Plessen and Thomas have recently treated Bloch oscillations in semiconductor superlattices.[12] In the following, we summarize

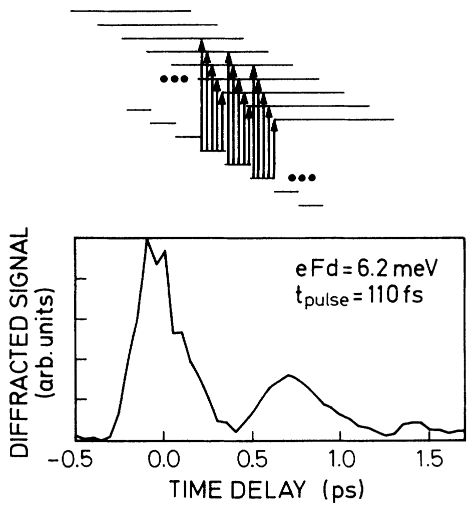

FIG. 2. DFWM signal vs time delay for $eFd = 6.2$ meV using 110-fs laser pulses. Under this condition, optical transitions from each localized *hh* state to about five conduction-band WS states takes place as illustrated in the upper part.

their main results. In k space, the ensemble of vertical optical transitions ($\Delta k = 0$) between the valence-band (miniband) states and the conduction-band (miniband) states can be viewed as an inhomogeneously broadened ensemble of two-level systems with transition frequencies $\omega(k)$. When a spectrally broad (short) laser pulse excites all of these two-level systems at $t = 0$, their dipole moments are initially in phase leading to a macroscopic (first order) polarization. Even without an applied electric field, the phases $\omega(k)t$ of the individual dipole moments evolve differently in time due to the different (but fixed) phase velocities $\omega(k)$. As a consequence, the macroscopic polarization vanishes. However, for uncoupled two-level systems the second pulse at $t = \tau$ leads to a zero phase shift at $t = 2\tau$. The recovered macroscopic (third order) polarization then leads to the emission of a photon echo. With an applied electric field, the temporal variation of the quasimomentum $k(t)$ [Eq. (1)] implies that the phases $\int \omega(k(t))dt$ of the dipole moments now evolve according to the temporally varying phase velocities $\omega(k(t))$. As a result, the nonlinear interaction due to the second laser pulse generally does not lead to a perfect zero phase shift at the instant the photon echo should arise ($t = 2\tau$). Only for specific values of the field, such that the electron is able to traverse the entire Brillouin zone and return to the initial state during the time between the pulses, i.e., for $\tau = \tau_b$, the radiators will again come to be in phase at $t = 2\tau$. The time-integrated photon echo signal should therefore be a periodic function of the time interval between the pulses with a time period equal to τ_b, as observed in the experiments presented here. For completeness, we note that this signal modulation can, alternatively, be understood in terms of quantum beats between different WS ladder transitions. However, in the WS ladder picture, it seems more difficult to anticipate the shape of the modulation, since the present experiment involves multiple optical transitions, which are coupled to each other and thus constitute a very complex system (see upper part of Fig. 2).

The above theoretical treatments were restricted to the case where the excitation pulses are much shorter than the period of Bloch oscillations. To analyze the experimental situation presented in Fig. 1 more quantitatively, we drop this restriction, using the formalism of Ref. 19 developed for transient DFWM. The result of this procedure is then specialized to the case of a two-band tight-binding model. For the computation of the DFWM signal, we use the following values realized in the experiment: electronic miniband width $\Delta_e = 21$ meV, hh miniband width $\Delta_{hh} = 2$ meV, and a laser pulse width of 500 fs (full width at half maximum); the laser frequency is centered in between the S_{-1} and S_{-2} transitions. In Fig. 3, the experimentally determined DFWM curve (solid line) taken at 0.55 V is redrawn from Fig. 1(a) together with the calculated DFWM signal (dashed line). The periodic modulations of the computed signal are due to the fact that the excited state created by the laser pulses executes Bloch oscillations in k space. Using a WS ladder spacing $eFd = 3.2$ meV and a dephasing time $T_2 = 2ps$,[20] the experimentally observed modulations, with time period equal to τ_b, are nicely reproduced in the calculated curve, strongly supporting our interpretation in terms of Bloch oscillations. In addition, the calculated curve exhibits a deviation from purely exponential damping in that the calculated height of the first peak at zero time delay is reduced, in agreement with the experiment. This reduction arises from the finite width of the laser pulses leading to a diminished diffraction efficiency when the pulses temporally overlap around zero time delay.[21] Using T_2 as the only fit parameter, we can reproduce not only the overall decay of the signal but also the relative height of the first two peaks.

We have neglected Coulomb effects in the model discussed so far. It is, however, clear that the periodic modulation of the DFWM will be retained in calculations considering excitonic effects, since the exciton wave function is just a superposition of one-particle electron and hole Bloch states, which do perform Bloch oscillations. However, a quantitative investigation of Coulomb effects on the temporal evolution of the DFWM signal is needed in future.

Finally, we briefly comment on the overall decay of the DFWM signals in Fig. 1(a), which reflects the dephasing

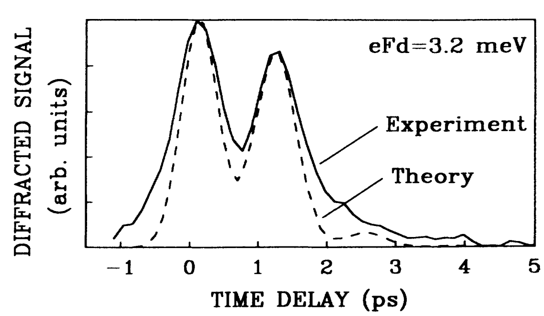

FIG. 3. Experimentally determined DFWM signal for an applied voltage of 0.55 V (solid line) together with the calculated DFWM curve (dashed line) using the experimental parameters as described in the text.

rate $\Gamma = 1/T_2$, as mentioned above. The sequence of DFWM curves in Fig. 1(a) shows that the dephasing becomes faster for larger electric fields. For an applied voltage of about 0.3 V, the observed decay is already close to our time resolution. This remains true also for even larger electric fields. This fast dephasing is due to scattering either within or out of the lowest miniband. The scattering rate between different WS states within a given miniband, however, is expected to decrease with increasing electric field, since the overlap of the initial and final scattering states decreases due to the Stark localization.[22] This is in contrast to our experimental findings. We therefore conclude that the enhanced dephasing rate is probably due to carrier escape out of the lowest miniband.

In summary, we have performed transient DFWM experiments to study optical dephasing of WS ladder excitations in a semiconductor superlattice. In the WS regime the DFWM signal exhibits a periodic modulation with a time period corresponding to the difference of the electrostatic potentials in adjacent quantum wells. We conclude that this modulation of the DFWM signal is a consequence of Bloch oscillations. Additionally, we find an increase of the dephasing rate $\Gamma = 1/T_2$ with increasing electric field. This is attributed to an efficient scattering out of the lowest miniband.

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