PHYSICAL REVIEW B VOLUME 47, NUMBER 11 15 MARCH 1993-I

Semiconductor superlattices as terahertz generators

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(Received 11 December 1992)

Avoiding any truncation of the Hamiltonian for independent electrons in both ideal and imperfect superlattices subject to a uniform electric field, we show that dipole radiation in the terahertz range should be detectable for many members of the $GaAs/Al_xGa_{1-x}As$ system. The radition can be attributed to periodic Bloch oscillations in the case of ideal superlattices, and to almost-periodic oscillations, with the dominant frequencies on the order of the Bloch frequency, in the case of imperfect superlattices.

Over sixty years ago, work by Bloch,[1] later clarified and elaborated upon by Zener,[2] suggested that independent electrons in a periodic lattice potential subject to a uniform electric field, henceforth referred to as "Wannier-Stark electrons," will undergo time-periodic oscillations with the period $\tau_B = h/(eFa)$, where h is Planck's constant, e is the magnitude of the electron charge, F is the electric-field strength, and a is the lattice constant. Over two decades ago it was suggested by Esaki and Tsu[3] that electrons undergoing Bloch oscillations in semiconductor superlattices could serve as a source of terahertz radiation, given that a and F can be on the order of 100 Å and 1 kV/cm, respectively. However, the theoretical validity of Bloch oscillations has from the outset been a matter of great controversy.[4] Every supposed demonstration of their existence has relied on truncating the independent-electron Hamiltonian, for example, by replacing the field-free portion of the Hamiltonian by that part of its spectral representation corresponding to a single band.[5] Furthermore, until the very recent work of Feldmann *et al.*,[6] there had been no convincing experimental evidence for their existence.

In this work we avoid any truncation of the independent electron Hamiltonian to show that Bloch oscillations are a bona fide component of the dynamics of Wannier-Stark electrons in many superlattices of the $GaAs/Al_xGa_{1-x}As$ system. As discussed below, the amplitude and frequency of oscillations in both perfect and imperfect superlattices are such that dipole radiation in the terahertz range should be detectable by existing[7] experimental methods. The present work thus provides a firm theoretical foundation for the Esaki-Tsu[3] proposal.

We investigate the dynamics of conduction electrons within a periodic superlattice, which we model, in the effective-mass approximation, by a periodic one-dimensional square-well/square-barrier potential, of the form $V(z)=0$ $(|z| \le w/2)$; $V(z)=V_0$ $(w/2<z<b+w/2)$; $V(z+a)=V(z)$. Here b and w are the barrier ($Al_xGa_{1-x}As$) and well (GaAs) widths, respectively, the superlattice period is $a=b+w$, and V_0 depends on the Al concentration x. The electric field is directed along the growth direction, $\mathbf{F}=F\hat{\mathbf{z}}$. The Hamiltonian we adopt is $H=H_0+eFz$, where $H_0=T+V(z)$, the kinetic energy operator is given by $T=-(\hbar^2/2)\partial/\partial z[1/m^*(z)\partial/\partial z]$, and $m^*(z)$ is piecewise constant, with the value $0.067m_e$ in the GaAs layers and a different value in the $Al_xGa_{1-x}As$ layers which depends on the Al concentration x.

We obtain the solution of the time-dependent Schrödinger equation (TDSE) based on the complete independent-electron Hamiltonian H using high-accuracy numerical methods[8] for an arbitrary choice of initial wave function, $\psi(z,t=0)$. In some situations, the wave function executes long-lived ($\sim 10\tau_B$) Bloch oscillations featuring time-periodic center-of-mass oscillations, as in the traditional textbook picture,[9] with a repeat time τ_B. In other situations the electron wave function can exhibit diverse dynamical phenomena which can coexist with, or even mask, the Bloch oscillations. (A comprehensive discussion of all of these dynamical phenomena is provided elsewhere.[8]) In this paper we focus on the regime in which Bloch oscillations are the dominant phenomenon. This regime is characterized by the following conditions[8] on the field-free minibands, the electric field strength, and the initial wave function: The initial wave function can reasonably be represented by a linear combination of states from minibands meeting the inequality $eFa < W < E_g/2$, where W is the width of a miniband and E_g is the energy gap separating it from the next-highest miniband.

We present several of our results for electrons in a $GaAs/Al_{0.3}Ga_{0.7}As$ superlattice for the choices $w=95$ Å and $b=15$ Å. This is the same superlattice used by Feldmann *et al.*[6] in their recent observation of Bloch oscillations. For this system the potential barrier height is $V_0=243$ meV, and the effective mass has the value $0.067m_e$ in the GaAs layers and $0.092m_e$ in the $Al_{0.3}Ga_{0.7}As$ layers. This superlattice has two bound minibands: The lowest is of width 21.6 meV, the first excited miniband is of width 75.7 meV, and they are separated by a gap of 54.5 meV. The second excited miniband, separated from the first excited miniband by a gap of 72.7 meV, is unbound. We have adopted values of F varying from 2700 ($eFa=2.97$ meV) to 15 000 V/cm ($eFa=16.5$ meV). The widths of the two bound minibands as well as their corresponding energy gap are very large compared to eFa. The choice of initial wave function given below is describable by a linear combination of states of the lowest miniband. Hence, the conditions listed above for the dominance of Bloch oscillations are satisfied, and, furthermore, the interband transition rate can be expected to be very low. In fact, our results given below show that Bloch oscillations persist without appreciable decay for many multiples of the period τ_B.

We choose an initial state designed to provide a reasonable approximation to the probability distribution of electrons in experiments where a laser is tuned to excite electrons selectively from the valence band into the lowest miniband of the conduction band. We assume that the initial probability density is large in the quantum wells, small in the barriers, and that there is roughly equal probability in each of several contiguous wells. Such initial states are conveniently described by utilizing a linear combination of several suitably defined[10] Wannier functions, $|n,l\rangle$. As an example, we choose a linear combination of Wannier function associated with six contiguous wells for the lowest ($l=0$) miniband, $\psi(z,0)=(1/\sqrt{6})\sum|n,0\rangle$, where the sum extends over the contiguous wells $n=-2,\ldots,3$. (Similar results are obtained for two or more contiguous wells.[11]) The initial normalized probability density $|\psi(z,0)|^2$ is shown in Fig. 1(a) along with the total potential energy function $V(z)+eFz$, for $F=2700$ V/cm.

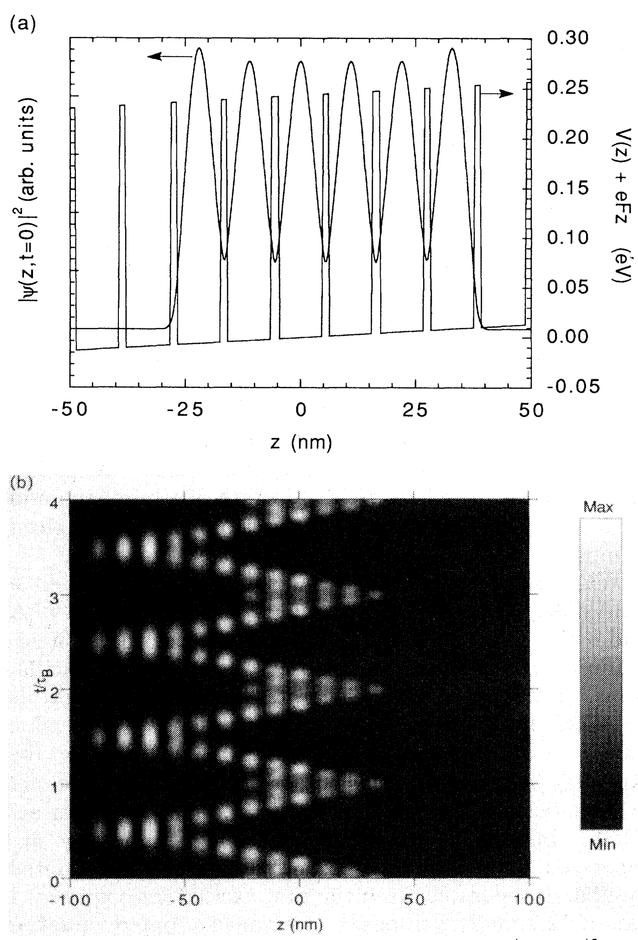

FIG. 1. (a) Initial electron probability density $|\psi(z,0)|^2$ (left ordinate), and total potential energy $V(z)+eFz$ (right ordinate), as a function of z for the periodic superlattice $GaAs/Al_{0.3}Ga_{0.7}As$, with $b=15$ Å, $w=95$ Å, and $F=2700$ V/cm. (b) Probability density as a function of z and t/τ_B for the initial state and potential energy shown in (a). The lighter the shading, the greater the probability density.

The subsequent dynamical behavior, as obtained from the numerical solution of the exact TDSE, is shown in Fig. 1(b), where the electron probability density is displayed as a function of z and t/τ_B. This is the classic Bloch oscillation picture:[9] The electron probability density moves first antiparallel to the electric field, appears to be reflected, and returns to its original form, and repeats this motion with the period τ_B. We stress that our results were obtained for the complete independent-electron Hamiltonian H free of all approximations. Thus, Bloch oscillations are a bona fide component of the exact dynamics of Wannier-Stark electrons, and not an artifact of a particular approximation method. Moreover, given the form of the wave function it is straightforward to calculate the position expectation value $\bar{z}(t)$ and from it the dipole radiation emitted by the accelerating electron. The amplitude and frequency of the Bloch oscillations shown in Fig. 1(b) are such that THz radiation should be emitted at a level detectable by existing techniques.[7]

It should be remarked that even though the above initial state is expressed in terms of Wannier states of the lowest miniband, the wave function evolves in time according to the complete independent-electron Hamiltonian H, and an electron is therefore free to make transitions to higher minibands. Nonetheless, for fields even as high as 15 kV/cm, the projection of the wave function onto the states of higher minibands is nearly zero for times up to $10\tau_B$.

The above results pertain to an idealized, periodic square-well/square-barrier superlattice potential. A more realistic model of a superlattice should allow for defects, or scattering centers, which can arise from imperfect crystal growth. In the following we describe our results for two kinds of defect configurations. In the first case, we add a small amount of Al (up to $\pm 0.5\%$) randomly throughout the superlattice, so that the potential energy function is now a roughened square-well/square-barrier function, and is no longer strictly periodic in z. We find that although the overall dynamical behavior is not strictly time periodic, the bulk of the wave packet does execute an oscillatory center-of-mass motion very near the Bloch frequency. Indeed, when we compare $\bar{z}(t)$ as a function of time for the roughened and idealized potentials, we find them to be virtually identical. This is because the random-potential fluctuations vary on a distance scale which is small compared to the width of the wave packet, so that the potential energy is in some average sense periodic in z. Thus, the electromagnetic radiation emitted by a superlattice with a small amount of random roughening should be virtually indistinguishable from that of an idealized superlattice.

A second, more interesting, defect configuration consists of introducing a single impure layer, for example, by adding uniformly a very small concentration ($<1\%$) of Al to one layer which nominally consists of GaAs. (We have also addressed multiple impure wells, but the major

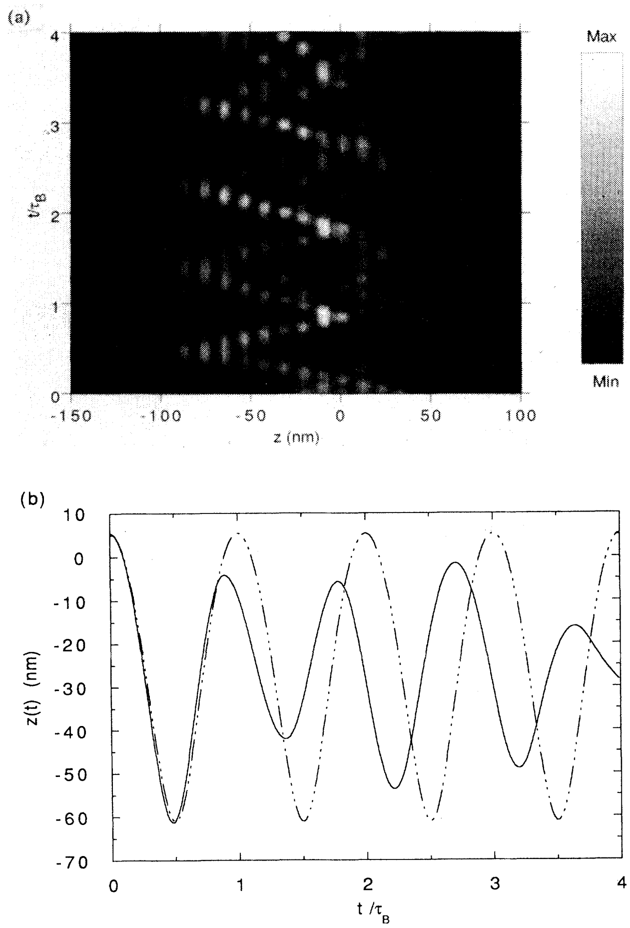

FIG. 2. (a) Probability density as a function of z and t/τ_B for the initial state shown in Fig. 1(a) and a superlattice differing from that in Fig. 1 by the addition of 0.5% Al, corresponding to $V(z)=4.05$ meV, in well $n=0$. (b) Expectation value of position $\bar{z}(t)$ as a function of t/τ_B, for the contaminated system described in (a) (solid curve), and the periodic superlattice described in Fig. 1 (dashed curve).

trends are already manifested for a single impure well.) This results in a potential energy function which is periodic everywhere except in a single well. Specifically, instead of $V=0$ as for the other GaAs wells, the lattice potential energy in the contaminated well is a few meV, depending on the concentration of Al contamination. For the same superlattice, electric field, and initial state shown in Fig. 1, but with 0.5% Al ($V=4.05$ meV) contamination in the $n=0$ GaAs well, the probability density evolves as shown in Fig. 2(a).

In contrast to the strictly periodic superlattice, for times which are integer multiples of τ_B the initial form of the wave function is not reproduced, even though there are similar qualitative features in Figs. 1(b) and 2(a). In actual fact, one can show[12] for the contaminated superlattice that the electron wave function can be described in terms of almost-periodic functions.[13] Specifically, the probability of the electron being found in any particular GaAs well can be written as an infinite Fourier series of terms of the form $\exp(i\omega_j t)$, where the frequencies ω_j ($j=0,\pm1,\pm2,\ldots$) are mutually incommensurate [ω_j/ω'_j is an irrational number for all $j,j'(\neq j)$], and their values depend on F and the concentration of Al in the defect layer. Thus, over the duration of any realistic measurement the initial form $\psi(z,0)$ is not repeated. We remark that a negligibly small fraction ($<1\%$) of the initial electron probability has made a transition to higher minibands by the time $t=4\tau_B$. Thus, the almost-periodic behavior should be attributed solely to the impurity well, and not to interband transitions.

Roskos *et al.*[7] have recently measured the dipole radiation emitted by an electron oscillating within a $GaAs/Al_xGa_{1-x}As$ asymmetric double quantum-well system, thereby essentially determining $\bar{z}(t)$. In Fig. 2(b) we show our calculated value of this quantity for both the strictly periodic superlattice as well as for the superlattice with the single contaminated well, in each case for the initial wave function shown in Fig. 1(a). We note that $\bar{z}(t)$ is periodic with period τ_B for the periodic superlattice, whereas it is expressible as an almost-periodic function for the contaminated superlattice. The amplitudes of the two curves are comparable to those measured by Roskos *et al.*,[7] indicating that even the imperfect superlattice should radiate at a detectable level, at frequencies on the order of the Bloch frequency, i.e., in the THz range. Measurement of the electromagnetic transients in the superlattice system could confirm the physical picture reported here. Alternatively, results differing significantly from those described here would indicate that the present choice of Hamiltonian, based on independent electrons, needs to be supplemented to include electron-hole, electron-phonon, or possibly electron-electron interactions. In short, such an experiment would pave the way for greater understanding of the dynamics of Wannier-Stark electrons in semiconductor superlattices.

Very recently, Feldmann *et al.*[6] have performed transient degenerate four-wave mixing (DFWM) experiments on a $GaAs/Al_{0.3}Ga_{0.7}As$ superlattice with $b=15$ Å and $w=95$ Å at the temperature 5 K. The DFWM signal exhibited modulations with period τ_B, thereby providing evidence[14] that Bloch oscillations occur in solids.[15] However, only one or two Bloch periods were observed before the signal had completely decayed. The rapid decay of the DFWM signal was attributed[6] to scattering and interband transitions. As we have described above, our results based on H for the very same superlattice show that interband transitions are entirely negligible over time intervals as lengthy as $\approx 10\tau_B$. Hence this mechanism cannot be responsible for dephasing in the DFWM experiment. However, Al contamination of a GaAs well, even at a relatively low level ($\leq 0.5\%$), excludes time-periodic behavior and gives rise to almost-periodic phenomena. We speculate that this might account for the rapid signal decay in the DFWM experiment.

More generally, there are interesting dynamical phenomena occurring in a superlattice that appear to be inaccessible to observation by the DFWM technique since the experiment is configured only to determine whether dynamical behavior is time periodic. It would appear that in the absence of time-periodic dynamical processes this technique does not provide significant information regarding the nature of other phenomena in progress. Such information could, however, be forthcoming from measurements of the electromagnetic transients.

In summary, in this work we have shown that in suitable circumstances Bloch oscillations are a bona fide component of the exact dynamics of Wannier-Stark electrons in superlattices of the $GaAs/Al_xGa_{1-x}As$ system. If one or more of the GaAs layers is contaminated, say with excess Al, the periodic Bloch oscillations are supplanted by almost-periodic oscillations. The amplitude and frequency of oscillations in both perfect and contaminated superlattices are such that radiation in the terahertz range should be detectable. The measurement of time-dependent electric dipole radiation would provide a direct probe of the dynamical behavior of the electrons. Such a technique could in principle verify the occurrence of either Bloch oscillations or almost-periodic oscillations in a direct manner and thereby greatly expand our understanding of the dynamical behavior of Wannier-Stark electrons.

We thank J. Shah for useful discussions. Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82.

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[10]For bound minibands we utilize the set of approximate Wannier functions defined by $|n,l\rangle = C_l F_l(z-na)$, where $F_l = \phi_l(z) - (J/2)[\phi_l(z-a)+\phi_l(z+a)]$, $|C_l|$ is fixed by the requirement that $|n,l\rangle$ is normalized to unity, and $\phi_l(z)$ denotes the lth normalized bound state orbital ($l=0,1,2,\ldots,k$) of the auxiliary Hamiltonian $\tilde{H} = T + \tilde{V}(z)$, where $\tilde{V}(z)=0$ ($|z|<w/2$); $\tilde{V}(z)=V_0$ ($|z|>w/2$), and J is the overlap integral for the pair of bound orbitals based on two adjacent sites. For integers $n \neq n'$ we have $\langle n'l|nl\rangle = 0$, apart from an error of order J^2.

[11]Remarkably different results are obtained if one chooses a *single* Wannier function as initial state, $\psi(z,0)=|n,0\rangle$. In the case of the periodic superlattice the position expectation value $\bar{z}(t)$ is constant, i.e., there is *no oscillatory center-of-mass motion,* in contrast to the traditional picture (Ref. 9). Thus, for this initial state, the Bloch oscillations have the character of *nonradiating time-periodic coherent breathing modes.* Thus, for this initial state, the Bloch oscillations have the character of *nonradiating time-periodic coherent breathing modes* with the repeat time τ_B and with spatial width of order $(a/2)[W(eFa)]$, where W is the energy width of the miniband. In the imperfect superlattice described below, having a single impure layer, for this same initial state, $\bar{z}(t)$ is no longer constant, but exhibits almost-periodic behavior. However, the amplitude of the oscillations is significantly smaller than for an initial state where multiple contiguous wells are occupied. A more detailed discussion of these issues is provided in Ref. 12.

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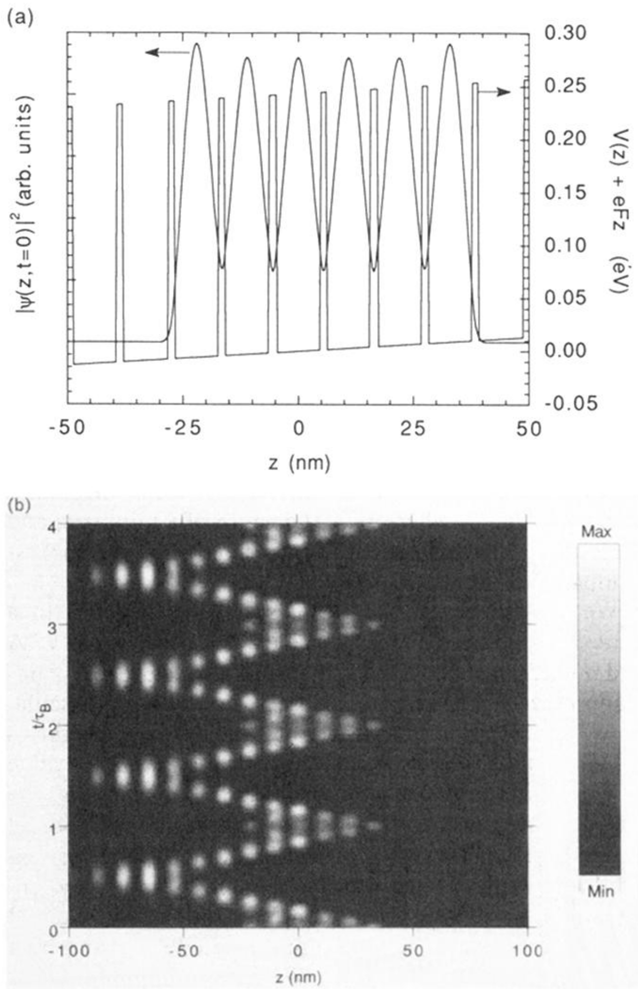
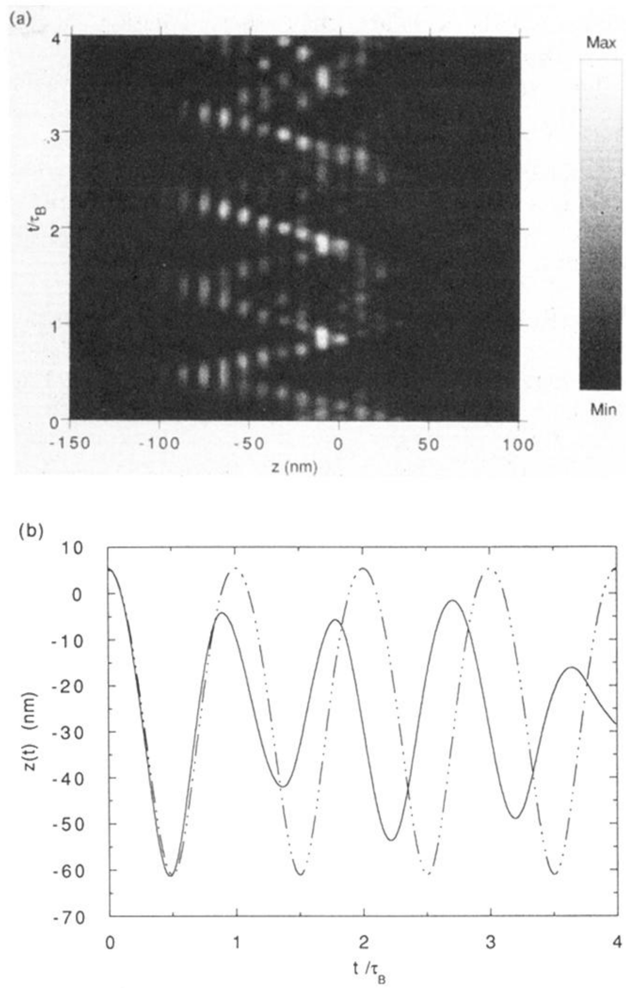


FIG. 1. (a) Initial electron probability density $|\psi(z,0)|^2$ (left ordinate), and total potential energy $V(z)+eFz$ (right ordinate), as a function of z for the periodic superlattice GaAs/$Al_{0.3}Ga_{0.7}As$, with $b=15$ Å, $w=95$ Å, and $F=2700$ V/cm. (b) Probability density as a function of z and t/τ_B for the initial state and potential energy shown in (a). The lighter the shading, the greater the probability density.



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