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Optical control of Bloch-oscillation amplitudes: From harmonic spatial motion to breathing modes

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We use a technique to measure the spatial dynamics of Bloch wave packets in semiconductor superlattices and investigate the dependence of the dynamics on the optical excitation conditions. For excitations well above or below the center of the Wannier-Stark ladder (WSL), the wave packets perform harmonic oscillations following the prediction of Zener; for excitations near the center of the WSL, the wave packets undergo a symmetric oscillation with virtually zero center-of-mass amplitude. [S0163-1829(98)51020-X]

In recent years, the dynamics of laser-generated wave packets have been investigated in detail in various physical systems. Due to the improvement of pulsed laser sources, detailed studies of the formation and the dynamics of wave packets are now possible in atoms, molecules, and semiconductors. Experiments in atoms and molecules show complex spatial and temporal dynamics due to the complexity of the underlying stationary states from which the wave packet is composed.

Semiconductor heterostructures have introduced the possibility of producing highly tailored structures. Historically, one of the first proposals of a wave-packet experiment was presented by Zener.[1] He proposed that a particle in a periodic potential subject to an electric field F will oscillate: undergoing so-called Bloch oscillations (BO). In the semiclassical picture, if an electron is ''put'' at $k=0$ and the field is switched on quasi-instantaneously, the electron moves with constant velocity in k space.[2] Due to the periodicity of the band, it starts to oscillate with a period

$$\tau_B = h/eFd. \tag{1}$$

For this transport Gedankenexperiment, an oscillating spatial motion with a total (left to right maximum) amplitude of

$$L = \Delta/eF \tag{2}$$

results. Here, d is the period of the potential and Δ the width of the band in which the electrons are moving.

Quantum mechanically, this behavior can be described as the time evolution of an electronic wave packet composed of a superposition of the eigenstates of the full Hamiltonian (including field): the so-called Wannier-Stark ladder (WSL) states. These states energetically form a ladder with a spacing given by

$$\Delta E = eFd. \tag{3}$$

Bloch oscillations are thus equivalent to quantum beats (QB) of the WSL, provided that the amplitudes of the constituent WSL states are chosen in such a manner *that the harmonic spatial motion results.*

In optical experiments in semiconductor superlattices (SL), the WSL manifests itself in the observation of a transition fan chart with energies

$$E_n = E_0 + neFd; \quad n = 0, \pm 1, \pm 2, \ldots, \tag{4}$$

when the field is swept[3,4] (E_0 is the energy of the transition between an electron and a hole in the same well).

Bloch oscillations in SL have recently been investigated by ultra-short-pulse photoexcitation of coherent superpositions of electron-hole WSL states (see, e.g., (Refs. 5–9). These time-resolved optical experiments have shown that the optical excitation of the wave packets leads to temporal oscillations with a period given approximately by Eq. (1).

We have recently introduced a method to directly trace the center-of-mass motion of the wave packets:[10] The detection principle of our experiment relies on the use of the WSL transition fan as a very sensitive field detector: the oscillating dipole field caused by the moving electrons and the static holes is superimposed on the static bias field, thus creating a small shift of the WSL transitions as a function of delay time. This shift allows us to directly obtain the displacement, with the excitation density as the only parameter.[10] The measurements provide the first direct proof of the harmonic spatial motion predicted by Bloch and Zener. When the wave packet is excited well below the center of the WSL, the total amplitude was found to be close to the theoretical prediction.[10]

Recently, a number of authors have theoretically investigated the wave-packet amplitude and its dependence on SL geometry, excitation and excitonic interactions.[11–13] The results have shown that the total spatial amplitude is strongly dependent on the properties of the wave packet. For certain excitation conditions, it was predicted that the wave packet

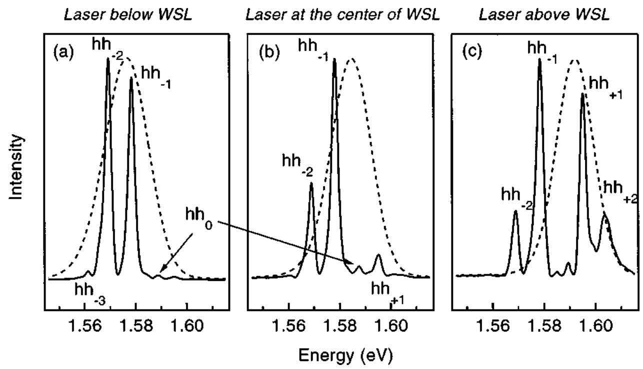

FIG. 1. Laser excitation conditions: FWM spectra (solid lines) showing the WSL heavy-hole (hh) transitions. Shown are three examples for laser excitation (dashed lines): below the center of the WSL (a), close to the center (b), and above the center (c).

would perform a symmetrical breathing-mode motion,[11,12] without any center-of-mass motion. Also, it was found that the single-particle transition energies of the WSL as given by Eq. (4) are strongly modified due to excitonic effects.[14] In combination with the excitonic modification of the WSL oscillator strengths, this modification of the energy levels has a pronounced influence on the wave-packet dynamics.[12]

In this Rapid Communication, we show that the spatial amplitude of the oscillating wave packets can be smoothly tuned between symmetric breathing-mode oscillations and the harmonic spatial motion with the amplitude as predicted by Zener. In agreement with theory, Bloch-oscillating center-of-mass motion is achieved for excitation well below and above the Wannier-Stark-ladder center whereas breathing-mode motion is achieved for excitation symmetric to the center.

The experiments are performed in GaAs/$Al_{0.3}Ga_{0.7}As$ superlattices which have been described previously.[9] We discuss here results which were taken on a SL with 67 Å well width and 17 Å barrier width. A Kronig-Penney calculation yields a miniband width of about 38 meV. The experiments are performed in a two-beam four-wave mixing (FWM) geometry using 120 fs pulses from a Ti-Sapphire laser. The first laser pulse in direction $\mathbf{k}_1$ excites the wave packet; a second pulse in direction $\mathbf{k}_2$ delayed by τ generates the FWM signal detected in the background-free direction $\mathbf{k}_3 = 2\mathbf{k}_2 - \mathbf{k}_1$. The detected signal is spectrally resolved by a CCD camera coupled to a monochromator. In all the experiments reported here, the samples are mounted in a close-cycle cryostat and held at about 10 K.

Figure 1 displays the laser excitation conditions. Shown are three examples for excitation: (a) below the center of the WSL, (b) close to the center, and (c) above the center. The solid lines are FWM spectra showing the WSL heavy-hole (hh) transitions labeled as hh_n, where n is the index as defined in Eq. (4).

Part (a) of Fig. 2 shows the spectrally integrated FWM signal as a function of delay time for various excitation conditions. For excitation near and above the WSL center, we observe double BO frequencies due to the fact that the hh_0 transition is nearly suppressed at this field (see Fig. 1). Such higher harmonics have been observed previously.[9] Part (b) of Fig. 2 shows the shift of the hh_{-1} peak vs delay time for various excitation conditions. The relative position of the

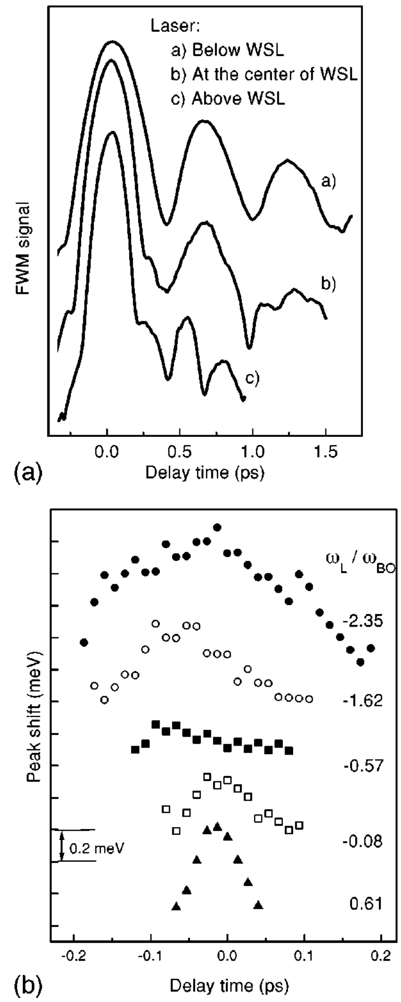

FIG. 2. (a) Spectrally integrated FWM signals for the different excitation conditions. (b) Spectral shift of the FWM hh_{-1} transition as a function of delay time.

laser is defined in units of the WSL splitting, where ω_L is the distance relative to the (experimentally observed) hh_0 transition. The time range shown is during the first oscillation when the laser excitation is still present. This has the disadvantage that the dynamics are still influenced by the further build-up of density. However, it is not possible to observe pronounced Bloch oscillations in FWM for the higher excitation density we need for observing the FWM peak shifts, at least for excitation above the center of the WSL. This is caused by the very fast loss of the interband coherence due to the large number of photo-excited free carriers.

From the field shift of the WSL peaks, it is possible to calculate the spatial displacement of the wave packet if the excitation density is known. This is performed (i) taking into account that the carrier density across the SL is not uniform due to the absorption of the exciting laser pulse, and (ii) using the experimentally determined cw field shifts of the WSL peaks.

The dots in Fig. 3 show the amplitude (as derived from the FWM peak shift) as a function of the excitation energy (i.e., the center of the laser spectrum). The energy is given in units of the WSL splitting relative to the experimentally observed energy of the hh_0 transition (ω_L/ω_{BO}). The data are taken at a field of 13 kV/cm and an excitation density (in the

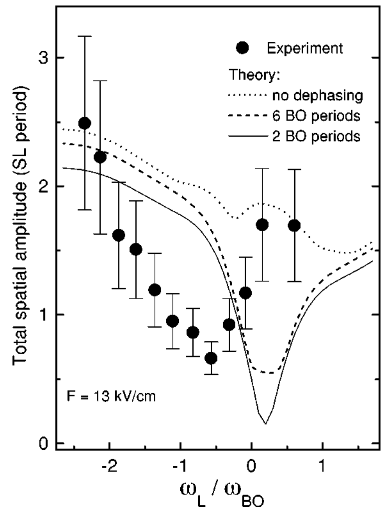

FIG. 3. Dots: Experimentally determined amplitude of the Bloch-oscillating wave packet as a function of the excitation energy at a field of 13 kV/cm. Lines: Theoretical calculation of the amplitude for various damping times, expressed in multiples of the BO time constant. The energy is given in units of the WSL splitting relative to the experimentally observed energy of the hh_0 transition.

first well) of 1.2×10^9 cm^{-2}.[15] The data clearly show that the dipole amplitude is close to zero when the excitation is near the center of the WSL. For excitation well below and above the center of the WSL, the amplitudes increase [the semiclassical amplitude as given by Eq. (2) would be about 3.5 times the SL period].

For comparison, we have performed theoretical calculations for our specific sample using a slightly modified version of the algorithm described previously.[12] This model calculates the amplitude of photoexcited wave packets taking excitonic interactions into account. We have augmented the previous model by including phenomenological dephasing of the off-diagonal elements of the resulting density matrix. The dephasing time constant was found from comparison to experiments to be approximately 1 ps. It was found, however, that the precise value was not crucial as long as it was between 2 to 6 BO periods. The comparison with theory (lines in Fig. 3) shows that rough qualitative agreement between experiment and theory is achieved: All theoretical curves show also a minimum for excitation close to the center of the WSL.

We note, however, that the inclusion of dephasing in the theory is essential for good agreement with experiment: The theoretical curve without dephasing (dotted line) predicts that the minimum amplitude for excitation close to the center of the WSL is still about 1.5 periods of the SL, whereas the experiment reaches nearly zero amplitude. With increasing dephasing, the theory agrees better with experiment: the minimum amplitude is reduced and shifted towards lower energies, as observed in the experiment.

The reason for this is as follows: Because the energy spacings of the excitonic WSL levels are not equal, there are a number of different frequencies which enter into the wave-packet dynamics. For excitation near the $n=0$ WSL level, the wave packet is initially created with a breathing-mode-type motion. However, after a time which is roughly equal to

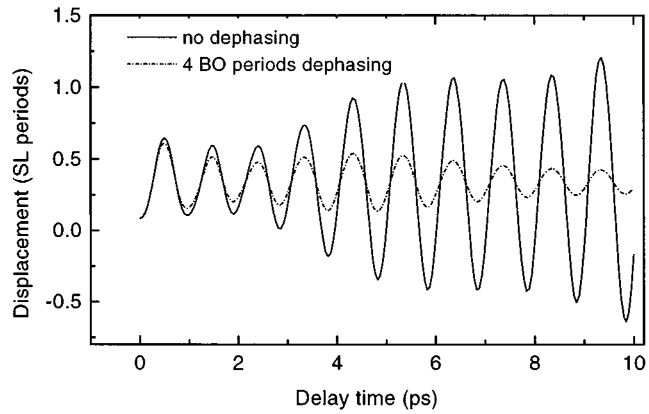

FIG. 4. Effect of the dephasing in the time evolution of displacement. Solid line: no damping; dashed line: damping time of 4 BO periods.

$\pi/\Delta\omega$, the states with energy difference $\hbar\Delta\omega$ are completely out of phase. The result is that the wave packet starts to exhibit BO with a considerable amplitude after times of this order. In the real system however, dephasing reduces the oscillation amplitude sufficiently by this time such that no large amplitude oscillation occurs. Figure 4 shows the effect of the dephasing in the time evolution of dipole for the wave packet excited with the laser central frequency at the $n=0$ WSL state. The development of the oscillating dipole when dephasing is neglected can clearly be seen.

It is interesting to note that due to the dephasing, the oscillation amplitude calculated using the full excitonic states does not differ dramatically from that predicted using the single-particle states. This is largely the result of the fact that the dipole does not oscillate long enough to display the beating effects arising from unequal level spacings. It is expected that this beating would be much more visible at lower fields, or in a superlattice with a larger period, where the difference in energy level spacing is more pronounced.

The major difference between the detailed behavior of experimental and theoretical results is that theory predicts that the minimum of the amplitude is reached for excitation *above* the center of the WSL, whereas the experiment observes minimum amplitude *below* the center. A smaller deviation in the minimum is reached when dephasing is included in the theory. The difference could be partly due to experimental error, which is relatively large (at least 30%) due to the difficulties in keeping field and excitation density constant. Possible other reasons are: (i) The theory does not take light-hole excitation into account. In the experiment, both heavy-hole and light-hole transitions are excited. (ii) In the experiment, higher minibands may play a role. They are visible by anticrossing of the transitions of the first miniband.[16] (iii) The dephasing is assumed to be the same for all off-diagonal elements of the density matrix under all excitation conditions. This is unlikely to be the case.

In conclusion, we have shown that the amplitude of photogenerated wave packets in superlattices can be controlled between true Bloch oscillations with center-of-mass dynamics and symmetric breathing modes. Further experiments and theoretical calculations are needed to obtain a full understanding of the problem and to resolve existing discrepancies between experiment and theory.

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