

## Direct Measurement of the Spatial Displacement of Bloch-Oscillating Electrons in Semiconductor Superlattices

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(Received 20 August 1996)

The absolute spatial displacement of Bloch-oscillating electrons in semiconductor superlattices is measured as a function of time with a few angstrom resolution using a novel experimental technique: The oscillating Bloch wave packet creates a small dipole field which can be determined using the field shift of the Wannier-Stark ladder transitions as a sensitive detector. The total amplitudes and their dependence on the static electric field are in good agreement with a theory including excitonic effects. [S0031-9007(97)03561-8]

PACS numbers: 78.66.Fd, 73.20.Dx

The theoretical work of Bloch [1] and Zener [2] has shown that electrons in a periodic potential subject to an electric field  $F$  will perform temporal and spatial oscillations. These oscillations are only present if the coherence of the electron will not be broken by scattering events [3]. The period and total (left to right maximum) amplitude of these so-called Bloch oscillations (BO) are given by

$$\tau_B = h/eFd, \quad (1)$$

and, respectively,

$$L = \Delta/eF, \quad (2)$$

with  $d$  as the period of the potential and  $\Delta$  as the width of the band in which the electrons are moving. A simple gedanken experiment for realizing these Bloch oscillations in a carrier transport sense is to “put” an electron at  $k = 0$ , switch on the field quasi-instantaneously, and monitor, e.g., the current oscillations.

The existence of such oscillations has been controversially debated. The situation for an experimental observation of this effect was much improved by the invention of the semiconductor superlattice (SL) by Esaki and Tsu [4]: Because of the small bandwidths in the artificial crystal made of alternating semiconductor layers with small and large band gap, the probability to perform an oscillation before scattering processes take place is much larger. A key experiment was the observation of the frequency-space equivalent of BO, the Wannier-Stark ladder (WSL), in SL by Mendez *et al.* [5] and Voisin *et al.* [6]: If a static field is present, the electron eigenstates of the superlattice form a ladder with energy spacing

$$\Delta E = eFd. \quad (3)$$

Usually, the holes can be assumed to be localized. In optical experiments, when the field is tuned, a fan chart is then observed with the transitions at the energies

$$E_n = E_0 + neFd; \quad n = 0, \pm 1, \pm 2, \dots, \quad (4)$$

where  $E_0$  is the energy of the transition between electron and hole in the same well. However, the transition energies given by Eq. (4) are strongly modified due to excitonic effects [7].

Bloch oscillations have recently been investigated with a number of techniques (see, e.g., [8–15]). Time-resolved optical experiments [8–12] have shown that the optical excitation of a wave packet consisting of WSL states with a short broadband laser pulse leads to oscillations with a period given by Eq. (1). It is not immediately obvious that these quantum beat oscillations are equivalent to the semiclassical transport picture [3] outlined above. The dynamics of optically created wave packets can be identified with the semiclassical transport picture if (i) the period of the oscillations [Eq. (1)] and (ii) if the spatial extension of the oscillations [Eq. (2)] are identical. The first requirement is always fulfilled since the spacing of the WSL as given by Eq. (3) will lead to quantum beats with the frequency given by Eq. (1). However, the *spatial amplitude* does not necessarily follow Eq. (2): Theoretical calculations [16,17] have shown that the total spatial amplitude is strongly dependent on the properties of the wave packet. For certain excitation conditions, the wave packet is not moving in one direction, but just performing a symmetrical “breathing” mode motion [16,18], without any center-of-mass motion.

The optical experiments performed up to now cannot be used to determine the spatial dynamics of the wave packet motion: Four-wave mixing (FWM) experiments [8–10] probe the temporal evolution of the excitonic interband dipole moment, which responds to both breathing mode motions and center-of-mass motions. Terahertz emission spectroscopy [11] is also strongly influenced by excitonic

correlations [19]. Furthermore, a precise determination of the amplitude would require an absolute measurement of the emitted radiation, which is not possible with sufficient accuracy. The detection by photorefractive effects [12] involves a broadband detection of the electro-optic effects of many excitonic WSL transitions and is thus also not directly linked to the oscillation amplitude.

Recently, BO have been directly observed by an inverse THz experiment [14]: The current shows resonances if the SL is illuminated with THz radiation. These experiments do not yield information about the spatial amplitudes of the BO motion. Very recently, BO have been impressively demonstrated in a lattice of atoms in a trap. Here, BO can be observed on a time scale 10 orders of magnitude longer than in semiconductors [15]. The  $k$ -space distribution of the electron wave packet was measured directly. The spatial amplitude of the oscillation was not determined.

In this Letter, we present a novel experimental method which we use to perform the *first direct measurement of the spatial dynamics of Bloch oscillations*. With a resolution of a few angstrom, we determine the spatial displacement of the Bloch oscillating wave packet as a function of time. The total amplitude is determined as a function of the static electric field. We show that optically excited BO are associated with a sinusoidal motion of the electrons and reach the amplitudes of the transport gedanken experiment.

The experiments are performed in GaAs/Al<sub>0.3</sub>Ga<sub>0.7</sub>As superlattices which have been described elsewhere [10]. In particular, 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 38 meV. The experiments are performed in a two-beam 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  $\tau$  is then either used to detect the transmission as a function of delay time or to generate the FWM signal detected in the background-free direction  $\mathbf{k}_3 = 2\mathbf{k}_2 - \mathbf{k}_1$ . In both cases, the detected signals are spectrally resolved by a charge-coupled device (CCD) camera coupled to a monochromator. In all the experiments reported here, the samples are mounted in a close-cycle cryostat and held at  $T = 8.5$  to 15 K. In most experiments, the excitation was chosen somewhat below the center of the WSL ( $E = 1.5765$  eV).

The detection principle of our experiment relies on the use of the WSL transition fan as a very sensitive field detector: The Bloch oscillations of the wave packet are associated with the oscillation of a dipole between the moving electrons and the static holes. The oscillating dipole field is superimposed to the static bias field, thus creating a small shift of the WSL transitions as a function of delay time. This shift is in a first approximation linearly proportional to the displacement of the electron wave packet.

Figure 1 shows spectrally resolved FWM signals in the region from the  $n = -1$  to the  $n = +2$  heavy-hole transitions {labeled  $hh_n$ , with  $n$  as the ladder index [Eq. (4)]}.

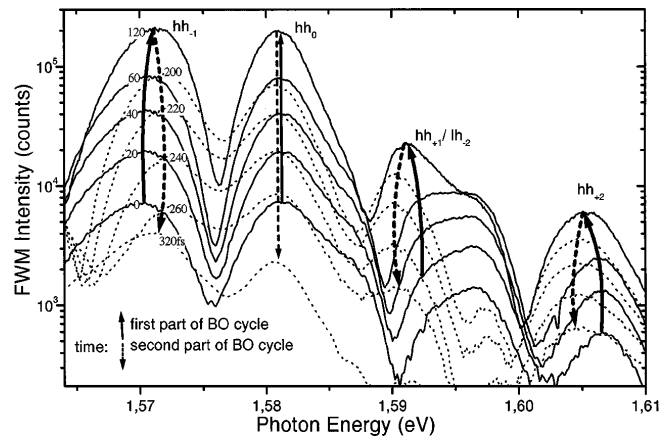


FIG. 1. Shift of different transitions of the Wannier-Stark ladder for various delay times. Solid arrows mark the behavior during the first part of a Bloch oscillation cycle, dashed arrows during the second. The relative delay times with respect to the start of the Bloch cycle (0 fs) are indicated.

The transient spectra are shown for a range of delay times which correspond to approximately one BO period. The  $hh_{-1}$  transition shows a shift to lower energies during the first half of the BO cycle (solid arrow); for the second half, it shifts first to higher energies and then back to its original position (dashed arrow). The  $hh_{+2}$  transition shows a similar oscillatory shift, *but into the opposite direction and with approximately twice the amplitude*. The  $hh_0$  transition shows nearly no shift. Obviously, the shifts of the WSL transitions allow one to *directly follow the dipole field created by the oscillating wave packet*.

The  $hh_{+1}$  transition shows a rather complicated behavior as a function of delay time: The transition splits up into two peaks, which both show a shift. A close inspection of the WSL fan chart (not displayed) shows that, in this field range, the  $hh_{+1}$  transition is subject to an anticrossing (with the light-hole transition  $lh_{-2}$ ). Thus, we observe a double peak and a nonlinear peak shift as a function of delay [20]. The results are another proof that the time-dependent peak shifts are an accurate measure of the internal field change in the sample [21].

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*. As displacement, we determine here the distance of the center of mass of the wave functions of the electrons and holes (which is identical to the distance of the charge dipole). Because of their large mass, the holes can be assumed to be fixed. The electron wave packets which are created in each well all move in parallel and create a negative charge sheet at one end of the SL. The fixed hole charges create a positive charge sheet at the other end [22]. The peak shift can be calculated by derivation of the field between these charge sheets. If the carrier density per well is  $n_{\text{well}}$ , the shift of the WSL  $\Delta E_n$  peak is given by

$$\Delta E_n = \frac{ne^2 n_{\text{well}}}{\epsilon_0 \epsilon_r} z(t), \quad (5)$$

where  $z(t)$  is the BO displacement as a function of time and  $n$  is the index of the WSL peak as defined by Eq. (4).

For a quantitative evaluation of the displacement, one has to take into account that the carrier density across the SL is not uniform due to the absorption of the exciting laser pulse. We thus use a numerical calculation of the peak shift which takes the inhomogeneous excitation into account. In this model, the photoexcited distribution of electrons and holes are first calculated. The electrons are then shifted by the Bloch oscillation amplitude (the holes are again assumed to be fixed), and the dipole field of the Bloch wave packets is calculated using the Poisson equation. Finally, the WSL peak is derived: For this purpose, we do not use the theoretical shift of the WSL peak as given by Eq. (4), but use the experimentally determined cw field shifts of the peaks. Figure 2 shows as typical results of this simulation the charge (solid line) and the field (dashed line) distribution across the intrinsic region of the SL. It is obvious that the absorption leads to different electron and hole charge sheets and to an inhomogeneous field. The absolute displacement of the electron wave packet is determined by comparing the experimentally observed field shift with the model taking the inhomogeneous field into account.

Figure 3 shows the FWM peak shift (right scale) as a function of delay time for the  $hh_{-1}$  heavy-hole transition at a field of approximately 16.5 kV/cm and an excitation density (in the first well) of  $1.7 \times 10^{10} \text{ cm}^{-2}$ . The solid line is the result of the numerical model as discussed above and includes a phenomenological damping time of 1.2 ps. By comparison of the model and the experiment, the peak shift can be directly related to the displacement shown as the left scale of Fig. 3 [23]. The electron wave packet performs a sinusoidal oscillation with a total amplitude of about 140 Å. This value is very close to the prediction including excitonic effects [16]. The semiclassical limit [as given by Eq. (2)] would be about 230 Å, i.e., the

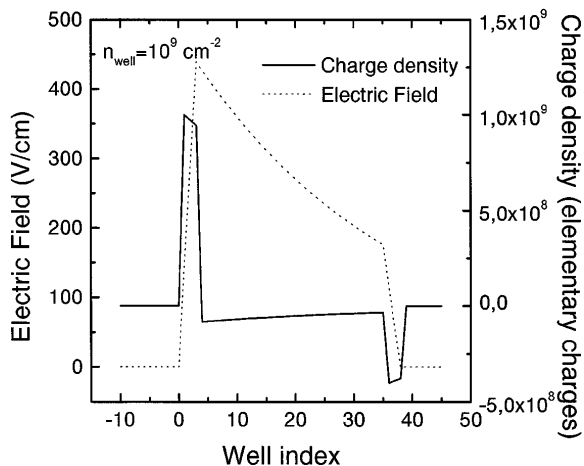


FIG. 2. Results of the simulation of the peak shift for an excitation density of  $10^9 \text{ cm}^{-2}$  and a displacement of six SL periods: Charge (solid line) and electric field (dashed line) created by the oscillating wave packet.

excitonic wave packet reaches about 2/3 of the maximum semiclassical amplitude. In principle, the peak shift can be followed down to less than a tenth of a meV, i.e., our technique is able to resolve displacements of less than 10 Å.

To verify the consistency of our results, we have checked the excitation density dependence: Part (a) of Fig. 4 shows the field shift vs excitation density. For low densities, the shift follows the expected behavior [Eq. (5)]: The peak shift should linearly increase with density (dashed line). For higher densities, we observe a saturation of the field shift. A possible reason is the screening of the internal field due to the increasing number of photoexcited free carriers, which reduce the field of the Bloch wave packet dipole by plasma screening.

As the next step, we have measured the total amplitude of the wave packet as a function of the electric field. For this purpose, we have kept excitation energy and intensity constant and have varied the electric field. The excitation was chosen somewhat below the center of the WSL ( $E = 1.5765 \text{ eV}$ ); the excitation density was  $4.3 \times 10^9 \text{ cm}^{-2}$  in the first well. The open circles in Fig. 4 are the experimentally determined oscillation amplitudes. The solid line is the semiclassical oscillation amplitude as given by Eq. (2). The experimental data agree well (within error [24]) with the semiclassical oscillation amplitude given by Eq. (2).

It has been shown that the oscillation amplitude of the wave packet is influenced by excitonic effects due to the presence of photoexcited holes [16]. The triangles in part (b) of Fig. 4 are the result of a theoretical calculation of the amplitudes including these excitonic effects for the excitation conditions chosen in the experiment. The calculation was based on the theory outlined in Ref. [16]. Within error, the theoretical predictions are in good agreement with the experiment.

Towards lower fields, we have observed that the amplitudes become very sensitive to the precise excitation conditions. This can be explained by the fact that the WSL in the low-field region is more strongly influenced by excitonic effects and shows rather complicated anticrossings

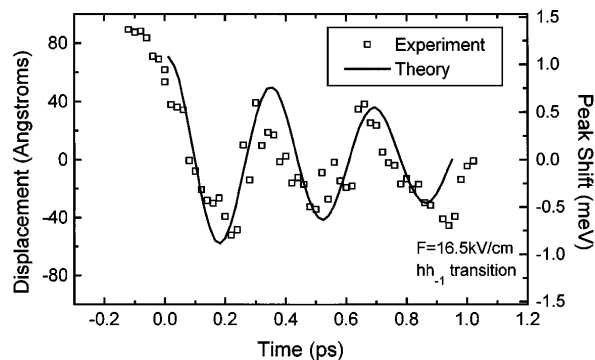


FIG. 3. Wannier-Stark ladder peak shift (right y axis) and Bloch electron displacement (left y axis) as a function of delay time for a 67 Å/17 Å superlattice. The solid line is a result of the model described in the text.

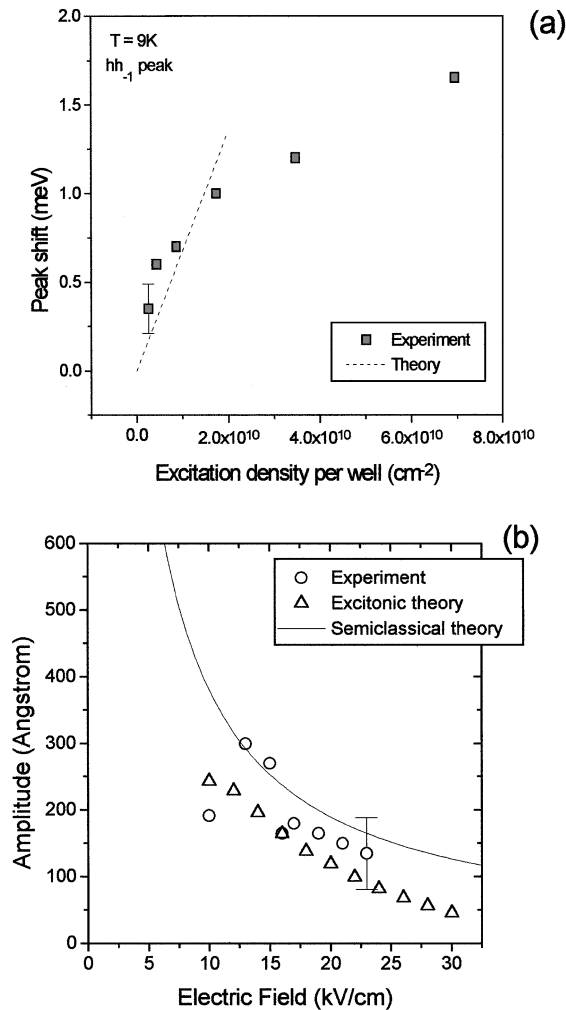


FIG. 4. (a) Peak shift (for one half of the oscillation) as a function of excitation density. The electric field is 12 kV/cm. The density displayed is the density in the first well. The dashed line is the prediction for a linear dependence assuming an amplitude of 50% of the semiclassical limit. (b) Total oscillation amplitude as a function of the electric field. Circles: experiment; line: semiclassical theory; triangles: theory including excitonic interactions.

between heavy-hole and light-hole or excitonic and free-carrier transitions, respectively. Thus, the composition of the wave packet and its spatial amplitude vary strongly with changing field and excitation conditions. We are currently performing a careful study of this field region.

We gratefully acknowledge J. Shah for helpful suggestions, C. Holfeld, D. Klatt, K.-H. Pantke, and M. Sudzius for experimental support, and fruitful discussions with J. Feldmann, H. Kurz, P. Leisching, H. Roskos, G. von Plessen, and D. Dunlap. Support from the Deutsche Forschungsgemeinschaft is gratefully acknowledged.

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- [21] Note that we determine the oscillating field shift by comparison with the cw WSL. Thus, the oscillating field (which yields the amplitudes) is determined absolutely *without errors due to excitonic effects on the Wannier-Stark ladder peak shift*.
- [22] We neglect here the edge effects which will prevent the outermost wave packets of the SL to move the full amplitude.
- [23] The field shift is decreasing during the oscillation due to the damping of the wave packet oscillation. This damping is caused by the reduction of the number of coherent electrons performing the oscillation, not by a decrease of the oscillation amplitude.
- [24] The experimental error of the amplitude determination is mainly given by the excitation density, since it is difficult to determine precisely and enters linearly into the amplitude. The absolute number of excited carriers can be determined with high precision from the photocurrent. The uncertainty in the carrier density is much larger, since (i) it is difficult to measure the spot diameter on the sample with high precision and (ii) the distribution is laterally inhomogeneous. We have imaged a pin hole on the sample and directly observed the Airy disk diameter on the sample. We estimate a total error of  $\pm 30\%$  for the absolute amplitude determination, and a relative error between data points below  $\pm 10\%$ .