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Quantum Dynamical Phenomena of Independent Electrons in
Semiconductor Superlattices Subject to a Uniform Electric Field

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To Paul and Virginia Bouchard

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GENERAL INTRODUCTION

A. Organization of the Dissertation

A general introduction and literature survey are given in Sec. II of this chapter. This is followed by four related papers which will be or have been submitted to scientific journals for publication. Each paper is preceded by an abstract and followed by any associated appendices, acknowledgments, and references, according to the journal format. The last chapter of the dissertation is the General Conclusion, which summarizes the key conclusions of Papers I-IV. The General Conclusion is followed by Literature Cited, which lists the literature referenced in the General Introduction and General Conclusion, and then by Acknowledgments.

B. Introduction and Literature Review

1. Survey of the Theoretical Literature

The dynamical behavior of independent electrons in a periodic potential subject to a uniform electric field, $F = F \hat{z}$, (henceforth referred to as "Wannier-Stark electrons") is a problem with a long and rich history replete with controversy. The issues are most simply presented in the context of a one-dimensional system described by the Hamiltonian $H = T + V(z) + eFz$, where T is the kinetic energy operator, $V(z) = V(z + a)$ is the periodic lattice potential, and e is the magnitude of the electron charge.

The earliest known work on this problem was by Felix Bloch¹ in 1928. In that work, Bloch simplified the problem by imposing the requirement that the wave function be expandable at all times in terms of the Bloch eigenfunctions

$$\Phi_k(z) = e^{ikz} u_k(z), \quad u_k(z) = (z + a)$$

of a *single* band of the field-free Hamiltonian, $H_0 = T + V(z)$, in the form

$$\psi(z, t) = \int_{-\pi/a}^{\pi/a} dk \, g(k, t) \Phi_k(z). \quad (1)$$

Employing several approximations, Bloch showed that a wave packet of the form (1) peaked about a quasimomentum $\hbar k$ moves in k space with a rate which is proportional to the field strength F . In addition, by solving the single-band time-dependent Schrödinger equation (TDSE), under the restriction (1), he showed that for each value of k the function g satisfies the equation

$$\frac{\partial}{\partial t} |g|^2 = \frac{eF}{\hbar} \frac{\partial}{\partial k} |g|^2. \quad (2)$$

In 1934, Clarence Zener² pointed out that (2) may be solved to yield

$$|g|^2 = G\left(k - \frac{eF}{\hbar} t\right), \quad (3)$$

where G is an arbitrary function. Zener argued that since $|g|^2$ moves in k space with the rate $-eF/\hbar$, and since $k = -\pi/a$ is equivalent to $k = +\pi/a$, when k reaches the Brillouin zone boundary, it is Bragg reflected. Hence, $|g|^2$ is a periodic function of t , with a period $\tau = \hbar / (eFa)$. Zener concluded that the "velocity" of the electron, defined by

$$\frac{\partial}{\partial t} \int_{-\infty}^{\infty} dz \psi^* z \psi,$$

is also time-periodic with the period τ , and he describes the electronic motion as follows:

Thus if we represent the electron by a wave packet confined to the first energy band, the electron moves in the direction of the field until it is reflected by the lattice, then moves in the opposite direction until it is stopped by the field, whereupon the motion is repeated.²

Since it was Zener who in fact pointed out that the field-dependent Bloch electron behavior should thus be periodic, this phenomenon would most aptly be called "Zener oscillations." However, virtually all investigators instead refer to the phenomenon as "Bloch oscillations," and to avoid confusion, we will do likewise. In addition, we will refer to the period of the electron motion as the "Bloch period," to be denoted

$$\tau_B = h / (eFa).$$

Note that Eq. (1) is not an expression of the usual completeness condition. Since the expansion does not include *all* of the complete set of Bloch functions, but only a small subset of them, namely, those associated with a single band, the function $\psi(z,t)$ as defined in (1) cannot be an *exact* solution of the time-dependent Schrödinger equation (TDSE). As shown below, the validity of Bloch oscillations has been a matter of controversy ever since Zener first proposed them in 1934.

We remark that the phenomenon of Bloch oscillations is a purely quantum mechanical one, with no analog in the corresponding classical problem. A *classical* particle

in a periodic potential subject to a uniform electric field, if its energy were sufficiently high to surmount a barrier of the combined (lattice plus field) potential, would be accelerated in the direction of the electric force. Its motion would thus be unbounded in the direction of decreasing electric potential energy, rather than oscillatory. By contrast, a particle with insufficient energy to surmount a barrier would be trapped in a single well and oscillate with a frequency which is characteristic of the shape of the well and the strength of the electric field, rather than the Bloch frequency. We might expect that in the weak lattice limit the unbound acceleration would be manifested in the corresponding quantum mechanical problem. Similarly, in the strong lattice limit we would expect some vestiges of the intra-well oscillations to be present in a fully quantum mechanical treatment. From these considerations, it seems unlikely, if the Bloch oscillations exist at all, that they would be the only dynamical behavior exhibited by Wannier-Stark electrons.

Zener recognized that the requirement used to derive the Bloch oscillations, that the electron motion be described in terms of eigenstates of a single band only, was an incomplete treatment of the full problem. He went on to estimate the probability per unit time, γ , that the electron will make a transition to an excited band. His approach was similar in spirit to a Wentzel-Kramers-Brillouin (WKB) theory of tunneling through an energy barrier. The so-called "Zener tunneling" rate, γ , was found to be²

$$\gamma = \frac{h}{eFa} \exp \left[- \left(\frac{ma^2}{4\hbar^2} \frac{E_g^2}{eFa} \right) \right], \quad (4)$$

where m is the mass of the electron, and E_g is the energy gap separating the occupied band from the next higher band.

Several other calculations of the interband tunneling rate followed Zener's work. W. V. Houston³ in 1940 used the fact that the wave vector associated with the peak of the wave

packet in k space changes according to $k = k_0 - eFt/\hbar$ to construct accelerated Bloch-like states, termed Houston functions,

$$\psi(z, t) = u_{k_0 - eFt/\hbar}(z) \exp[i(k_0 - eFt/\hbar)] \exp\left[-\frac{i}{\hbar} \int_0^t E_{k_0 - eFt'/\hbar} dt'\right]. \quad (5)$$

From these, he computed the tunneling rate to be

$$\gamma = \frac{\hbar}{eFa} \frac{4\pi^2 e^{-\alpha}}{(1 + e^{-\alpha})^2}$$

where

$$\alpha = \frac{ma^2}{4\hbar^2} \frac{E_g^2}{eFa}.$$

Paula Feuer⁴ in 1952 extended a single-band tight-binding model to calculate the interband tunneling rate, and E. O. Kane⁵ in 1959, using Bloch functions as basis functions, computed the tunneling rate via Fermi's golden rule. Neither result agreed exactly with either Zener's² or Houston's³ results, but all four calculations supported one common trend, that the interband transition rate decreases very rapidly as $E_g/(eFa)$ is increased. In 1951, K. B. McAfee *et al.*⁶ measured the Zener breakdown current in germanium p-n junctions and showed that the measurements were in good agreement with calculations from Zener's² tunneling rate. At this point, then, one has the physical picture that for low fields, the interband transition rate should be low and Bloch oscillations should occur, whereas at higher fields the interband transition rate increases and periodic Bloch oscillations cannot occur.

However, in 1955, G. H. Wannier⁷ published some work that began an argument in the literature which has persisted until the present day. Wannier argued that if one constructs

an auxiliary equation from the TDSE by replacing z by $z + i \partial/\partial k$, one can show that interband transitions "are rigorously excluded by a selection rule." He states that "the entire change of the wave function in time is summed up by the linear increase of k with t ," and concludes that "the result of Zener^[2] is wrong and no tunneling though the forbidden band is possible. It is possible to show, however, that for very high fields the band gaps all have disappeared," which implies "a smooth acceleration from band to band, as for a free particle."

A year later Wannier⁸ wrote that there was in fact an inconsistency in his treatment, and that the Zener effect does exist. However, as shown below, he later returned to his position that Zener tunneling is rigorously excluded, and for decades much of the controversy surrounding the problem of Bloch oscillations has centered on whether the electron motion is actually physically confined to a single band, or whether this result comes about only as an artifact of inconsistent arguments or uncontrolled approximations.

In 1957, E. N. Adams⁹ claimed to show that one can define field-dependent energy bands of the Hamiltonian H , even though the field potential energy term, eFz , is not periodic. Adams concluded that for weak fields, Wannier's work⁷ was actually valid.

In a series of papers beginning in 1960, Wannier^{10,11} and collaborators,^{12,13} claimed that it is possible to modify the field-free Bloch states in such a way that there is no interband coupling. The modified Bloch functions are similar to Houston functions in that they depend on a time-dependent wave vector. They are given by

$$\psi_l(z,t) = b_l(z; k_0 - eFt/\hbar) \exp \left[-\frac{i}{eF} \int_{k_0 - eFt/\hbar}^{k_0} W_l(k) dk \right], \quad (6)$$

where b_l and W_l are the Bloch functions and dispersion relation for band index l of the auxiliary equation

$$\left[T + V(z) + eF(z + i \partial/\partial k) \right] b_l(z; k) = W_l(k) b_l(z; k). \quad (7)$$

Wannier and collaborators¹⁰⁻¹³ offered what they described as a rigorous proof that if the electron is described at time $t = 0$ by a modified Bloch function with wave vector k_0 , then at all later times it will be describable by a modified Bloch function of the same band with wave vector $k_0 - eFt/\hbar$. Thus, they argued that an electron moves in a single band, and no interband transitions occur.

Another important result obtained by Wannier and collaborators¹⁰⁻¹³ was on the related problem of the energy eigenvalues of the system. They showed that if the electric field is in the direction of a reciprocal lattice vector (in the case of a one-dimensional system, this condition is automatically satisfied), the energy eigenvalue spectrum, E_n , of the Hamiltonian is a Stark ladder with equal spacing eFa ,

$$E_n = E_0 + neFa, \quad (8)$$

where E_0 is a constant depending on the field strength and the dispersion relation $W_l(k)$.

It is important to note that, apart from the single constant term, E_0 , the so-called "Wannier-Stark ladder" (WSL) spectrum of equally spaced energy eigenvalues (8), only depends on site index n , and not on the band index l . This is because, according to Wannier,¹⁰⁻¹³ the wave function is at all times representable by states of a single band. Thus, any band other than the one initially occupied never enters into either the dynamics or the eigenvalues of the time-independent problem.

This WSL (8) was interpreted¹¹ by symmetry arguments as follows: the first term is a mean value of the energy of the band, and the second gives the shift in energy from one lattice site to the next. i.e., All lattice sites in the system are equivalent apart from a

difference in electric potential energy of eFa from one site to the next. Thus, if E_0 is an eigenvalue of the system associated with some eigenfunction $\phi_0(z)$, then $E_n = E_0 + neFa$ is also an eigenvalue associated with the eigenfunction $\phi_n(z) = \phi_0(z - na)$, where $\phi_n(z)$ is localized about the lattice site n . The set of eigenstates $\{\phi_n(z)\}$ have come to be called Wannier-Stark states.

If, as according to Wannier,¹⁰⁻¹³ the electron wave function is describable at all times by a linear combination of Wannier-Stark states of a *single* band, we can write

$$\psi(z,t) = \sum_n \phi_n(z) \exp(-iE_n t / \hbar).$$

Clearly, if the energy spectrum is given by (8), where the energy levels E_n are equally spaced, the wave function would be periodic with the Bloch period. Thus the verification of the WSL and the verification of Bloch oscillations are intimately related.

Even though Wannier^{10,11} claimed to provide rigorous proofs of these results, Wannier and Van Dyke¹³ later pointed out a feature of the theory that casts some doubt on its validity. In 1968,¹³ they showed that in most cases, the field-dependent modified Bloch bands used in their theory did not connect adiabatically to those at zero field. That is, in the limit as F approaches zero, the field-dependent Bloch bands of their theory do not merge smoothly with the bands of the field-free Hamiltonian.

Before this limitation was pointed out, however, the idea of a discrete equally-spaced WSL spectrum became fairly well accepted. For example, in 1963, J. Callaway¹⁴ calculated the optical absorption coefficient based on the theory by Kane⁵ and assuming the existence of the WSL spectrum, and predicted oscillations in absorption as a function of incident photon energy due to the WSL. Chester and Fritzsche¹⁵ in 1965 performed similar calculations of phonon-assisted optical absorption in indirect-bandgap materials in an electric field. They

also found oscillations in the absorption spectrum as a result of assuming the WSL. As shown in Sec. B, a number of experiments were performed around the same time and were interpreted as supporting the notion of the WSL.

Wannier was not without skeptics, however. In 1968, J. Zak¹⁶ pointed out that a key assumption in Wannier's^{10,11} arguments leading to the WSL is actually erroneous. Wannier's crucial assumption, according to Zak, was that the quantum number l in equation (7) is discrete. Zak rigorously derived¹⁷ an equation which is identical to (7) with the exception that the quantum number l is actually *continuous*. Zak¹⁶ concluded that the equation (7) with discrete index l is an approximation, rather than exact. He further argued¹⁶ that if one does not assume a discrete l , the quantity ϵ_0 in (8) is actually continuous in l , rather than a single well-defined constant, and therefore the WSL is replaced by a continuous distribution of energy eigenvalues.

Wannier¹⁸ promptly offered another proof of (7), rejecting Zak's critique that the equation was only approximate. Wannier did, however, concede that the Stark ladder states may not be truly discrete states but are metastable or resonance states limited by interband tunneling. Zak¹⁹ replied by pointing out that Wannier's new "proof" of (7) again involved the assumption that the discrete band index is a good quantum number. "[Wannier] assumes, therefore, what he wants to prove."¹⁹ Zak also made a point of noting that Wannier, by admitting that "'a truly discrete spectrum' for a Bloch electron in an electric field 'is unlikely,'"¹⁹ finally concurred with Zak's original result.¹⁶

Another important point was made by Zak concerning many previous calculations^{3,5,10,11,14,15} regarding Wannier-Stark electron. Each of the previous calculations employed some type of Bloch functions as a basis and either assumed or "proved" that electrons moved in a single band. The proper use of Bloch functions requires either that the system be infinite in spatial extent or that periodic boundary conditions (PBC) be used by

bending the chain into a circle to connect one end of the system to the other. Zak¹⁶ pointed out that when an electric field is applied, PBC cannot be used, because the potential energy eFz would have a discontinuity where the two ends of the chain were joined. However, an infinite system "will lead to an infinite number of levels in the Stark ladder [(8)] . . . which is meaningless because the ladder then covers the whole energy range from $-\infty$ to $+\infty$." Zak concluded that the single-band derivations based on Bloch functions^{3,5,10,11,14,15} were inconsistent.

This issue of boundary conditions was then examined closely by A. Rabinovitch²⁰ in 1970. He showed that if one applied the Born-von Karman PBC to a one-dimensional finite crystal of length L , by requiring that $\psi(z + L) = \psi(L)$, no solutions of the time-independent Schrödinger equation (TISE) exist because of the discontinuity of eFz at $z = L$. If, however, one employs the less restrictive set of PBC by requiring that $\psi(0) = \psi(L)$ and $\partial\psi(0)/\partial z = \partial\psi(L)/\partial z$, then the symmetry argument that leads to the WSL is no longer valid. Thus for either type of boundary condition, for a finite crystal, no WSL is obtained.

Shortly thereafter, Rabinovitch and Zak²¹ obtained numerical solutions of the TISE for eigenvalues and eigenfunctions for a finite Mathieu-type model crystal in an electric field, i.e., the periodic potential had the form $V(z) = 2W_1 [1 - \cos(2\pi z/a)]$ over a finite range $0 \leq z \leq L$. They found that the eigenfunctions were dramatically influenced by the choice of boundary conditions, whereas the eigenvalues were relatively insensitive. In all cases studied, the eigenvalue spectrum showed a complete absence of the WSL.

In 1972 Rabinovitch and Zak²² extended Zak's¹⁶ earlier arguments to the question of Bloch oscillations. They pointed out that the interband matrix elements neglected by Houston³ were comparable to the intraband matrix elements which were retained. They argued that if the interband coupling is also retained, then a discrete band index cannot be assigned, and the eigenvalue spectrum is continuous. They further concluded that Houston's

functions can only satisfy the TDSE for times shorter than one Bloch period and that "there is no reason to believe that the oscillations exist."²²

A number of works followed which refuted various previous arguments regarding the WSL. Avron and Zak²³ in 1974 again showed that the discrete band index associated with a periodic potential does not survive the application of an external electric field. In 1976, Avron²⁴ argued that the ladder states were actually Stark *resonances*, rather than real discrete stationary states. This finding was supported much later (in 1991) by G. Nenciu.²⁴

Others attempted to compute the spectrum of a multi-band system, but still it was necessary to use some approximations. C. A. Moyer²⁶ solved for the eigenstates of a Kronig-Penney potential in an electric field rigorously to first order in F , and found multiple Stark ladders for each band with the number of ladders increasing with decreasing field. This result brings into question the importance of terms of higher order in F . H. Fukuyama *et al.*²⁷ worked with a two-band tight-binding model, taking into account electric field matrix elements between the two bands at the same site, but neglecting matrix elements of electric field between neighboring sites. They claimed that the eigenvalue spectrum consisted of two intertwined WSLs, each WSL separately having the uniform spacing eFa .

Avron *et al.*²⁸ proved by means of a theorem²⁹ from the analysis of linear operators that the Hamiltonian H has a continuous spectrum from $-\infty$ to $+\infty$ for any nonzero value of the field. In particular, it has neither a band nor a ladder structure. If, however, one were to restrict the problem to a system of N bands, and if the interband coupling between those bands were taken into account, they argued that the spectrum would consist of N intertwining WSLs, each of which would not have equally spaced levels. In the limit as the interband coupling is reduced to zero, each WSL recovers the equal level spacing eFa .

However, not all arguments were against the existence of the single WSL spectrum and Bloch oscillations. Over the years many more arguments have been made on both sides of the issue. In some cases arguments were made on both sides by the same investigator.

Nenciu and Nenciu³⁰ in 1980 claimed to prove that if the wave function is initially in a Bloch state at $t = 0$, it remains a Bloch function for all times. Their viewpoint was softened somewhat in an article published the following year³¹ which stated that in the weak field case, the wave function remains in the same band and Bloch oscillations occur, but with stronger fields, Zener tunneling takes place. They also claimed to show that the type of effective Hamiltonian employed by Wannier [e.g., see Eq. (7)] is valid.

Churchill and Holmstrom in 1981³² made some qualitative remarks regarding the existence of Bloch oscillations. They pointed out, as we have earlier in this Section, that Bloch oscillations were inconsistent with the unbounded acceleration of an electron in a uniform electric field in the absence of a lattice potential.³³ They asserted that at least some component of the dynamics of Wannier-Stark electrons should include the unbounded acceleration seen in the lattice-free case. They then showed that if, rather than requiring *no* interband transitions, one instead required that interband transitions take place at *every* Brillouin zone boundary, then the resulting velocity did converge to the lattice-free case. They took these results to cast doubt on the validity of Bloch oscillations.

In a subsequent paper,³⁴ they constructed time-dependent Bloch states which were exact solutions of the TDSE by taking linear combinations of the solutions of the TISE with energies corresponding to the WSL. They showed that such solutions of the TDSE remain in a single band and repeat after the period of one Bloch oscillation. However, these functions did not connect adiabatically with the field-free Bloch states, in agreement with the result of Wannier and Van Dyke.¹³ Finally, they argued that the wave function is actually a standing

wave and that the time-periodic behavior does not necessarily imply the existence of center-of-mass oscillations as in the traditional picture of Bloch oscillations.

In 1985, M. Luban³⁵ solved the TDSE within the single-band tight-binding approximation utilizing localized Wannier functions as a basis, rather than Bloch states, which is an important distinction. Rather than examining the behavior of a Bloch function, time-dependent or otherwise, which is a totally extended state (and leads to inconsistencies for finite systems^{16,20}), he examined the behavior of a *localized* electronic wave packet. Luban showed that all solutions of the single-band TDSE were periodic in time with the Bloch period, providing evidently the first fully quantum mechanical derivation of the phenomenon of Bloch oscillations by a localized wave packet, the only other work on a localized packet being done by Bloch¹ himself.

Krieger and Iafrate³⁶ found a different way to sidestep the numerous criticisms¹⁶ of earlier work^{3,5,10-13} using Bloch functions. By representing the field by a time-dependent *vector* potential rather than a scalar potential, the Hamiltonian they used was invariant under a crystal lattice translation. Therefore, they could use PBC without inconsistency, since there would be no discontinuity in electric potential energy. They also avoided assuming *a priori* that the energy eigenvalues formed a WSL. For low fields or short times, they found that the electron undergoes Bloch oscillations within a single band. At higher fields or longer times, the interband transition rate was found to agree with that computed by Kane.⁵ In addition, in computing the optical absorption, oscillations associated with discrete Stark-like levels emerged as a natural result of selection rules, even without assuming or determining the eigenvalue spectrum. Thus, although no *new* results were obtained, Krieger and Iafrate claimed that the results finally were supported by a convincing derivation, since they had avoided all of the criticisms of the earlier work which were pointed out particularly by Zak.¹⁶

Zak³⁷ later wrote a comment on Krieger and Iafrate's work, saying that they had not actually removed the difficulties by utilizing this alternate representation in terms of a time-dependent vector potential, but had only transferred them from the spatial domain to the time domain. Therefore, their approach was not truly new, and was faced with essentially all of the same problems as earlier work. Krieger and Iafrate³⁸ responded by noting an error in Zak's³⁷ derivation of the problem in the time domain, and by showing that the "inconsistency" could trivially be removed by including a time-dependent phase factor if desired. They concluded by maintaining that their approach did successfully avoid the problems inherent in Houston's³ and others' methods, and that they had finally provided convincing evidence for the earlier conclusions.

Emin and Hart³⁹ in 1988 found yet another way to represent the problem. They broke up the electric field potential eFz into two components. The first is a periodic "sawtooth" potential, which distorts the shape of each well equally. The second is a "staircase" potential, which shifts the energy of each equivalent well relative to each other by a multiple of eFa . The sawtooth portion of the potential is periodic in z and is incorporated into the lattice periodic potential. The Bloch eigenfunctions of this new combined periodic Hamiltonian thus depend on the electric field by the degree to which each well is distorted (i.e. the slope $-eF$ of the potential energy in each well). The remaining staircase potential was then shown, so they claimed, to have vanishing interband matrix elements. They concluded that the energy eigenvalues were interpenetrating WSLs. In a subsequent paper,⁴⁰ they took the same approach toward the time-dependent problem, concluding that since there were no matrix elements mixing the field-dependent bands, an electron initially prepared in a field-dependent Bloch state will execute Bloch oscillations within the same field-dependent band.

A number of criticisms of Emin and Hart's work^{39,40} were made in the next few years. L. Kleinman,⁴¹ J. Zak,⁴² and Leo and MacKinnon⁴³ each pointed out a different error in Emin and Hart's^{39,40} derivation and showed that the interband matrix elements do not in fact vanish, so that their approach was equivalent to a single-band approximation. Emin and Hart responded to each comment⁴⁴ with an alternative proof of the relation questioned. The validity of Emin and Hart's^{39,40,42} work is still a matter of controversy.

One key reason why the debate has persisted has been the intractability of the problem. To date the solution of the TDSE for the complete Hamiltonian with no approximations has been inaccessible to analytical methods. Each "new" and "exact" approach is met with criticism of errors or inconsistency. A totally new approach is required, which is immune to mathematical errors, approximations, and inconsistencies. As shown in Sec. III and in the articles that follow, our numerical approach to the TDSE provides just that.

Another key reason why the theoretical debate has continued for so many decades has been the remarkable lack of experimental evidence for either the WSL or for Bloch oscillations. As shown in the next section, evidence for the WSL was not forthcoming until the 1960's, and the first experiments in support of Bloch oscillations were only reported late last year, in 1992.

2. Survey of Experiments, Proposed and Reported

The earliest successful experiments on electrons in a crystal subject to a uniform electric field were on detecting the WSL. As early as 1960, A. G. Chynoweth, *et al.*⁴⁵ observed oscillations in conductance measurements as a function of voltage and interpreted the oscillations as evidence of the WSL. S. Maekawa⁴⁶ in 1970 detected oscillations in conductivity in ZnS as a function of field strength. This was later explained by the theory of M. Saitoh⁴⁷ as due to electron-LO phonon coupling among electrons in the Stark ladder.

Saitoh showed that the oscillations provided evidence of the two-dimensional density of states in the WSL.

In 1967, V. S. Vasilov *et al.*⁴⁸ observed oscillations in absorption as a function of field strength, which was in accord with the oscillations due to the WSL, predicted by Callaway¹⁴ and Chester and Fritzsche.¹⁵ Koss and Lambert⁴⁹ performed electroabsorption experiments in 1972 which showed a "staircase" in optical absorption as a function of incident photon energy in qualitative and quantitative agreement with theoretical predictions¹⁴ assuming Kane's⁵ wave functions and a WSL energy spectrum.

In 1988, Bleuse *et al.*⁵⁰ performed tight-binding calculations of the electroabsorption spectrum in semiconductor multiple quantum wells. They predicted not only the absorption oscillations as a function of F and the staircase as a function of incident photon energy, but also a blue-shift of the optical absorption edge associated with the strong localization of eigenstates. A number of experiments followed which revealed the blue shift,⁵¹⁻⁵⁴ providing evidence for Wannier-Stark localization even at room temperature^{53,54} and demonstrating the transition from miniband extended states to localized WSL states.⁵¹

In parallel with experiments on the WSL, attempts to observe Bloch oscillations have met with less success. However, interest in this problem has intensified in recent years, with the advent of epitaxial crystal growth techniques and the development of high-mobility semiconductor superlattices (particularly of the III-V class of semiconductors). Given that a and F can be on the order of 100Å and 1 kV/cm, respectively, the Bloch period is sufficiently small (~ 1 psec) that the electron should be able to undergo several Bloch oscillations in less than estimated⁵⁵ electron scattering times.

In 1970 Esaki and Tsu⁵⁶ suggested that electrons undergoing Bloch oscillations in semiconductor superlattices would emit terahertz radiation. Such radiation could serve as a

means of detecting the Bloch oscillations, and may have applications in microwave generator devices.

Other means of detecting Bloch oscillations were proposed by R. O. Grondin *et al.*⁵⁷ in 1985. They suggested that structure could be measured in the velocity-field characteristic when incident external radiation was harmonically tuned to the Bloch frequency. Alternatively, they proposed that the velocity fluctuation noise spectra should have a peak at a field tunable Bloch frequency. However, to our knowledge neither type of experiment has been reported to date.

In 1992, von Plessen and Thomas⁵⁸ developed a theory for detecting Bloch oscillations by degenerate four-wave mixing (DFWM) experiments. They predicted that if electrons in a semiconductor superlattice were coherently undergoing Bloch oscillations, then the detected DFWM signal would have photon echo beats, or peaks, for time delays equal to integer multiples of the Bloch period. Later in 1992, J. Feldmann⁵⁹ and collaborators⁶⁰ reported just such experiments. Thus, the work of Feldmann *et al.*^{59,60} provided the first experimental evidence for Bloch oscillations.

Feldmann's^{59,60} data was not, however, periodic, as predicted by von Plessen and Thomas.⁵⁸ The signal showed definite peaks at integer multiples of the Bloch period, but also showed a rapid dephasing, or strong decay, of the detected signal. In fact, the dephasing was so rapid that only one or two oscillations were observable before the signal had completely decayed. Feldmann attributed the rapid dephasing in part to interband transitions from the lowest miniband of the superlattice, where the electrons were initially injected, to higher minibands. Later in 1992, K. Leo *et al.*⁶¹ performed similar DFWM experiments and reported as many as five Bloch oscillations, but again the detected signal evidenced strong decay. Thus, it was still unclear whether the rapid dephasing was evidence that Bloch oscillations are not strictly valid, or whether it was due to elastic and inelastic scattering

processes, etc., in the real semiconductor superlattice. In short, the experiments were not able to answer the long-standing theoretical question as to whether strictly periodic Bloch oscillations occur, or not.

In 1993, Bouchard and Luban⁶² (Paper I of this dissertation) solved the time-dependent Schrödinger equation by high-accuracy numerical methods for an electron in an idealized model of the very same superlattice used by Feldmann *et al.*^{59,60} in their DFWM experiments. Bouchard and Luban showed that robust periodic Bloch oscillations occur with negligible interband transitions for times on the order of at least *ten* Bloch periods, significantly longer than those observed in the experiments. They concluded that interband transitions could not be responsible for the rapid dephasing in the DFWM experiments. They further conjectured that scattering from static impurities in the superlattice could give rise to the type of signal decay observed in the experiments, and proposed that the detection of the electromagnetic transients,⁶³ in the terahertz range, from electrons oscillating in the sample could confirm this picture.

Later in 1993, C. Waschke *et al.*⁶⁴ reported just such an experiment. They directly detected the electromagnetic radiation emitted by coherently oscillating electrons in semiconductor superlattices and showed that the frequency of the oscillations corresponded to the energy spacing of the WSL detected by photoconductivity measurements on the same sample. The work of Waschke *et al.*⁶⁴ was thus the first direct observation of the radiation originally predicted by Esaki and Tsu⁵⁶ and confirmed by the theoretical treatment of Bouchard and Luban.⁶² However, in these radiation experiments, the oscillations decayed rapidly, as in the DFWM experiments. As we show in Paper IV, scattering from impurities may also give rise to the electromagnetic transients detected by Waschke *et al.*⁶⁴

3. A New Approach

When the present work was initiated, in 1990, the recent experiments^{59-61,64} on Bloch oscillations had not yet been published. Thus, our original goal was to undertake an entirely new theoretical investigation of the problem in an attempt to clarify whether Bloch oscillations were strictly valid or not, and to hopefully put an end to the controversy. Since the first report on the experiments by Feldmann *et al.*,^{59,60} however, we have found that our technique also has immense predictive power with regard to experiments.

By solving the TDSE by high-accuracy *numerical* methods, we have simultaneously sidestepped the numerous difficulties which have plagued previous investigators' attempts to solve the problem analytically, *and* have greatly expanded the range of problems which can be solved. Not only can we perform "numerical experiments" for *any* initial wave packet in *any* one-dimensional periodic potential subject to *any* electric field strength, but, unlike analytical methods, it is straightforward to modify the Hamiltonian to include additional interactions *without having to develop an entirely new formalism*.

For example, not only do we demonstrate that under certain conditions electrons in a periodic potential plus an electric field will execute Bloch oscillations with no noticeable deterioration for as long as we care to run the simulation (say, $50\tau_B$), but we also show its behavior in the presence of any number of static impurities. In the future, we can extend the technique to include electron-hole, electron-electron, or electron-phonon interactions, in order to develop more and more realistic models of an actual semiconductor superlattice. Thus, not only can we address the questions regarding the fundamental quantum mechanical problem of Bloch oscillations, we can continually develop and modify an increasingly realistic model of a genuine experimental system. In fact, because we view the numerical approach as having so many advantages over an analytical treatment, it is surprising to us that this approach to the problem has not been taken before.

Although we have solved the TDSE for many different periodic potentials, we focus here on results for a commonly employed model of ideal GaAs/Al_xGa_{1-x}As superlattices, because of their appeal for use in experiments. We find that Bloch oscillations most decidedly are not ubiquitous, as claimed by a number of earlier investigators.^{10-13,30,34,40} The electron wave function can exhibit a rich variety of dynamical phenomena, including long-lived Bloch oscillations, small-amplitude high-frequency intra-well oscillations, and the unbounded acceleration of a portion of the electron wave function anti-parallel to the electric field. Under certain conditions, the intra-well oscillations and/or acceleration can coexist with, or even mask, the Bloch oscillations. The precise blend of the three basic dynamical elements depends on the periodic potential, the strength of the electric field, and the detailed form of the initial electron wave function.

When the electron wave function initially occupies bands characterized by the inequality $E_g \gg eFa$, the interband transition rate²⁻⁵ is exponentially small. Thus, the electron is effectively confined to a single band. Under these conditions, we find by numerical solution of the exact time-dependent Schrödinger equation that Bloch oscillations are a *bona fide* component of the exact dynamics of Wannier-Stark electrons. The period is given by the Bloch period $\tau_B = h / (eFa)$, and the amplitude is on the order of $(a/2)[W / (eFa)]$, where W is the energy width of the occupied band.

Strictly speaking, however, the Bloch oscillations are not *exactly* periodic. There is a non-zero, but exponentially small, transition rate from the occupied energy band to the next higher band, which over long times damps out the Bloch oscillations. However, the deterioration from essentially periodic behavior can be so slow that on the time scale of any realistic measurement, the oscillations would be observed as periodic. For example, in one superlattice considered in Paper II, we find that Bloch oscillations occur for some fifty periods without any significant deterioration.

These results can also be interpreted as supporting the notion of a WSL-like eigenvalue spectrum of equally spaced, sharply peaked resonance states, whose width, or lifetime, is limited by Zener tunneling. Thus, in the case where fifty Bloch periods are observed, the spectrum must consist of very nearly discrete states, whereas when significant interband transitions occur, the resonances are broadened, corresponding to a shorter lifetime.

When more than one band is initially occupied, but when the condition $E_g \gg eFa$ is satisfied for all the occupied bands, intra-well oscillations are found to coexist with the Bloch oscillations. Specifically, Bloch oscillations associated with each occupied band are independently observable, with amplitude $\sim (a/2)[W/(eFa)]$ corresponding to each band width W . In addition, high-frequency oscillations are seen superimposed on the Bloch oscillations, with amplitude approximately one-quarter to one-half of the width of the quantum well. We are able to qualitatively explain this phenomenon by a simple model in which the electric field serves as a small perturbation coupling the different quantum states of a single well. This phenomenon can also be understood by considering the limit as the lattice strength becomes large. In that case, an electron initially in a particular well is essentially isolated from any other well, and the wave function would simply rock back and forth within that single well which is distorted by the electric field. We can also recognize this phenomenon as the quantum analog of the classical intra-well oscillations mentioned in Section II A.

In the regime in which the Zener tunneling rate is high, that is, when $E_g \lesssim eFa$, a large portion of the electron wave function is accelerated anti-parallel to the electric field. The remaining portion of the wave function undergoes Bloch oscillations which are damped out with time as more and more probability is accelerated. This acceleration is linked to the cascade of probability from one energy band to the next higher band, and so on. That is,

when we actually observe the high Zener tunneling rate, the real-space manifestation is the unbounded acceleration of a portion of the wave function anti-parallel to the electric field.

Thus with our numerical approach, we have been able, for the first time, to map out all of the quantum dynamical behavior of Wannier-Stark electrons in semiconductor superlattices in the complicated parameter space of different periodic potentials, electric fields, and initial wave functions. We have determined that long-lived Bloch oscillations are a real component of the exact dynamics of Wannier-Stark electrons, and not simply an artifact of a particular approximation scheme. This is accomplished in the first half of this dissertation.

In the latter half of this dissertation, we consider a different, but related, problem. We investigate the dynamical behavior of independent electrons in semiconductor superlattices *with static impurities* in the presence of a uniform electric field. We employ the Hamiltonian $H = T + V(z) + U(z) + eFz$, where $U(z)$ is an aperiodic term associated with the impurities. In particular we explore the effect of impurities on modifying the Bloch oscillatory behavior which would occur in a perfectly periodic, or ideal, superlattice, i.e., when $U(z) = 0$.

The main motivation for studying this second problem is to begin to develop a model which is a more realistic representation of a genuine superlattice. The periodic potentials we have investigated represent fairly simple models of ideal superlattices. Whereas the solutions of the TDSE for a periodic lattice demonstrate that as many as fifty Bloch oscillations should occur in the systems used in the experiments,^{59-61,64} the experimental results indicated at most eight oscillations before complete signal decay. We would conclude therefore, that the dephasing must be due to effects such as elastic and inelastic scattering processes in the superlattice. To make the model more realistic, one could augment the Hamiltonian, for example, with electron-electron, electron-phonon, or electron-hole⁶⁵ interactions, and with static scattering centers.

The experiments^{59-61,64} were conducted at sufficiently low temperatures (5-15K) that electron-phonon interactions are not likely to play an important role. Likewise, the density of electrons per quantum well ($\sim 10^9/\text{cm}^2$) in the experiments^{59-61,64} is such that electron-electron interactions should be minimal. The effects of electron-hole interactions⁶⁵ may play an important role, and would certainly be worthy of investigation in the future. However, the static impurities are most easily incorporated into our model, by a straightforward modification of the total potential energy function. This work represents a first, and significant, step in investigating the effect of employing a modified superlattice Hamiltonian.

Our approach is to solve the TDSE with the new independent-electron Hamiltonian, $H = T + V(z) + U(z) + eFz$, by high-accuracy numerical methods. Under conditions where Bloch oscillations would occur in ideal superlattices, i.e., when $E_g \gg eFa$, we find that in the aperiodic superlattices, the Bloch oscillations are supplanted by "almost-periodic" oscillations.⁶⁶ Specifically, the probability amplitude for the electron to be found within any particular unit cell can be written as an infinite series of terms of the form $\exp(i\omega_j t)$, where the frequencies ω_j ($j = 0, \pm 1, \pm 2, \dots$) are mutually incommensurate [$\omega_j / \omega_{j'}$ is an irrational number for all $j, j' (\neq j)$], and their values depend on F , the superlattice potential, and the impurity potential. Thus, the dynamical behavior is not characterized by a single frequency, as is the case when no impurity is present. Rather, it evidences many different "competing" frequencies, so that on the time scale of any realistic measurement, the exact form of the wave function is not repeated.

In the case of weak impurities (the meaning of the term "weak" is clarified in Paper IV), the dynamical behavior resembles Bloch oscillations for several (~ 5 -10) periods before the almost-periodic oscillations are clearly evident. We conjecture that scattering from weak impurities could give rise to the damped Bloch oscillation-like behavior observed in experiments.^{59-61,64}

In addition we show that in most cases, electrons occupying a wide miniband are less disturbed by the impurity than electrons occupying a narrow miniband. In some cases the damping due to impurities can be so slow that the behavior is virtually indistinguishable from pure Bloch oscillations for ten Bloch periods or more. We propose that an experiment based on superlattices with wide (~ 150 meV) lowest miniband could help elucidate the nature of the signal decay observed in experiments. In addition, and perhaps more importantly, such an experiment may provide an opportunity to observe Bloch-like oscillations for many more periods than have been seen before.

One disadvantage of using a numerical approach is that one has no closed-form solution to aid in qualitatively understanding the results. For this reason, we have undertaken a second complementary investigation of each of the problems discussed above. For electrons in superlattices with and without impurities, we also adopt a single-band tight-binding (SBTB) approach to the TDSE, which, although not exact, is valuable for the interpretation of the exact numerical results for cases where the electron wave function is effectively confined to a single band. For example, in the case of ideal superlattices, the SBTB model gives us insights into the different behaviors associated with different initial wave functions. In the impure superlattices, it is the SBTB results that enable us to identify the dynamical behavior as "almost-periodic,"⁶⁶ rather than random, or chaotic, as it may appear in the numerical simulations. We stress, however, that under conditions where a single-band approximation is not valid, our numerical approach allows us to explore a rich tapestry of dynamical phenomena which, regrettably, are inaccessible to investigation by analytical methods.

In Paper I we report briefly two of the most significant results of this dissertation. One result is that Bloch oscillations are a real component of the dynamics of Wannier-Stark electrons, and that the oscillations should occur for many more periods than had been seen in

experiments.⁵⁹ Secondly, we show that electrons in impure superlattices undergo almost-periodic oscillations which could give rise to the rapid dephasing in experiments.⁵⁴ The amplitude and frequency are on the same order as the Bloch oscillations, so that electronic oscillations in both ideal and impure superlattices should generate radiation in the terahertz regime. The detailed development of these results is spelled out in the remaining three papers. This paper contains essentially the same text as an article which has been published in the Rapid Communications section of Physical Review B.⁶² A few minor modifications were made so that it would blend better with the following papers.

In Paper II, we give a detailed analysis of all of the dynamical phenomena exhibited by Wannier-Stark electrons in ideal superlattices. To aid in interpreting the numerical results, analytical work is provided in Appendices. A very similar text will soon be submitted to Physical Review B.

Paper III gives an extensive development of the SBTB model for an electron in a periodic potential with a single impurity, in the presence of a uniform electric field. It is in this article that we obtain the result that the behavior is almost-periodic. This work is significantly newer than that discussed in the previous two articles. Paper III is a draft of an article which will eventually be submitted to Physical Review B or Journal of Mathematical Physics.

In Paper IV, we give an exact numerical treatment of electrons in an impure superlattice subject to an electric field. We show that under certain conditions the SBTB results of Paper III can reasonably well describe the exact dynamics of electrons in these systems. We also show that electrons in a wide superlattice miniband undergo oscillations with approximately the amplitude and frequency of Bloch oscillations for several periods before giving way to almost-periodic oscillations, whereas the almost-periodic oscillations are evident almost immediately in a wave function in a narrow miniband. The paper

concludes by proposing an experiment which may provide the opportunity to observe Bloch-like oscillations of longer lifetimes than have been observed before. Paper IV is a draft of an article which will be submitted to Physical Review B.

PAPER I

SEMICONDUCTOR SUPERLATTICES AS TERAHERTZ GENERATORS

Semiconductor superlattices as terahertz generators

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ABSTRACT

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 radiation 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.

SEMICONDUCTOR SUPERLATTICES AS TERAHERTZ GENERATORS

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 Tsu³ 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 existing⁷ experimental methods. The present work thus provides, for the first time, a firm theoretical foundation for the Esaki-Tsu³ 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 $V(z) = 0, (|z| \leq w/2); = V_0 (w/2 < z < b + w/2);$

$V(z+a) = V(z)$. Here b and w are the barrier ($\text{Al}_x\text{Ga}_{1-x}\text{As}$) 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, $\vec{F} = F\hat{z}$. 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 piece-wise constant, with the value $0.067m_e$ in the GaAs layers and a different value in the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ 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 methods⁸ for an arbitrary choice of initial wave function, $\psi(z, t=0)$. In some situations, the wave function executes long lived ($\sim 25\tau_B$) 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 article we focus on the regime in which Bloch oscillations is the dominant phenomenon. This regime is characterized by the following conditions⁸ on the initial wave function and the field-free superlattice mini-bands: (1) The initial wave function can reasonably be represented by a linear combination of states from minibands which are separated from the next higher miniband by an energy gap which is large compared to eFa ; (2) the widths of the occupied minibands are comparable to or greater than twice eFa .

We present several of our results for electrons in a $\text{GaAs}/\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ superlattice for the choices $w = 95\text{\AA}$ and $b = 15\text{\AA}$. 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 $V_0 = 243\text{ meV}$, and the effective mass has the value $0.067m_e$ in the GaAs layers and $0.092m_e$ in the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers. This superlattice has two bound minibands, i.e., minibands whose

energies lie below V_0 : 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 V/cm ($eFa = 2.97$ meV) to 15000 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.² 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 defined¹⁰ Wannier functions, $|n, l\rangle$. As an example, we choose a linear combination of Wannier functions associated with six contiguous wells for the lowest ($l = 0$) miniband, $\psi(z, 0) = (1/\sqrt{6}) \sum_n |n, l\rangle$, where the sum extends over the contiguous wells $n = -2, \dots, 3$. (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.

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

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 a periodic GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95 \text{ \AA}$, $b = 15 \text{ \AA}$, and $F = 2.7 \text{ kV/cm}$. (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.

density moves first anti-parallel to the electric field, appears to be reflected and returns to its original form, and repeats this motion with the period τ_B . The appearance in Fig. 1(b) of light regions (high probability density) separated by dark bands (low probability density) indicates that the wave function tunnels from quantum well to quantum well without ever have appreciable probability density in the barriers.

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, $z(t) \equiv \langle \psi(z,t) | z | \psi(z,t) \rangle$, and from it the dipole radiation¹² 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 exactly 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 $z(t)$ 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 trends are already manifested for a single impure well.¹³) 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 show¹³ for the contaminated superlattice that the electron wave function can be described in terms of almost-periodic functions.¹⁴ 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 ($j = 0, \pm 1, \pm 2, \dots$) are mutually incommensurate [$\omega_j/\omega_{j'}$ is an irrational number for all $j, j'(\neq j)$], and their values depend on F , the concentration of Al in the defect layer, and

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 $n = 0$. The lighter the shading, the greater the probability density. (b) Position expectation value, $z(t)$, 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).

the energy width of the occupied miniband. 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/AlGaAs asymmetric double quantum well system, thereby essentially determining $z(t)$. In Fig. 2(b) we show our calculated value of this quantity for both the ideal 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 $z(t)$ 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 $b = 15\text{\AA}$ and $w = 95\text{\AA}$ at the temperature 5K. The DFWM signal exhibited modulations with period τ_B , thereby providing evidence¹⁵ for the first time¹⁶ that Bloch oscillations occur in solids. However, only one or two Bloch periods were observed before the signal had completely decayed. The rapid decay of the DFWM signal was attributed⁶ 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 $\sim 10\tau_B$. 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 the 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/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ 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.

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expectation value, $z(t)$, 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 *non-radiating time-periodic coherent breathing modes*, with width of order $(a/2)[W/(eFa)]$ and with repeat time τ_B . (See Ref. 8 for more details.) In the imperfect superlattice described below, having a single impure layer, for this same initial state, $z(t)$ 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. 13.

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PAPER II

**BLOCH OSCILLATIONS AND OTHER DYNAMICAL PHENOMENA
OF ELECTRONS IN SEMICONDUCTOR SUPERLATTICES**

**Bloch oscillations and other dynamical phenomena
of electrons in semiconductor superlattices**

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ABSTRACT

The dynamical properties of independent electrons in a periodic potential subject to a uniform electric field, F , henceforth termed "Wannier-Stark electrons," has been a matter of great controversy for over six decades. The debate has largely centered on whether time-periodic Bloch oscillations are a real phenomenon of the exact dynamics of Wannier-Stark electrons, or merely an outcome upon adopting simplifying approximations or questionable analytical arguments. In this work, we solve the time-dependent Schrödinger equation (TDSE) for Wannier-Stark electrons in ideal GaAs/Al_xGa_{1-x}As superlattices in the conventional one-dimensional flat-band picture. The TDSE, based on the complete independent-electron Hamiltonian, i.e., avoiding any truncation, is solved using high-accuracy numerical methods. In suitable circumstances the electrons exhibit Bloch oscillations in the form of long-lived very nearly time-periodic sinusoidal center-of-mass oscillations. Depending on the miniband structure of the superlattice, the value of F , and the form of the initial wave function, other dynamical phenomena can occur which can coexist with or even totally mask the Bloch oscillations. These include a time-periodic coherent breathing mode, an unbounded acceleration of a portion of the electron wave packet anti-parallel to the electric field, and intra-well oscillations. We provide the conditions under which each of these basic dynamical elements occurs. This work thus constitutes the first systematic investigation of the dependence of the dynamics of Wannier-Stark electrons on the entire parameter space of periodic potentials, field strengths, and initial wave functions.

I. INTRODUCTION

The seemingly straightforward question, what are the dynamical properties of independent electrons in a periodic lattice potential subject to a static, uniform electric field (henceforth referred to as "Wannier-Stark electrons"), has been a matter of great controversy for decades. The issues are most simply presented in the context of one-dimensional periodic potentials, $V(z)$, (lattice constant a), with electric field $\vec{F} = F\hat{z}$ and the electron Hamiltonian given by $H = H_0 + eFz$, $H_0 = T + V(z)$, where T is the kinetic energy and e is the magnitude of the electronic charge. Work by Bloch,¹ clarified and elaborated upon by Zener,² suggested a scenario where a class of normalized electron wave functions would exhibit time-periodic oscillations, termed "Bloch oscillations," with period $\tau_B = h/(eFa)$, where h is Planck's constant. The traditional textbook approach³ is to argue that an electron initially describable by a Bloch function of wave vector k of the field-free Hamiltonian, H_0 , will at later times be describable by a Bloch function of wave vector $k(t)$, satisfying the equation

$$\hbar \frac{dk}{dt} = -eF, \quad (1a)$$

i.e.,

$$k(t) = k(0) - eFt / \hbar. \quad (1b)$$

Similarly each k component of an electron wave packet evolves according to (1). Since $k = -\pi/a$ is equivalent to $k = +\pi/a$, as $k(t)$ decreases according to Eq. (1b) and attains the value $-\pi/a$, it jumps discontinuously to the value $+\pi/a$. This is equivalent to a Bragg reflection of the electron at the Brillouin zone boundary. In real space, then, the electron is accelerated by the field, is Bragg reflected, runs in the opposite direction until it is again caught by the field and accelerated, and the motion is repeated with the period τ_B given above.

This scenario would in fact be strictly valid if it were possible to exclude interband transitions, for example by approximating the field-free portion, H_0 , of the Hamiltonian by that part of its spectral representation corresponding to a single band.⁴ The theoretical debate⁵ questions whether the phenomenon of Bloch oscillations is a feature of the *exact* dynamics of Wannier-Stark electrons, i.e., where the complete Hamiltonian H is dealt with consistently, free of all approximations, or whether it is only an artifact of a particular approximation scheme, e.g., by excluding interband transitions. A number of efforts have been made to solve the problem exactly by analytical methods,⁶⁻⁸ many of which have claimed to have shown that in fact the coupling between bands vanishes,^{6,8} so that Bloch oscillations are rigorously valid. However, each of these arguments has met with severe criticism,^{9,10} and to date the debate continues.

The closely related problem regarding the eigenvalues of the Hamiltonian, H , has also been a matter of controversy over the years. Wannier⁶ claimed that the eigenvalue spectrum is a Stark ladder of discrete equally spaced levels, with spacing eFa , which has become known as the "Wannier-Stark ladder" (WSL), $E_n = E_0 + neFa$. He further argued that if the electron wave function initially occupies only states of a single band, it remains within that band at all later times. If Wannier's claims are valid, the wave function initially occupying a single band can be written as

$$\psi(z,t) = \sum_{n=-\infty}^{\infty} c_n u_n(z) e^{-iE_n t / \hbar}, \quad (2)$$

where u_n is the eigenfunction of the time-independent Schrödinger equation associated with the eigenvalue E_n , and $c_n = \langle u_n | \psi(t=0) \rangle$. It is clear that if the WSL is valid, specifically, if the energy levels are equally spaced, the electron probability density obtained from (2) is time-periodic with the Bloch period $\tau_B = \hbar/(eFa)$. Thus, the validity of the WSL is intimately related to the validity of Bloch oscillations.

However, Zak⁹ pointed out some inconsistencies in Wannier's arguments, and thus began a long series of articles¹¹ arguing on both sides of the issue as to whether the WSL was the spectrum of the *exact* time-independent Schrödinger equation, or whether it comes about only as a result of a single-band treatment. Over the years a number of experiments¹² have provided evidence for the existence of a spectrum of equally spaced energy levels, but it is still unclear as to whether the states are truly discrete or whether they are simply energy resonances limited by Zener tunneling, as was first emphasized by Avron.¹¹

If one considers intuitively the time-dependent problem in two limited regimes, it seems clear that Bloch oscillations cannot be the only phenomenon exhibited by Wannier-Stark electrons. In the empty lattice regime, i.e., where the periodic potential, $V(z)$, is taken to be identically zero, the solution of the time-dependent Schrödinger equation (TDSE) for a localized initial wave function is a diffusing wave packet with a center-of-mass acceleration given by the classical expression¹³ $\ddot{z} = -eF/m$, where m is the mass of the electron. One must therefore expect that in cases where the lattice potential is weak, the dynamical behavior of a Wannier-Stark electron should exhibit some vestiges of this unbounded acceleration behavior, and *not* just Bloch oscillations.⁵

By contrast, in the regime where the lattice potential is very large, an electron in any particular well is essentially isolated from any other well, and the wave function should be describable as rocking back and forth within that well. One should therefore expect, for strong lattice potentials, some component of intra-well oscillation, with a frequency which is characteristic of the shape of the well (*not* the Bloch frequency).

We remark that the acceleration and intra-well oscillations are simply the quantum analogs of the behavior one would find classically. A particle with sufficient energy to surmount a local potential energy maximum (lattice plus field) would be accelerated, whereas a particle with insufficient energy would be confined to a single well and oscillate.

In short, there are arguments for a *number* of phenomena to be exhibited by Wannier-Stark electrons under different conditions. The issue of whether Bloch oscillations are valid or not has been debated most enthusiastically, but the intra-well oscillations and acceleration are intuitively reasonable as well. The question as to how exactly Wannier-Stark electrons will behave under certain conditions remains to be clarified, and that is the goal of the present work.

In this article, we provide the solution of the TDSE based on the complete Hamiltonian H for Wannier-Stark electrons given any choice of normalized initial wave function $\psi(z, t=0)$. Our approach consists of using high-accuracy numerical methods, thus enabling us to entirely avoid the numerous, often subtle, theoretical difficulties which result from invoking various analytical representations and approximations, whose impact it is usually difficult to assess. Using this approach, we are able to examine the dynamics of *any* electron wave packet in *any* one-dimensional periodic potential subject to *any* electric field strength, with no approximations.

We find that the electron wave function exhibits a rich variety of dynamical phenomena, including Bloch oscillations, time-periodic coherent breathing modes which we term "Bloch breathing modes," intra-well oscillations, and unbounded acceleration. The precise blend of these basic dynamical elements depends on the superlattice potential, the field strength, and the detailed form of the initial wave function.

Although we have applied the technique to a wide variety of periodic potentials, we focus here on results for semiconductor superlattices of the III-V class. These systems are of particular interest for the experimental detection of Bloch oscillations, because given the relatively large value of the superlattice constant a and the high electron mobility, the electron should be able to complete several Bloch oscillations in less than estimated¹⁴ electron scattering times. In addition, it has been proposed^{15,16} that electrons undergoing Bloch oscillations in superlattices, in which a and F can be on the order of 100 Å and 1

kV/cm, respectively, could serve as a source of terahertz radiation. Thus, Bloch oscillations in GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ superlattices may have potential application in novel microwave generator devices.

Recent experiments,¹⁷⁻²⁰ based on GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ superlattices, have provided the first convincing experimental evidence for the existence of Bloch oscillations. Anywhere from one to eight Bloch oscillations were observed,¹⁷⁻²⁰ but in all cases the detected signal decayed rapidly over time. This decay has been attributed^{17,18} to loss of coherence due to scattering and interband transitions. As we show in Sec. III, however, under conditions similar to those of the experiments, the wave function undergoes Bloch oscillations for as many as twenty-five periods with no noticeable decay and negligible interband transitions, i.e., less than 1% of the probability density has made a transition to higher bands after $25 \tau_B$.

The goal of this article is thus twofold. First, we demonstrate the various dynamical phenomena and determine the conditions for each to occur. In essence, we map out the dynamical behavior of Wannier-Stark electrons in the entire parameter space of periodic potentials, electric field strengths, and initial wave functions. Secondly, in light of our findings, we comment on the results of recent experiments¹⁷⁻²⁰ to detect Bloch oscillations. We conclude that the rapid decay that has been observed must be due either to effects which have not been taken into account in the independent-electron Hamiltonian we have employed, or to imperfections in the laboratory superlattices, i.e., deviations from the ideal periodic square-wave picture.

In the following Section, the various ingredients of our treatment are presented. Results and discussion are given in Sec. III. Some of our results are interpreted with the aid of analytical calculations, which are found in the Appendices. Specifically, in Appendix A, we present results of a single-band tight-binding model of the TDSE to elucidate some features of the Bloch oscillations and Bloch breathing modes. In Appendix B, we develop a simple time-dependent perturbation theory of a single quantum well so as to give a

quantitative basis for to interpreting the intra-well oscillations. A summary and conclusions are given in Sec. IV.

II. THEORY

A. Numerical Method

The time-dependent Schrödinger equation (TDSE),

$$i \hbar \frac{\partial \psi}{\partial t} = H \psi, \quad (3)$$

has an elegant formal solution:

$$\Psi(t + \Delta t) = U(\Delta t) \Psi(t). \quad (4a)$$

where

$$U(\Delta t) = \exp(-iH\Delta t/\hbar) \quad (4b)$$

if the Hamiltonian, H is time-independent. However, the evaluation of (4b) is problematical, in that one cannot retain all of the infinite number of terms in the series expansion of the exponential, but truncating the series results in an approximate evolution operator which is not strictly unitary, so that norm conservation of the wave function is violated.

Therefore, to solve the TDSE, we utilize a modified Cayley method,^{21,22} in which the strictly unitary operator,

$$U_c(\Delta t) = \left(1 + \frac{i\Delta t}{2\hbar} H\right)^{-1} \left(1 - \frac{i\Delta t}{2\hbar} H\right), \quad (5)$$

is used as an approximation to the exact evolution operator (4b), with an error which is only of order $(\Delta t)^3$. Thus by choosing Δt sufficiently small, the error introduced by using the Cayley operator (5) in place of the exact evolution operator (4b) is exceedingly small and grows extremely slowly over time.

To monitor the dynamical evolution of the system we obtain a sequence of snapshots of the wave function, at uniformly spaced times, $t_k = k \Delta t$, ($k = 0, 1, 2, \dots$). The system is described at the time t_k by a column vector, $\Psi(t_k)$, whose elements are the instantaneous values of the electron wave function for the uniformly spaced set of spatial mesh points

$z_j = z_0 + j \Delta z$, ($j = 0, \dots, N$), where Δz is the mesh spacing. The choices $\Delta z = a/200$ and $\Delta t = \tau_B/2000$ generally ensure accurate results for time intervals up to $50\tau_B$. We employ "hard wall" boundary conditions by requiring that the wave function at the fictitious $j = -1$ and $j = N + 1$ mesh points be equal to zero at all times. We then choose N sufficiently large that the wave function never comes close to the boundaries for the duration of the simulation. Depending upon the value of F and the duration of the run, N was chosen in the range 5×10^3 to 10^5 .

A potential weakness of any discretization procedure is that there is no *a priori* method for deciding how small Δz and Δt need be in order to obtain accurate results. In addition to decreasing the mesh spacing and time step until a convergent solution is reached, we have also developed a procedure to check the accuracy of our numerical solution of the TDSE by testing to what extent the Ehrenfest theorem is satisfied. Ehrenfest's theorem consists of two equations,

$$\frac{d}{dt} z(t) = \frac{p(t)}{m} \quad (6a)$$

$$\frac{d}{dt} p(t) = \mathcal{F}(t) \quad (6b)$$

whose derivation implicitly assumes the TDSE to be satisfied. Here, for any operator O , the expectation value $O(t)$ is given by $O(t) \equiv \langle \Psi(z, t) | O | \Psi(z, t) \rangle$. Thus, for example $p(t)$ is the expectation value of the momentum operator, $-i\hbar \partial/\partial z$, and $\mathcal{F}(t)$ is the expectation value of the "force operator," $-dV/dz - eF$. By combining (6a,b), we have

$$\frac{d}{dt} z(t) = \frac{1}{m} \left[p(0) + \int_0^t dt' \mathcal{F}(t') \right]. \quad (7)$$

The procedure by which we test the accuracy of the Cayley method consists of computing each of the left and right hand sides of (7), using the numerical solution $\Psi(z, t)$, and comparing them at each time step. If the two quantities do not agree, Ψ cannot be a reliable solution of the TDSE. This situation signals, for example, that the choice for Δz and/or Δt is

too large, or that the wave function has encountered the artificial boundaries imposed on the system. If, however, the left- and right-hand sides of (7) do agree (typically we consider them to "agree" when the relative difference at any time step is no more than a percent or so), then Ψ is a solution of the TDSE to high accuracy.

We checked the validity of this accuracy assessment method by comparing the numerical and exact solutions of the TDSE in two cases where the exact solution of the TDSE is known, the first, a free electron wave packet²³ of arbitrary form, and the second, an electron wave packet of arbitrary form in a uniform electric field.¹³ We found that when the numerical solution of the TDSE departed from the exact solution, the left- and right-hand sides of (7) also departed from each other, whereas when the numerical results for the wave function were in close agreement with the exact results, (7) was satisfied to high accuracy.

The appeal of this method lies in the fact that it uses a *physical* criterion, Ehrenfest's theorem, to assess the *physical* validity of the solution. It is possible to obtain a numerical solution to a mathematical equation which converges when the mesh spacing is fine-tuned, but yet the solution is unphysical. Verifying that Ehrenfest's theorem is satisfied ensures that the solution obtained not only is numerically accurate, but also is a *physical* solution. This gives us more confidence in the validity of our results than can be obtained solely from fine-tuning the mesh spacing and time step.

B. Superlattice Model

We focus in this article on electrons in a GaAs/ $\text{Al}_x\text{Ga}_{1-x}\text{As}$ superlattice which we model in the effective mass approximation by a one-dimensional square-well/square-barrier potential of the form $V(z) = 0, (|z| \leq w/2); = V_0, (w/2 < z < b + w/2); V(z+a) = V(z)$, where V_0 depends on Al concentration x . Here b and w are the barrier ($\text{Al}_x\text{Ga}_{1-x}\text{As}$) and well (GaAs) widths, respectively, and the superlattice period is $a = b + w$. The effective mass is piecewise constant with a value depending on the Al concentration x in each layer. We

remark that in order to incorporate a spatially varying effective mass, $m^*(z)$, we utilize the kinetic energy operator of the form²⁴ $T = -(\hbar^2/2) \partial/\partial z \{ [1/m^*(z)] \partial/\partial z \}$.

As shown below, the superlattice band structure strongly influences which of the three basic dynamical elements, i.e., Bloch oscillations, intra-well oscillations, or acceleration, will dominate the dynamical behavior. By adjusting the Al concentration, x , the barrier width, b , and the well width, w , one can tailor, to a large extent, the band structure of the superlattice in order to design a system to display a particular combination of dynamical properties.

It should be stressed, however, that the particular use of a square-well/square-barrier potential function is non-essential to our conclusions regarding Wannier-Stark electrons in general. We have investigated other periodic potentials, including double-barrier/double-well superlattices and cosine potentials, and the basic conclusions are the same. What is significant in determining the electron dynamics is the band structure generated by the periodic potential, rather than the specific form of the potential itself.

C. Initial Wave Functions

We find that the electron dynamics depends strongly on the form of the initial wave function and its relationship to the states of the field-free minibands of the superlattice. One feature of the initial wave function which is of key importance is the number of contiguous superlattice wells with significant initial probability density. As shown below, dramatically different results are obtained if only one well is initially occupied than if two or more contiguous wells are occupied.

The second crucial property is the projection of the initial wave function onto states of the superlattice minibands and the properties of the minibands which are occupied. The behavior of the wave function thus depends on whether only a single miniband is occupied,

or more than one miniband. It also depends on the energy width of each occupied miniband as well as the band gap separating it from the next higher miniband.

It is important to remark that the *specific* shape of the initial wave function, e.g., whether it is a Gaussian, or a Lorentzian, or any other normalized function, is *not so important*. As remarked above, what *is* important is how many contiguous wells are occupied, and the properties of the minibands which are occupied. We demonstrate this claim below, by showing that significantly different initial wave functions can exhibit very similar time evolution if only they occupy the same minibands and approximately the same number of wells.

We have employed a variety of initial states in our investigations in order to explore the effect of different spatial configurations and energy components of the initial state on the electron dynamics. We frequently utilize a normalized traveling Gaussian initial wave function of the form

$$\psi(z, 0) = C \exp[iq_0 z - (z - z_0)^2 / (2\sigma^2)], \quad (8)$$

where C is the normalization constant.

In other instances we construct an initial state from a finite linear combination of Wannier functions, $\{|n, l\rangle\}$, defined by

$$|n, l\rangle = \phi_l(z - na) = \frac{a}{2\pi} \int_{-\pi/a}^{\pi/a} dq e^{-inqa} \Phi_{l,q}(z), \quad (9)$$

where $\Phi_{l,q}(z)$ is the Bloch eigenfunction of the field-free Hamiltonian associated with the band l and wave vector q and normalized according to

$$\int_0^a dz |\Phi_{l,q}(z)|^2 = 1. \quad (10)$$

The Wannier functions so defined are orthonormal, i.e., $\langle n, l | n', l' \rangle = \delta_{n,n'} \delta_{l,l'}$, and if one considers all integers n and l , constitute a complete set. Thus, they may be used to construct any arbitrary initial state.

The state $|n, l\rangle$ may be identified with a particular band l and is well localized about the quantum well centered about $z = na$, or "site n ." This set of functions is particularly convenient for constructing an initial state, because we may utilize a particular linear combination of Wannier functions to explicitly control which sites and minibands are occupied. As we have said previously, these are the properties of the initial state which are of importance to the electron dynamics.

To compute the Wannier functions (9), we numerically solve the time-independent Schrödinger equation for the given superlattice in the absence of the electric field subject to the Bloch boundary condition, $\Phi_{l,k}(z+a) = e^{ika} \Phi_{l,k}(z)$, for a specific value of wave vector q in the range $-\pi/a \leq k \leq \pi/a$. The Bloch eigenfunctions $\Phi_{l,k}(z)$ are normalized according to (10). This process is repeated for those values of k which are required for evaluating the integral in (9) by either Gaussian or Simpson integration.²⁵ We find that in most cases Gaussian integration converges efficiently. However, in cases where $\partial \Phi_{l,k}(z)/\partial k$ changes rapidly as a function of k , Simpson integration, with a fairly fine mesh of k -points (~ 100 points in the interval $0 \leq k \leq \pi/a$), usually proves more successful. The resulting Wannier functions are then checked for orthonormality. We find that the magnitude of the overlap integral between Wannier functions, $\langle n, l | n', l' \rangle$, for $n \neq n'$ and/or $l \neq l'$ is no greater than 10^{-6} and frequently is on the order of 10^{-15} or 10^{-12} , while for $n = n'$, $l = l'$ the overlap integral differed from unity by approximately the same amount.

Fig. 1 shows the Wannier function, $\phi_l(z-na)$, at site $n = 0$ for the lowest three minibands of two different superlattices. The first, superlattice A, is a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95$ Å and $b = 25$ Å. The second, superlattice B, is a GaAs/Al_{0.2}Ga_{0.8}As superlattice with $w = 200$ Å and $b = 5$ Å. (The Wannier function $\phi_l(z-na)$ associated with an arbitrary site n is obtained from $\phi_l(z)$ by a spatial translation through a distance na .)

Fig. 1 Wannier functions, $\phi_l(z)$, of band index l and site index 0, as a function of z for (a)-(c) superlattice A: a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95\text{\AA}$ and $b = 25\text{\AA}$, and (d)-(f) superlattice B: a GaAs/Al_{0.2}Ga_{0.8}As superlattice with $w = 200\text{\AA}$ and $b = 5\text{\AA}$.

Note that the Wannier functions for a given superlattice become less localized and have more oscillations as the band index is increased. Also note that the Wannier functions associated with the superlattice A are much more localized than those of the superlattice B, due to the relative size of the quantum wells and barriers. These observations will be valuable for understanding the differences between some of the initial wave functions employed in Sec. III.

III. RESULTS AND DISCUSSION

A. Overview of Results

In this section, we provide a brief overview of the dynamical phenomena displayed by Wannier-Stark electrons and state the conditions under which each occurs. In subsequent sections, we examine each phenomenon in detail, and construct, piece by piece, the overall picture summarized here.

We find that the dynamical behavior of Wannier-Stark electrons is intimately related to the rate of interband transitions from initially occupied minibands to higher minibands. The probability, γ , per Bloch period that an electron will make a transition from one band to the next higher band was estimated first by Zener² to be

$$\gamma = \exp\left(-\frac{ma^2}{4\hbar^2} \frac{E_g^2}{eFa}\right), \quad (11)$$

where E_g is the energy gap above the occupied band. Other calculations²⁶ of the so-called "Zener tunneling" rate, although not agreeing exactly with (9), give the same trend, that the interband transition rate rapidly decreases as the ratio $E_g/(eFa)$ is increased.

If for any occupied miniband $E_g \gg eFa$, so that the Zener tunneling rate is low, we find that long-lived Bloch oscillations do occur with lifetime $\geq 10\tau_B$. The form of the Bloch oscillations depends on the number of contiguous wells that have significant initial probability. If two or more contiguous wells are occupied, then the wave packet exhibits a sinusoidal center-of-mass motion similar to the traditional textbook²⁷ picture. The amplitude of the oscillations is $\sim (a/2)[W/(eFa)]$, where W is the energy width of the occupied miniband. Thus, if W is at least twice eFa , the amplitude of the oscillations will be on the order of the superlattice constant a .

If, however, only a single well is occupied, or if each occupied well is separated from other occupied wells by at least one empty lattice site, then a very different, special class of Bloch oscillations occurs. We will distinguish it from the wider class of Bloch oscillations by referring to it as a "Bloch breathing mode," as it has the form of a time-periodic coherent breathing mode with the period τ_B , i.e., it has no center-of-mass motion. The width of the wave packet oscillates in time with an amplitude $\sim (a/2)[W/(eFa)]$

If more than one band is occupied initially, each satisfying the condition $E_g \gg eFa$, then in addition to the Bloch oscillation (or Bloch breathing mode) associated with each occupied band, we find that high-frequency, small-amplitude intra-well oscillations also occur. The frequency of the intra-well oscillations can be identified with the difference between the mean energy of the occupied minibands, and the amplitude depends on the field strength, but is typically on the order of half the well width. Thus, if conditions are such that $eFa \gg W$, but still $E_g \gg eFa$, then the Bloch oscillations are suppressed [recall that the Bloch oscillation amplitude depends on $W/(eFa)$], and the intra-well oscillations can actually be the dominant phenomenon.

As the field strength is increased, or the band gap is decreased, we find that the Zener tunneling rate increases. When eFa is comparable to or greater than E_g , the probability density cascades from the initially occupied miniband to the next higher miniband, and so on. The real-space manifestation of the Zener tunneling is a portion of the electron wave function undergoing unbounded acceleration anti-parallel to the electric field. A small portion of the wave packet may also exhibit Bloch oscillations and/or intra-well oscillations which are damped over time as more and more of the probability density is accelerated.

Thus, by modifying the superlattice, the field strength, and the initial wave function, we can design a system which will demonstrate one particular dynamical phenomenon or a specific mixture of phenomena. We now do precisely that, in order to demonstrate each behavior and examine it in detail.

B. Bloch Oscillation Regime

We first present several of our results for electrons in a GaAs/Al_{0.3}Ga_{0.7}As superlattice for the choices $w = 95 \text{ \AA}$ and $b = 25 \text{ \AA}$ ($a = 120 \text{ \AA}$), henceforth referred to as "superlattice A." For this system the potential barrier height is $V_0 = 243 \text{ meV}$, 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. The energy minimum and maximum of each of the four lowest minibands of this superlattice are given in Table I, along with the corresponding widths and gaps. The Wannier functions associated with the three lowest minibands of this superlattice are shown in Fig. 1(a)-(c) (superlattice A). We choose $F = 2.5 \text{ kV/cm}$, so that $eFa = 3 \text{ meV}$, and $\tau_B = 1.38 \text{ psec}$.

TABLE I Minimum and maximum energy values, E_{min} and E_{max} , respectively, for the miniband of band index l of superlattice A: a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95 \text{ \AA}$ and $b = 25 \text{ \AA}$. Also included are the miniband widths, W , and the energy gap, E_g , between minibands. All energy values are given in meV.

l	E_{min}	E_{max}	W	E_g
0	27	38	11	72
1	110	153	43	93
2	246	346	100	85
3	431	619	188	

Note that the lowest miniband is well separated from the next higher miniband (band gap $E_g = 72 \text{ meV}$), so that eFa (3 meV) is very small compared to E_g . The Zener tunneling rate is thus very low,^{2,26} on the order of 10^{-4} per τ_B . As stated in Sec. I, this is the regime where a single-band treatment, in which Bloch oscillations have been shown^{2,4} to be valid, is likely to provide a good approximation to the exact problem. Therefore, those initial wave

functions which are reasonably well represented by a linear combination of Wannier states of the lowest miniband are prime candidates for exhibiting Bloch oscillations.

The first results we will show are for just such an initial state. Specifically, we consider a linear combination of several, for example, six, Wannier functions of the $l = 0$ miniband and centered about contiguous sites, $\psi(z, 0) = (1/\sqrt{6}) \sum_n |n, 0\rangle$, where the sum extends over $n = -2, \dots, 3$. (The equality of the coefficients employed here is non-essential. Other linear combinations of contiguous Wannier functions give qualitatively similar results.) The initial normalized probability density $|\psi(z, 0)|^2$ is shown in Fig. 2(a) along with the total potential energy function $V(z) + eFz$.

The rationale for choosing such an initial state is as follows. In experiments,¹⁷⁻²⁰ where electrons are selectively injected into the lowest miniband of the superlattice by means of a tuned laser excitation from the valence band, the initial electron probability density is likely to be large in the quantum wells, and small in the barriers. Additionally, we would expect that each of several contiguous wells would have roughly the same probability of being occupied. The choice of initial wave function shown in Fig. 2(a) thus may provide a reasonable representation of the probability distribution of electrons generated in experiments.

The evolution of the wave function, as obtained from the numerical solution of the exact TDSE, is shown in Fig. 2(b), where the electron probability density is displayed as a function of z and t/τ_B . This is very similar to the traditional Bloch oscillation picture.²⁷ The electron probability density moves first anti-parallel to the electric field (i.e., in the direction of the classical force due to the field), appears to be reflected and returns to its original form, repeating this motion with the period τ_B . The appearance in Fig. 2(b) of light regions (high probability) separated by dark bands (low probability) indicates that the wave packet tunnels from quantum well to quantum well without ever having appreciable probability in the barriers.

Fig. 2 (a) Total potential energy, $V(z) + eFz$ (right ordinate) as a function of z for a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95$ Å, $b = 25$ Å, and $F = 2.5$ kV/cm. Initial probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to a linear combination of Wannier functions of the lowest field-free miniband. (b) Probability density as a function of z and t/τ_B for the initial state and total potential energy shown in (a).

Fig. 3 Position expectation value, $z(t)$, as a function of t/τ_B , for the same initial state and potential energy shown in 2(a).

Fig. 3 shows the position expectation value,

$$z(t) \equiv \int_{-\infty}^{\infty} dz \, z |\psi(z, t)|^2,$$

as a function of time, on a longer time scale than shown in Fig. 2(b). As indicated in Fig. 3, the Bloch oscillations are virtually time-periodic and very long-lived with negligible deterioration.

We stress that these results were obtained for the complete Hamiltonian, H , free of all approximations. Thus, long-lived Bloch oscillations are a genuine component of the dynamics of Wannier-Stark electrons at least in the idealized superlattices under consideration. As shown below, a wide class of initial wave functions in this superlattice will

produce Bloch oscillations of a similar character as those seen in Figs. 2-3. Furthermore, the amplitude and frequency of these oscillations are such that dipole radiation in the terahertz regime should be detectable^{15,16} by existing experimental techniques.²⁸

Very recently, Waschke *et al.*²⁰ reported the observation of coherent terahertz radiation emitted from electrons undergoing Bloch oscillations in a GaAs/Al_xGa_{1-x}As superlattice with similar properties to superlattice A considered above. However, at most only eight oscillations were observed which decayed rapidly over time, in contrast to the much longer-lived periodic behavior seen in our solutions of the TDSE for an ideal superlattice. As discussed in Sec. IV, we conclude that the decay observed in the experiments,²⁰ must be due to features of the real superlattice system which are not included in the idealized problem under consideration here.

We now examine the dynamical behavior of the initial state shown in Fig. 4(a), a broad Gaussian function, of the form of Eq. (8), with no initial momentum, i.e., $q_0 = 0$, and with width parameter $\sigma = 200 \text{ \AA}$, so that three contiguous quantum wells have appreciable initial probability density. The sum of the projections of this initial wave function onto Wannier states of the lowest miniband turns out to be equal to 0.91. Thus, this initial state can also be reasonably well represented by a linear combination of Wannier states from the lowest band.

The probability density as a function of z and t/τ_B is shown in Fig 4(b). Although Figs. 2(b) and 4(b) differ in their details, the oscillating center-of-mass motion and amplitude are nearly the same. We also find qualitatively similar results from other linear combinations of contiguous Wannier functions of the lowest miniband. Thus, we can conclude that the specific shape of the initial wave function is not crucial to the subsequent dynamical behavior. What is important is that more than one contiguous well is occupied. We find that in general, when the initial state consists of *at least two adjacent Wannier states of a band which is well isolated from higher bands*, the amplitude of the position expectation value of

Fig. 4 (a) Total potential energy, $V(z) + eFz$, (right ordinate) as a function of z for a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95$ Å, $b = 25$ Å, and $F = 2.5$ kV/cm. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to a stationary Gaussian function. (b) Probability density as a function of z and t/τ_B for the initial state and total potential energy shown in (a).

the Bloch oscillation is on the order of $(a/2)[W/(eFa)]$, where W is the energy band width. This finding is in agreement with single-band tight-binding (SBTB) predictions for the amplitude of the oscillations of the position expectation value, $z(t)$, as discussed in Appendix A.

C. Bloch Breathing Mode

In contrast to the above results, in which the Bloch oscillations involve an oscillatory center-of-mass motion, a remarkably different result is seen when $\psi(z,0)$ consists of only a single Wannier state, or of a linear combination of Wannier states which are not contiguous, i.e., where each occupied site is separated from another by at least one empty site. In Fig. 5(a) is shown the initial probability density for the choice $\psi(z,0) = |0,0\rangle$, a single Wannier function at site $n = 0$ in band $l = 0$. The spatial and temporal dependence of the probability density for this case is shown in Fig. 5(b). Although the motion is periodic with a period τ_B , *there is no oscillatory center-of-mass motion*. This special class of Bloch oscillations, henceforth referred to as "Bloch breathing modes," has the character of a time-periodic coherent breathing mode with width of order $(a/2)[W/(eFa)]$.

These Bloch breathing modes are perhaps surprising, in that they depart from the traditional picture²⁷ of Bloch oscillations as sinusoidal center-of-mass oscillations. In the special case of a Bloch breathing mode, the probability density is virtually symmetric about $z = 0$ at all times, i.e., in the language of classical physics, the electron is equally likely to move *against* the force due to the electric field as it is to move with it. It should be remarked, however, that this symmetry is not seen in the site amplitudes, but only in the site probabilities. That is, the site amplitude at site n differs from that at site $-n$ by a phase factor, but the site probabilities at sites n and $-n$ are equal. These properties of Bloch breathing modes are predicted by the SBTB treatment given in Appendix A.

Fig. 5 (a) Total potential energy, $V(z) + eFz$, (right ordinate) as a function of z for a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95 \text{ \AA}$, $b = 25 \text{ \AA}$, and $F = 2.5 \text{ kV/cm}$. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to a single Wannier function of the lowest field-free miniband. (b) Probability density as a function of z and t/τ_B for the initial state and total potential energy shown in (a).

It should be remarked that, in all three of the above cases (Figs. 2-5), even though the initial state is chosen explicitly to be representable by Wannier states of a single band, the wave function at later times evolves according to the complete Hamiltonian, H , and therefore is free to make transitions to higher bands. However, the energy gap between the occupied lowest miniband and the next higher miniband is sufficiently large that the projection of the wave function onto states of higher bands, although not strictly zero, is entirely negligible for as long as we have monitored the system. This finding is in accordance with the Zener tunneling prediction^{2,26} that the interband transition rate should be low for $E_g \gg eFa$. For example, for the case shown in Figs. 2-3, greater than 99% of the probability density is still associated with the lowest band, even for times as long as $25\tau_B$. This is why the SBTB model (Appendix A) provides excellent quantitative predictions for dynamical behavior of the exact wave function for this system. We stress, however, that for conditions where the SBTB approximation is inadequate, such as for the systems we consider below, our computational approach provides a unique opportunity to explore a rich tapestry of dynamical phenomena which, regrettably, is not accessible to investigation by analytical techniques.

D. Coexisting Bloch and Intra-well Oscillations

We again consider superlattice A with $F = 2.5$ kV/cm, but now our choice of initial state is explicitly constructed of Wannier states of more than one band. It consists of six equally weighted Wannier functions centered at adjacent sites from each of the lowest ($l = 0$) and first excited ($l = 1$) superlattice bands, $\psi(z,0) = (1/\sqrt{12}) \sum_n (|n, 0\rangle + |n, 1\rangle)$, where again the sum extends over sites $n = -2, \dots, 3$. In this case, a single-band picture obviously does not apply. The initial probability density is shown in Fig. 6(a) along with the total potential energy function.

Fig. 6 (a) Total potential energy, $V(z) + eFz$, (right ordinate) as a function of z for a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95$ Å, $b = 25$ Å, and $F = 2.5$ kV/cm. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to a linear combination of Wannier functions of the two lowest field-free minibands. (b) Probability density as a function of z and t/τ_B for the initial state and total potential energy shown in (a).

Fig. 6(b) shows the probability density as a function of z and t/τ_B . A remarkable result is seen: Bloch oscillations occur, even though the initial wave function occupies *two* minibands. Note that a localized portion of the wave packet (yellow in color) oscillates between approximately $z = 0$ and $z = -50$ nm with the period τ_B , closely resembling the motion in Fig. 2(b). We conclude that this is a Bloch oscillation due to the portion of the wave function associated with the lowest miniband.

In addition, there is a large amplitude oscillation (dark green) running from approximately $z = 0$ to $z = 200$ nm (and a much weaker component in the opposite direction), with the Bloch period. By comparison to a case with an initial wave function consisting of the Wannier states associated with sites $n = -2, \dots, 3$ from only the $l = 1$ miniband, we have determined that the large amplitude oscillation in Fig. 6 is due to the portion of the wave packet associated with the wider (43 meV) $l = 1$ miniband. The projection of the wave function onto Wannier functions of each miniband reveals that, for times up to $\sim 10 \tau_B$, half of the probability density lies in the $l = 0$ miniband and half lies in the $l = 1$ miniband. Thus, the Bloch oscillations in the two occupied minibands appear to evolve essentially uncoupled from and independent of each other.

Note that the majority of the probability density in the $l = 1$ miniband is initially accelerated in the direction *opposite* to the classical electric force, a result which was at first surprising to the authors. Intuition, and predictions from earlier work¹⁻⁸ on Bloch oscillations, would suggest that the electron would be accelerated *in the direction* of the classical force until a Bragg reflection compelled it to change direction. We have found that in general, any portion of the wave function occupying a miniband with an even band index l is accelerated in the direction of the electric force, whereas the wave function occupying an odd- l miniband is accelerated opposite to the electric force. These findings are also borne out in the SBTB results (see Appendix A).

After the fact, we can rationalize these results in terms of the following qualitative arguments. Consider the miniband structure of the superlattice, $\epsilon(k)$, shown in Fig. 7. We assign to an electron an "overall effective mass", M^* , obtained not from the crystal band structure of the constituent GaAs or AlGaAs materials, but from the *superlattice* miniband structure, $\epsilon(k)$, by the conventional formula²⁹

$$\frac{1}{M^*} = \frac{1}{\hbar^2} \frac{d^2\epsilon}{dk^2}. \quad (12)$$

We would argue that, in the same way the crystal band structure is used to normalize the electron mass so that it may be considered as having an effective mass m^* , so also can the superlattice band structure renormalize the mass, so that it has an overall effective mass M^* .

The linear combination of Wannier functions we have chosen as initial state tends to weight the Bloch eigenfunctions associated with wave vector k near zero most heavily, so that the initial wave packet has a quasimomentum centered near $\hbar k = 0$. Note that the curvature of the even- l minibands is positive around $k = 0$, whereas in the odd- l minibands it

Fig. 7 Miniband dispersion relation, $\epsilon(k)$ as a function of k , for a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w=95\text{\AA}$ and $b=25\text{\AA}$.

is negative. By this definition of the overall effective mass (10), an electron near $k = 0$ in any even- l miniband would have a positive M^* , whereas near $k = 0$ in an odd- l miniband it would have a *negative* M^* . Hence, an electron in an even- l miniband is initially accelerated in the *same* direction as the electric force, but in an odd- l miniband it is accelerated in the *opposite* direction. A more rigorous treatment of the issue is warranted, but these qualitative arguments do provide a rather simple explanation of the acceleration direction in each band.

In addition to the Bloch oscillations associated with each of the occupied bands, another feature is also seen in Fig. 6. When the probability density maximum is near $z = 0$ (near the times $t = 0, \tau_B, 2\tau_B, \dots$), there are some small-amplitude high-frequency intra-well oscillations (evident in the orange-yellow color range), which are reminiscent of the classical intra-well oscillations mentioned in Sec. I. These oscillations are more striking when the probability density is viewed in animated form. Then one can actually see a small portion of the wave function in each quantum well rocking back and forth at a high frequency.

The major features of this phenomenon can be elucidated by a simple time-dependent perturbation theory (see Appendix B), where the potential energy due to the electric field, $H' = eFz$, is treated as a small perturbation on the system of an electron confined within a single potential well, $H_0 = T + U(z)$, where $U(z) = 0$ ($z \leq w/2$); ∞ ($z > w/2$). Specifically, this simple theory predicts that for the short time scales considered here, on the order of a few picoseconds, the frequency of the intra-well oscillation is given by

$$\nu = (E_1 - E_0) / h, \quad (11)$$

where E_1 and E_0 are two bound state energy eigenvalues of the unperturbed Hamiltonian, H_0 , and h is Planck's constant. In the numerical solutions of the TDSE for Wannier-Stark electrons, we find that the intra-well oscillation frequency can also be described by (11), with E_1 and E_0 lying approximately in the center of the higher and lower minibands, respectively. In addition, the amplitude of the intra-well oscillations observed in the numerical simulations of Wannier-Stark electrons is in accord with qualitative predictions of the perturbation

treatment. Specifically, with an initial wave function consisting of the Wannier states from the lowest band only, the amplitude of the intra-well oscillations is so small as to be unobservable ($< 1 \text{ \AA}$ for superlattice A and $F = 2.5 \text{ kV/cm}$), whereas initial wave functions consisting of Wannier states from more than one band result in intra-well oscillations with observable amplitudes ($\sim 10 \text{ \AA}$).

We remark that the Bloch oscillation amplitude is not always larger than the intra-well oscillation amplitude, as it is in Fig. 6. In cases where eFa is large compared to the energy width, W , of any occupied band, but where still $eFa \ll E_g$, the Bloch oscillations are suppressed [recall that their amplitude depends on $W/(eFa)$], and the intra-well oscillations dominate the dynamical behavior.

Fig. 6 illustrates the seemingly paradoxical dual role of the electric field. On the one hand, Bloch oscillations are an effect of the electric field's giving the electron a small acceleration so that it may be Bragg reflected and repeat its motion. In order for the Bloch oscillations to occur there must be *negligible* coupling between states of different bands. On the other hand, the intra-well oscillations are a direct result of the electric field's coupling the local states within a particular well, so that intra-well oscillations only occur when there is *significant* coupling between states of different bands. These two conditions seem to contradict each other, yet the two dynamical phenomena can exist simultaneously.

E. Acceleration Regime

The dynamical phenomena discussed in the previous sections, Bloch oscillations, Bloch breathing modes, and intra-well oscillations, arise when the band gap energy is large compared to eFa so that the Zener tunneling rate is very low.^{2,26} We turn now to a superlattice with sufficiently small band gaps that the Zener tunneling rate is quite high,^{2,26} and in which the Bloch oscillations, Bloch breathing modes, and intra-well oscillations are supplanted by unbounded acceleration behavior.

We present selected results for superlattice B: a GaAs/Al_{0.2}Ga_{0.8}As superlattice with $w = 200 \text{ \AA}$ and $b = 5 \text{ \AA}$ ($a = 205 \text{ \AA}$). For this system, the potential barrier height is $V_0 = 162 \text{ meV}$, and the effective mass has the value $0.067m_e$ in the GaAs layers and $0.084m_e$ in the Al_{0.2}Ga_{0.8}As layers. Table II gives the minimum and maximum energy of each of the four lowest minibands of this superlattice, along with the band widths and band gaps. The associated Wannier functions for the lowest three minibands, one of which will be used to construct the initial state below, are shown in Fig. 1(d)-(f).

TABLE II Minimum and maximum energy values, E_{min} and E_{max} , respectively, for miniband of band index l of a GaAs/Al_{0.2}Ga_{0.8}As superlattice with $w = 200 \text{ \AA}$ and $b = 5 \text{ \AA}$. Also included are the miniband widths, W , and the energy gaps, E_g , between minibands. All energy values are given in meV.

l	E_{min}	E_{max}	W	E_g
0	3	13	10	7
1	20	53	33	28
2	61	119	58	69
3	127	212	85	

Note that the band gaps in Table II are substantially smaller than those given in Table I. For the data that follow, we utilize a field strength $F = 3.9 \text{ kV/cm}$, or $eFa = 8 \text{ meV}$ ($\tau_B = 0.52 \text{ psec}$). The Zener tunneling rate^{2,26} [see Eq. (11)] in this superlattice should be quite high, ~ 0.5 per Bloch period.

For an initial state, we choose a linear combination of six contiguous Wannier functions associated with the lowest band, $\psi(z,0) = 6^{-1/2} \sum_n |n,0\rangle$, $n = -2, \dots, 3$. [The Wannier function $|0,0\rangle$ is shown in Fig. 1(d).] The initial probability density is shown in Fig. 8(a), along with the total potential energy function.

Fig. 8 (a) Total potential energy, $V(z) + eFz$, (right ordinate) as a function of z for a GaAs/Al_{0.2}Ga_{0.8}As superlattice with $w = 200$ Å, $b = 5$ Å, and $F = 3.9$ kV/cm. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), consisting of six contiguous Wannier functions of the lowest field-free miniband. (b) Probability density as a function of z and t/τ_B for the initial state and total potential energy shown in (a).

The time evolution of the probability density is shown in Fig. 8(b). Although there is a faintly recognizable damped Bloch oscillation seen in Fig. 8(b), the dominant behavior is acceleration, as indicated by the large probability which moves in the negative z direction (in the direction of the electric force) in an approximately parabolic trajectory starting at time $t = 0$. In addition, after one Bloch time, another portion of probability is again accelerated, while only a very small probability density remains in a damped Bloch oscillating mode.

Some insight may be gained into the relationship between the Zener tunneling and the observed dynamical phenomena by examining the projection of the wave function onto the Wannier states of several of the lowest bands. Recall that in the cases where Bloch oscillations are long-lived, as, for example, in Fig. 3, the projection of the wave function onto the initially occupied bands remains essentially unchanged for as long as we monitor the system, even up to fifty Bloch periods.

Fig. 9 shows the projection of the wave function onto the Wannier states of the l th miniband, $P_l(t)$, defined by

$$P_l(t) = \sum_{n=-\infty}^{\infty} |\langle \psi(t) | n, l \rangle|^2,$$

for each of the four lowest minibands in the present case, in which the gap between the occupied miniband and the next higher miniband is slightly smaller than eFa . Initially the wave function is associated with only the lowest miniband, since it is constructed explicitly of Wannier functions of that band. However, after $0.5\tau_B$, the projection onto that band has decreased to approximately 0.5, while the projection onto the first excited miniband has grown from zero initially to approximately 0.5. Then, at approximately τ_B , the projection onto the first excited band drops off while that onto the second excited band begins to grow. Later still, the projection onto the second excited band drops off while that onto the third excited band begins to grow. The solid circles in Fig. 9 indicate the sum of the projections onto the four lowest bands. After approximately $1.8\tau_B$, this sum departs from unity,

Fig. 9 Projection, $P_l(t)$ of the wave function onto Wannier functions of bands $l = 0$ (open circles), $l = 1$ (open diamonds), $l = 2$ (open triangles), $l = 3$ (open squares), and the sum of the projections onto bands $l = 0, 1, 2$, and 3 (solid circles), as a function of t/τ_B .

indicating that some probability has succeeded in making a transition to bands higher than $l = 3$. Thus the probability density is seen to cascade from one band to the next, and the real-space manifestation of this cascade (Fig. 9) is an unbounded acceleration of a portion of the wave function anti-parallel to the electric field (Fig. 8(b)).

It should be remarked that Zener tunneling and acceleration are not always the dominant phenomenon in a narrow gap superlattice such as that summarized by Table II. For example, with the same initial wave function as that used in Fig. 6, but with a weaker electric field (e.g., $eFa = 0.5$ meV), we find that Bloch oscillations occur which are as robust as those seen in Fig. 2, but with a much greater amplitude [$W/(eFa) = 20$].

One might expect, by analogy to the behavior of a classical particle in a periodic potential subject to a uniform electric field, that the unbounded acceleration evident in Fig. 8(b) would only occur for that portion of the probability density which has succeeded in

making a transition to bands which lie above the barrier height V_0 . In classical terms, once the electron has energy sufficient to surmount a local maximum of the combined (lattice plus field) potential, it should be free to accelerate.

To test this hypothesis, we return to the superlattice summarized in Table I, with $eFa = 6$ meV, and with an initial state consisting of two Wannier functions associated with the $l = 2$ band, $\psi(z, 0) = (1/\sqrt{2})(|0, 2\rangle + |1, 2\rangle)$. This initial probability density and potential energy function are shown in Fig. 10(a). Note that the energy minimum of the $l = 2$ band ($E_{min} = 256$) is greater than $V_0 = 243$ meV. If the hypothesis is true, that what enables the electron probability to accelerate is that it is occupying minibands which are unbound, then this entire wave function should accelerate, since it all lies in an unbound band.

The probability density for this system as a function of z and t/τ_B is shown in Fig. 10(b), and one notes an unmistakable Bloch oscillatory behavior. Indeed, acceleration effects are entirely absent. Thus, even for an initial wave function occupying a band which lies higher in energy than the potential energy barriers, Bloch oscillations are a robust dynamical phenomenon. Examination of the projection of the wave function onto the four lowest minibands indicates that greater than 99% of the probability density remains within the $l = 2$ miniband for times as long as $10\tau_B$. We conclude, therefore, that the acceleration behavior is not a result of probability density occupying unbound bands, but rather it is a manifestation of the Zener tunneling cascade from one band to the next.

Put simply, the description first given by Zener² provides a useful way of characterizing the electron dynamics even when the complete Hamiltonian H is dealt with free of approximations. Namely, the electron begins to accelerate due to the electric field until it reaches the zone boundary. If interband transitions occur, the electron *continues* to accelerate. If, however, no transition occurs, the electron is Bragg reflected and undergoes Bloch oscillations. It is the very absence of interband transitions that allows the curious phenomenon of Bloch oscillations to occur.

Fig. 10 (a) Total potential energy, $V(z) + eFz$, (right ordinate) as a function of z for a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 95$ Å, $b = 25$ Å, and $F = 2.5$ kV/cm. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), consisting of two adjacent Wannier functions of the $l = 2$ field-free miniband. (b) Probability density as a function of z and t/τ_B for the initial state and potential energy shown in (a).

IV. CONCLUSIONS

In this work we have presented the first systematic investigation of the full range of dynamical phenomena exhibited by Wannier-Stark electrons. By solving the time-dependent Schrödinger equation based on the complete Hamiltonian, H , using high-accuracy numerical methods, we have been able to map out the dynamical behavior of Wannier-Stark electrons in the complicated parameter space of periodic potentials, electric field strengths, and initial wave functions. Specifically, we have presented some of our major results for the exact time-dependent behavior of Wannier-Stark electrons in idealized semiconductor superlattices.

We have demonstrated that under certain restricted conditions Wannier-Stark electrons do, in fact, execute long-lived Bloch oscillations, for twenty-five or more periods, with no noticeable decay and negligible interband transitions. Thus, the long-standing question as to whether Bloch oscillations exist or not in an ideal quantum mechanical system has finally been answered. Additionally, these results may be interpreted as supporting the notion of a WSL-like spectrum of equally spaced, sharply peaked energy resonances, with lifetimes which are limited by Zener tunneling.

In different situations the electron wave function can exhibit other dynamical phenomena, which can coexist with, or even mask, the Bloch oscillations. These include Bloch breathing modes, intra-well oscillations, and unbounded acceleration. The precise blend of these different basic elements depends upon the field-free minibands associated with the superlattice, the field strength, and the form of the initial wave function. The conditions under which each phenomenon occurs were summarized in Sec. IIIA.

Within the past year, four experiments¹⁷⁻²⁰ have been reported which have provided the first convincing experimental evidence for the existence of Bloch oscillations. All four

experiments¹⁷⁻²⁰ were based on electrons selectively excited into the lowest miniband of a GaAs/Al_xGa_{1-x}As superlattice. For all the choices of field strength employed, the band gap separating any occupied miniband from the next higher miniband was much greater than eFa .

Feldmann¹⁷ and collaborators¹⁸ and Leo *et al.*¹⁹ have performed degenerate four-wave mixing experiments and detected photon echo beats at time delays corresponding to integer multiples of the Bloch period, in agreement with the theory of von Plessen and Thomas.³⁰ More recently, Waschke *et al.*²⁰ have directly detected the electromagnetic radiation emitted by electrons undergoing Bloch oscillations in the electrically biased superlattice structure. Anywhere from one to eight Bloch oscillations were observed,¹⁷⁻²⁰ but in all cases the detected signal decayed rapidly over time. Feldmann *et al.*^{17,18} attributed this decay to loss of coherence due to scattering processes and interband transitions.

As we have shown, however, under conditions similar to those in the experiments, the wave function undergoes Bloch oscillations for as many as twenty-five periods with no noticeable decay and negligible interband transitions. Thus, interband transitions cannot be responsible for the signal decay seen in the experiments. In fact, we conclude that the decay observed in the experiments is due to effects not included in the independent-electron Hamiltonian we have employed. For example, in a real superlattice the electron could undergo elastic or inelastic scattering processes from static impurities, phonons, holes, or other electrons.

We have pointed out previously¹⁶ that static impurities could give rise to the signal decay observed in experiments.¹⁸ This issue is investigated more thoroughly in a forthcoming publication.³¹ Dignam and Sipe³² have undertaken studies of exciton states in semiconductor superlattices, by including the Coulomb interaction between an electron and a hole. Such investigations may also prove useful for interpreting the recent experimental results.¹⁷⁻²⁰ In any event, the present challenge for theorists is to expand the problem to

include more realistic features of the genuine experimental system in order to understand the dynamics of electrons in real semiconductor superlattices.

APPENDIX A: SINGLE-BAND TIGHT-BINDING MODEL

The single-band tight-binding (SBTB) model provides an excellent approximation for superlattices of the type considered in Secs. III B, C, and D (i.e., where the band gap separating a single occupied miniband from the next higher miniband is large compared to eFa), and can elucidate many features of the dynamical behavior found in our simulations. The SBTB method consists of, first, approximating the complete Hamiltonian $H = H_0 + eFz$, by that portion of its spectral representation corresponding to a single band l ,

$$\tilde{H}_l = \sum_{m,n=-\infty}^{\infty} \langle m,l | H | n,l \rangle |m,l\rangle \langle n,l|,$$

and second, choosing the initial wave function as expandable exclusively in terms of Wannier functions $|n,l\rangle$ of the single band l . Then the SBTB version of the TDSE,

$$i\hbar \frac{\partial \psi}{\partial t} = \tilde{H}_l \psi,$$

ensures that the wave function at all later times can also be so expanded, as

$$\psi(z,t) = \sum_{n=-\infty}^{\infty} f_n(t) |n,l\rangle,$$

where $f_n(t)$ is termed the site amplitude for site n . Henceforth, we shall suppress the band index l .

The Wannier functions $|n\rangle$ utilized in the SBTB model are related to those used for the numerical solution of the TDSE by

$$|n\rangle_{SBTB} = i^n |n\rangle_{num}.$$

In particular, the $|n\rangle_{num}$ are real functions of z for all integers n and l , whereas the $|n\rangle_{SBTB}$ are in general complex functions of z . For brevity, we shall employ the notation $|n\rangle$, with no

subscript, to denote for the SBTB Wannier functions and $|n\rangle_{num}$ for those associated with the numerical simulations.

The site amplitudes, $f_n(t)$, were obtained in Ref. 4, Eq. (42), as

$$f_n(t) = \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} f_m(0) e^{-im\omega_B t} \int_0^{2\pi} d\varphi \exp \left\{ -i \left[(n-m)\varphi + \frac{1}{\hbar} \int_0^t dt' V(\varphi - \omega_B t') \right] \right\}. \quad (A1)$$

For an arbitrary choice of the initial site amplitudes, $f_m(0)$, we obtain, by analogy to the calculations performed in Ref. 4,

$$f_n(t) = \sum_{m=-\infty}^{\infty} f_m(0) (-1)^{n-m} e^{-\frac{i}{2}(n+m)\omega_B t} J_{n-m}(\xi), \quad (A2)$$

where note carefully that ξ is a *time-dependent* quantity defined by

$$\xi \equiv [4V_1 / (eFa)] \sin(\omega_B t / 2), \quad (A3)$$

$V_1 = \langle 0 | H | 1 \rangle_{num}$, and J_n is the Bessel function of order n . In the tight-binding scheme, the width, W , is equal to $4V_1$.

We now proceed to calculate the position expectation value, $z(t)$:

$$\begin{aligned} z(t) &= \int_{-\infty}^{\infty} dz \psi^*(z,t) z \psi(z,t) \\ &= \sum_{n,n'=-\infty}^{\infty} f_n^*(t) f_{n'}(t) \langle n | z | n' \rangle \\ &= \sum_{n,n'=-\infty}^{\infty} f_n^*(t) f_{n'}(t) [na \delta_{n,n'} + \langle 0 | z | n - n' \rangle]. \end{aligned}$$

The matrix element $\langle 0 | z | n - n' \rangle$ can be written as

$$\langle 0 | z | n - n' \rangle = \langle 0 | z - (n - n')a/2 | n - n' \rangle + (n - n')a/2 \langle 0 | n - n' \rangle \quad (A4)$$

The second term is identically zero, since the Wannier functions are orthonormal. If the Wannier function $|n\rangle$ has even parity about $z = na$, as is the case for the $l = 0$ miniband (see

Fig. 1), then the first term on the right side of (A4) is also zero, since the integrand is odd about $z = (n - n')/2$. Thus, the position expectation value is seen to be

$$z(t) = a \sum_{n=-\infty}^{\infty} n |f_n(t)|^2. \quad (\text{A5})$$

If the Wannier function $|n\rangle$ does not have even parity about $z = na$, as is the case in other minibands (see Fig. 1), then the off-diagonal matrix elements $\langle 0 | z | n - n' \rangle$ are non-zero. However, due to the localized character of the Wannier functions, the off-diagonal matrix elements are small compared to the diagonal elements. We therefore neglect the off-diagonal elements and use (A5) as an approximation in the investigation of bands with index $l \neq 0$.

Inserting (A2) into (A5) gives

$$z(t) = a \sum_{m, m'=-\infty}^{\infty} f_m^*(0) f_{m'}(0) (-1)^{m+m'} e^{-\frac{i}{2}(m'-m)\omega_B t} \sum_{n=-\infty}^{\infty} n J_{n-m}(\xi) J_{n-m'}(\xi). \quad (\text{A6})$$

We rewrite the sum on n as

$$\sum_n n J_{n-m}(\xi) J_{n-m'}(\xi) = m \sum_n J_n(\xi) J_{n-p}(\xi) + \sum_n n J_n(\xi) J_{n-p}(\xi), \quad (\text{A7})$$

where we have defined $p \equiv m - m'$, in order to take advantage of a useful Bessel function addition theorem due to Graf,³³

$$J_\nu(\omega) \cdot \left(\frac{Z - ze^{-i\varphi}}{Z - ze^{i\varphi}} \right)^{\frac{1}{2}\nu} = \sum_{m=-\infty}^{\infty} J_{\nu+m}(Z) J_m(z) e^{im\varphi}, \quad (\text{A8})$$

where $\omega \equiv \sqrt{Z^2 + z^2 - 2Zz \cos \varphi}$ and φ is an arbitrary angle. A special case of this identity is

$$\sum_n J_n(\xi) J_{n-p}(\xi) = J_p(0) = \delta_{p,0}.$$

This gives rise to a contribution to $z(t)$ in (A6), due to the first sum in (A7), given by

$$a \sum_m m |f_m(0)|^2 = z(0). \quad (\text{A9})$$

To evaluate the second sum in (A7), we utilize the Graf identity (A8) in the form

$$\begin{aligned} \sum_{n=-\infty}^{\infty} J_n(\xi) J_{n+p}(\xi) e^{in\varphi} &= J_p(2\xi \sin \varphi/2) \cdot \left(\frac{1 - e^{-i\varphi}}{1 - e^{i\varphi}} \right)^{p/2} \\ &= (-e^{-i\varphi})^{p/2} J_p(2\xi \sin \varphi/2). \end{aligned}$$

We now differentiate with respect to φ and let $\varphi \rightarrow 0$ to obtain

$$\sum_{m=-\infty}^{\infty} n J_n(\xi) J_{n+p}(\xi) = (\xi/2)(\delta_{p,1} + \delta_{p,-1}). \quad (\text{A10})$$

Combining (A7), (A9), and (A10) with (A6), we find

$$z(t) = z(0) - \xi \frac{a}{2} \left[\sum_{m=-\infty}^{\infty} f_m^*(0) f_{m-1}(0) e^{\frac{i}{2} \omega_B t} + \sum_{m=-\infty}^{\infty} f_{m-1}^*(0) f_m(0) e^{-\frac{i}{2} \omega_B t} \right]. \quad (\text{A11})$$

If we define

$$S \equiv \sum_{m=-\infty}^{\infty} f_m^*(0) f_{m-1}(0), \quad (\text{A12})$$

(A11) reduces to

$$z(t) = z(0) - \xi a |S| \cos \left(\arg(S) + \frac{1}{2} \omega_B t \right). \quad (\text{A13})$$

Substitution of (A3) into (A12) gives

$$z(t) = z(0) - \frac{4V_1}{eFa} \frac{a}{2} |S| [\sin(\omega_B t + \arg(S)) - \sin(\arg(S))]. \quad (\text{A14})$$

The amplitude of the oscillations depends on the values of $f_m(0)$ at nearest neighbor sites via (A12). It is easily seen, however, that the modulus of S is less than or equal to unity. Thus, recalling that the band width is $W = 4V_1$, the maximum possible amplitude is $(a/2)[W/(eFa)]$.

If the initial state is given by a single Wannier function, i.e., $f_n(0) = \delta_{n,0}$, or several Wannier functions separated by at least one vacant site, the quantity S is identically zero, and $z(t) = z(0)$ for all times. That is, there is no center-of-mass motion. This is consistent with the results of Fig. 5, which shows the wave function undergoing a time-periodic Bloch breathing mode, with no center-of-mass motion. By contrast, if at least two adjacent sites have non-zero amplitude initially, the position expectation value oscillates with period τ_B , as shown below.

Consider the specific example of the initial wave function shown in Fig. 2(a), $\psi(z,0) = 6^{-1/2} \sum_n |n,0\rangle_{num}$, $n = -2, \dots, 3$. The initial site amplitudes are thus given by

$$f_n(0) = \langle n, 0 | \psi(z, 0) \rangle = (-i)^n \text{num} \langle n, 0 | \psi(z, 0) \rangle,$$

i.e.,

$$f_n(0) = \begin{cases} \frac{1}{\sqrt{6}} (-i)^n, & n = -2, \dots, 3 \\ 0, & \text{otherwise.} \end{cases}$$

The quantity S for this case is found to be $S = -(5/6)i = (5/6)\exp(-i\pi/2)$. Upon substitution of S into (A14) we finally obtain

$$z(t) = z(0) + \frac{5}{6} \frac{a}{2} \left(\frac{4V_1}{eFa} \right) (1 - \cos \omega_B t). \quad (\text{A11})$$

From the $l = 0$ Wannier functions [shown in Fig. 1(a)], we compute $V_1 = -2.64$ meV. Using $eFa = 3$ meV and $a = 120\text{\AA}$, we find that the amplitude of the Bloch oscillation is $176\text{\AA}$, so that the position expectation value should oscillate between $60\text{\AA}$ and $-292\text{\AA}$. Comparison of this prediction with the position expectation value as computed from the exact solution of the TDSE, Fig. 3, indicates extremely good agreement. Similar results are obtained for any initial state from the $l = 0$ miniband which has at least two adjacent sites with non-zero amplitude. The amplitude and phase of the center-of-mass oscillation are modified only through the quantity S .

For the $l = 1$ Bloch oscillation, which is a component of the behavior depicted in Fig. 6, we find that the position expectation value should oscillate between $60\text{\AA}$ and $1364\text{\AA}$, in good agreement with the exact results. In that case the motion is initially in the positive z direction because $V_1 = 10.68$ meV is positive.

We again emphasize that the SBTB model is an excellent approximation to the exact problem in these cases because the Zener tunneling from the occupied band to higher bands is entirely negligible for many multiples of the Bloch period. Clearly, the intra-well oscillations and acceleration effects, which are features of the exact dynamics of Wannier-Stark electrons could not be predicted by such an approximate treatment.

APPENDIX B: TIME-DEPENDENT PERTURBATION THEORY FOR AN INFINITE POTENTIAL WELL WITH A PERTURBING ELECTRIC FIELD

In order to gain an intuitive understanding of the major features of the intra-well oscillations, we consider the dynamics of an electron of effective mass m^* and charge $-e$ in an infinite potential well of width w , so that $V(z) = 0, |z| < w/2; = \infty, |z| > w/2$. The effects of the electric field originate from a term in the Hamiltonian of the form eFz . We will consider fields such that eFw is small compared to the quantum well energy level spacing, and thus it is justified to treat this term as a small perturbation. In the following we solve the time-dependent Schrödinger equation for this system. This model should provide reasonable qualitative predictions for the local electronic behavior in superlattices where the potential energy due to the electric field is very small compared to the superlattice barriers.

The unperturbed eigenstates for the field-free infinite well are of the well-known form

$$\begin{aligned} |n^e\rangle &= \phi_n^e = \sqrt{\frac{2}{w}} \cos(2n+1) \frac{\pi z}{w} \quad (n = 0, 1, 2, \dots) \\ |m^o\rangle &= \phi_m^o = \sqrt{\frac{2}{w}} \sin \frac{2m\pi z}{w} \quad (m = 1, 2, 3, \dots) \end{aligned}$$

with energy eigenvalues

$$\begin{aligned} E_n^e &= \frac{\hbar^2 \pi^2}{2m^* w^2} (2n+1)^2 \\ E_m^o &= \frac{\hbar^2 \pi^2}{2m^* w^2} (2m)^2, \end{aligned}$$

where the superscripts e and o indicate even and odd parity states, respectively.

Utilizing the perturbing Hamiltonian $H' = eFz$, we find that the first order energy correction is identically zero for all states. The first order perturbed eigenstates, denoted u_n^e and u_m^o , are given by

$$u_n^e = \phi_n^e + A \sum_{m=1}^{\infty} B_{mn} \phi_m^o \quad (\text{B1})$$

$$u_m^o = \phi_m^o - A \sum_{n=1}^{\infty} B_{mn} \phi_n^e, \quad (\text{B2})$$

where $A = eF 2m^* w^2 / (\hbar^2 \pi^2)$ and $B_{mn} = \langle m^o | z | n^e \rangle [(2n+1)^2 - 4m^2]^{-1}$. The functions u_n^e and u_m^o of course do not have any parity properties, yet it is convenient to maintain the e, o designations.

Now we proceed to solve the TDSE,

$$i \hbar \frac{\partial \psi}{\partial t} = (H_0 + eFz) \psi, \quad |z| < w/2$$

with the boundary condition $\psi(\pm w/2, t) = 0$. We expand the wave function in terms of the first-order perturbed eigenstates given by (B1) and (B2),

$$\psi = \sum_{n=0}^{\infty} c_n^e \exp(-iE_n^e t / \hbar) u_n^e + \sum_{m=1}^{\infty} c_m^o \exp(-iE_m^o t / \hbar) u_m^o, \quad (\text{B3})$$

where $c_n^e = \langle u_n^e | \psi(z, 0) \rangle$ and $c_m^o = \langle u_m^o | \psi(z, 0) \rangle$.

We first consider the behavior when the system is initially in the ground state of the unperturbed Hamiltonian, $\psi(z, 0) = \phi_0^e$. This would most closely resemble the superlattice system with an initial state consisting of a single Wannier function of the lowest band.

The coefficients of (B3) are found to be $c_n^e = \delta_{n,0}$ and $c_m^o = -A B_{m0}$, so that the wave function is given by

$$\psi = e^{-iE_0^e t / \hbar} \left\{ \phi_0^e + A \sum_{m=1}^{\infty} B_{m0} [1 - e^{-i(E_m^o - E_0^e)t / \hbar}] \phi_m^o \right\}. \quad (\text{B4})$$

It is straightforward to obtain the position expectation value to order F ,

$$z(t) = 2A \sum_{m=1}^{\infty} B_{m0} \langle 0^e | z | m^o \rangle [1 - \cos(E_m^o - E_0^e)t / \hbar] + O(F^2). \quad (\text{B5})$$

Utilizing the reasonable numerical values $m^* = 0.067m_e$, $w = 95 \text{ \AA}$, and $F = 2.5 \text{ kV/cm}$, and the fact that

$$\langle 0^e | z | m^o \rangle = (-1)^{m+1} \frac{16w}{\pi^2 (4m-1)^2},$$

we find that the $m = 1$ term dominates the sum, so that the leading term of $z(t)$ is given by

$$z(t) \approx 0.2 \text{ \AA} [1 - \cos (E_m^e - E_0^e) t / \hbar]. \quad (\text{B6})$$

The amplitude of the intra-well oscillation is very small compared to the width of the well. In fact, if intra-well oscillations are occurring along with the Bloch oscillations of Fig. 2 or Fig. 4, they would not be observable with such a small amplitude. However, as shown in the following, intra-well oscillations arising from different initial states can have much larger amplitudes, so as to be observable on the same scale as the Bloch oscillations.

Consider an initial wave function which is an equally weighted linear combination of the ground state and the first excited state of the unperturbed Hamiltonian, $\psi(z,0) = 2^{-1/2}(\phi_0^e + \phi_1^o)$. This system is analogous to the superlattice with an initial state which is a linear combination of Wannier functions from the two lowest minibands. In this case the coefficients are $c_n^e = 2^{-1/2}(\delta_{n,0} + A B_{0n})$ and $c_m^o = 2^{-1/2}(\delta_{m,0} - A B_{m0})$, so that

$$\begin{aligned} \psi = 2^{-1/2} & \left\{ e^{-iE_0^e t / \hbar} \left[\phi_0^e + A \sum_{m=1}^{\infty} B_{m0} \phi_m^o \right] \right. \\ & + A \sum_{n=0}^{\infty} B_{0n} e^{-iE_n^e t / \hbar} \left[\phi_n^e + A \sum_{m=1}^{\infty} B_{mn} \phi_m^o \right] \\ & + e^{-iE_1^o t / \hbar} \left[\phi_1^o + A \sum_{n=0}^{\infty} B_{0n} \phi_n^e \right] \\ & \left. + A \sum_{m=1}^{\infty} B_{m0} e^{-iE_m^o t / \hbar} \left[\phi_m^o + A \sum_{n=0}^{\infty} B_{mn} \phi_n^e \right] \right\} \quad (\text{B7}) \end{aligned}$$

In calculating the position expectation value, it is clear that the dominant terms will contain matrix elements of the form $\langle 0^e | z | 1^o \rangle$, since any terms containing the factor AB_{mn} will be smaller by at least a factor of order 10^{-4} for any m,n . The leading term of $z(t)$ turns out to be

$$z(t) \approx 17 \text{ \AA} \cos (E_1^o - E_0^e) t / \hbar. \quad (\text{B8})$$

Here the intra-well oscillations are much larger than in (B6). In fact, such oscillations are observable on the same scale as the Bloch oscillations. The qualitative predictions of the difference in amplitude of intra-well oscillations from this simple time-dependent

perturbation theory are borne out in the superlattice simulations. In Fig. 6, in which the initial state consists of Wannier functions from the two lowest bands, the intra-well oscillations are observable, but in Figs. 2 and 5, they are not, because the corresponding initial states contained only states from the single lowest band. Some intra-well oscillations are also observable in Fig. 4, in which the projection of the initial wave function onto Wannier states of the lowest and first excited bands equal 0.91 and 0.09, respectively. Despite the low occupancy (9%) in the first excited band, it is sufficient for the intra-well oscillations to be observable.

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PAPER III

**SOLVABLE DYNAMICAL MODEL OF AN ELECTRON
IN A ONE-DIMENSIONAL APERIODIC LATTICE
SUBJECT TO A UNIFORM ELECTRIC FIELD**

**Solvable dynamical model of an electron in a one-dimensional aperiodic lattice
subject to a uniform electric field**

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ABSTRACT

We provide the exact solution of the time-dependent Schrödinger equation for an electron in a one-dimensional aperiodic lattice in the presence of a static uniform electric field, in the single-band tight-binding approximation. This aperiodic lattice can be described as an otherwise periodic lattice with the form of the potential modified in a single unit cell. We find that the eigenvalue spectrum is discrete and may be characterized as a Stark ladder, but of nonuniform spacing. The electron's dynamical behavior may be described in terms of functions which are "almost-periodic" in the time variable. We examine the departure from time-periodic Bloch oscillations, which occur in the corresponding periodic system, and its dependence on impurity strength, electric field, lattice parameters, and initial wave function.

This aperiodic lattice potential may provide a reasonable model of a semiconductor superlattice with a single impure unit cell. We speculate that almost-periodic oscillations associated with weak impurities in semiconductor superlattices could give rise to the damped oscillatory behavior observed in recent experiments. Elsewhere we provide high-accuracy numerical solutions of the exact time-dependent Schrödinger equation for an electron in a superlattice with any number of added impurities, subject to a uniform electric field. The results of the current analytical SBTB treatment of the single-impurity problem prove to be of great value for interpreting the results of the numerical calculations in the case of multiple impurities.

I. INTRODUCTION

The study of the static and dynamic properties of an electron in a periodic lattice has been among the fundamental problems in the development of the physics of solids. One particular problem of long-standing interest¹⁻³ has been the dynamical behavior of independent electrons in a periodic lattice subject to a uniform electric field, henceforth referred to as "Wannier-Stark electrons." Bouchard and Luban^{4,5} recently solved the exact time-dependent Schrödinger equation for Wannier-Stark electrons in idealized semiconductor superlattices, using high-accuracy numerical methods. They demonstrated⁵ that under certain restricted conditions, a Wannier-Stark electron can undergo sustained time-periodic Bloch oscillations with period $\tau_B = h/(eFa)$ for many (> 25) Bloch periods with negligible deterioration. Here h is Planck's constant, e is the magnitude of the electronic charge, and a is the superlattice constant. The time-periodicity of the dynamical behavior can be associated with the spectrum of equally spaced energy eigenvalues, $E_l = E_0 + leFa$, termed the "Wannier-Stark ladder"^{6,7} (WSL). Bouchard and Luban^{4,5} modeled the idealized GaAs/Al_xGa_{1-x}As superlattices in the effective mass approximation in the conventional manner, namely, by a one-dimensional square-well/square-barrier potential energy function.

However, in many cases, a real semiconductor superlattice, indeed any real solid, may not be adequately described by such a simplistic, idealized model. In particular, the lattice may not be periodic, but rather be *aperiodic*. Fig. 1 shows some especially simple examples of one-dimensional aperiodic lattices. In 1954, Koster and Slater⁸ considered an aperiodic lattice of the type shown in Fig. 1(a), in which a single atom of type A is substituted into an otherwise periodic lattice of type-B atoms. Figs. 1(b)-(e) show different aperiodic Kronig-Penney-like potentials, with the modification of the depth of a single well [Fig. 1(b)], the

Fig. 1 Schematic illustrations of the spatial dependence of five one-dimensional aperiodic lattices with lattice constant a . (a) One type-A atom is substituted at a single site in a B-atom lattice. (b) The depth of a single well is modified in this Kronig-Penney-like potential (w is the well width, b is the barrier width). (c) A single barrier height is modified. (d) A single interface is translated by a distance δ . (e) A single well is widened by a width δ .

modification of a single barrier height [Fig. 1(c)], the translation of a single interface [Fig. 1(d)], or the modification of a single well width [Fig. 1(e)]. Figs. 1(b)-(d) may also be described by a Koster-Slater⁸ single-impurity model, since only a single unit cell of an otherwise periodic potential is distorted. Fig. 1(e), however, cannot be described by a Koster-Slater⁸ model, since the altered width of the central well renders the two half-lattices to the left and right to be out of register with each other. The examples in Fig. 1(b)-(e) could

also be used to model possible simple configurations of static impurities in semiconductor superlattices which could be introduced either intentionally or unintentionally during the growth process.

In this article we will explore the dynamics of electrons in Koster-Slater⁸ single-impurity aperiodic potentials, such as those shown in Fig. 1(a)-(d), in the presence of a uniform electric field. The system is described by the Hamiltonian $H = T + V(z) + U(z) + eFz$, where T is the kinetic energy operator, $V(z) = V(z+a)$ is the periodic lattice potential energy function, $U(z)$ is the aperiodic potential and is nonzero only within the $n = 0$ unit cell, and eFz is the potential energy associate with the electric field. Our purpose is to assess the role of the aperiodic potential in modifying the Bloch oscillations which occur in the corresponding Wannier-Stark problem, i.e., where $U(z) \equiv 0$.

Bouchard and Luban⁵ have shown that Bloch oscillations dominate the dynamical behavior of Wannier-Stark electrons if, for any occupied band, the following two conditions are met: (i) the band gap separating it from the next higher band is large compared to eFa , and (ii) the energy width is comparable to or greater than twice eFa . Under these conditions, if the initial wave function can reasonably be described by a linear combination of states associated with a single band, the rate of electron transitions to higher bands is nearly zero (for example, $\sim 10^{-4}$ per τ_B). Under these circumstances, a single-band tight-binding (SBTB) model provides a remarkably good approximation to the exact solution of the time-dependent Schrödinger equation (TDSE).⁵ The only difference between the present problem, of an electron in an aperiodic lattice subject to a uniform electric field, and the Wannier-Stark problem is the single impurity unit cell. Thus, a SBTB approach is likely to provide a good approximation to the TDSE for this aperiodic system, as well.

In this article, we provide the exact solution of the TDSE in the SBTB approximation for an electron in a one-dimensional Koster-Slater⁸ single-impurity aperiodic lattice in the presence of a static uniform electric field. For convenience we choose the impurity to be

located at lattice site $n = 0$. The solution of this problem for the special case of an electron occupying the impurity unit cell was reported earlier by Luban and Luscombe⁹. In this work we consider an arbitrary initial wave function.

We find that the dynamical behavior can be described in terms of functions which are "almost-periodic"¹⁰ in time. Specifically, the probability amplitude for the electron being found within any particular unit cell can be written as an infinite Fourier series of terms of the form $\exp(\omega_l t)$, where the frequencies ω_l ($l = 0, \pm 1, \pm 2, \dots$) are mutually incommensurate [$\omega_l / \omega_{l'}$ is an irrational number for all $l, l' (\neq l)$], and their values depend on F and the form of the impurity potential. Thus, over the duration of any realistic measurement, the exact form of the wave function is not repeated.

This almost-periodic time-dependence is related to the eigenvalue spectrum of the SBTB Hamiltonian in the following manner. We find that the impurity serves to distort the WSL, so that the eigenvalue spectrum consists of discrete energy levels, $E_l = E_0 + z_l eFa$, having a nonuniform spacing. Specifically, a small number of levels with l near zero may depart significantly from the equally spaced WSL of the corresponding periodic problem, but as l differs significantly from zero, the eigenvalues E_l approach the corresponding WSL eigenvalue, $E_0 + leFa$. If the eigenvalue spectrum were equally spaced, the corresponding dynamical behavior would be time-periodic Bloch oscillations. The distortion of the WSL away from equally spaced levels results in oscillations which are not time-periodic, but which have a frequency and amplitude which are on the same order of magnitude as those for the corresponding periodic system.

In the limit as the impurity strength approaches zero, we of course find that periodic Bloch oscillations are recovered. In fact, for weak impurity strengths (the meaning of the term "weak" will be made clear in Sec. III), the electron dynamics appears to be very nearly time-periodic for an extended time interval (several Bloch periods), before the effects of the

impurity become noticeable. This behavior thus has the *appearance* initially of Bloch oscillations, which then give way to almost-periodic oscillations.

Recent degenerate four-wave mixing (DFWM) experiments of J. Feldmann¹¹ and collaborators¹² and K. Leo *et al.*¹³ provided the first experimental evidence of Bloch oscillations in GaAs/Al_xGa_{1-x}As superlattices. The DFWM technique probes the extent to which the original wave form is maintained. If the electron wave function is coherently reproduced after each Bloch time τ_B , then a strong photon echo signal is detected at time delays corresponding to integer multiples of the Bloch period.¹⁴ The experiments¹¹⁻¹³ showed rapid dephasing of the signal, indicating that the Bloch oscillatory behavior was short-lived ($\sim 1-4$ periods). This is in striking contrast to the findings of Bouchard and Luban,⁵ that electrons in the very same superlattices used in the experiments would undergo as many as twenty-five Bloch oscillations with negligible deterioration. We speculate that almost-periodic oscillations due to weak impurities in such superlattices could give rise to the observed rapid dephasing. This and other features of the dynamical behavior of electrons in imperfect semiconductor superlattices will be presented in a forthcoming publication.¹⁵

We remark that although aperiodic potentials such as those shown in Fig. 1(b)-(d) are too simplistic to be accurate models of realistic impurities in semiconductor superlattices, nonetheless, the intuition which may be gained from an analytical solution of this simple single-impurity problem is of great value in understanding the effects of impurities on the dynamics of the electron wave function. In Ref. 15, we employ the complete Hamiltonian, H , rather than a SBTB approximation, to solve the TDSE for an electron in a periodic superlattice potential with any number of added impurities in the presence of a uniform electric field. The solution of the TDSE is obtained using high-accuracy numerical methods. Many of the results obtained in the present work by the SBTB method are valuable for interpreting the exact numerical results.

In Sec. II of this article, we derive the solution of the TDSE within the SBTB approximation. In Sec. III, we present selected results and discuss their implications for a special class of GaAs/Al_xGa_{1-x}As superlattices for which the SBTB model has been shown⁵ to provide a good approximation. A summary and conclusions are given in Sec. IV. The Appendix provides a detailed procedure for numerically obtaining the poles and residues discussed in Sec. II.

II. THEORY

A. Formulation

We consider independent electrons in a one-dimensional potential whereby a periodic background potential, $V(z) = V(z + a)$, is supplemented by an arbitrary aperiodic term, $U(z)$. As an example of such a potential energy function, see Fig. 2, which corresponds to the potential shown in Fig. 1(b). In the following, we consider the Koster-Slater⁸ single-impurity model, i.e., we assume that $U(z)$ is non-zero only within the unit cell centered about $z = 0$.

Fig. 2 Total potential energy $V_{total}(z)$ (top), periodic potential energy $V(z)$ (center), and aperiodic potential $U(z)$ (bottom) versus z . $V_{total}(z) = V(z) + U(z)$.

We now introduce a uniform external electric field directed along the positive z axis, $\vec{F} = F\hat{z}$. The TDSE is given by

$$i\hbar \frac{\partial \psi}{\partial t} = H \psi, \quad (1)$$

where the Hamiltonian H has the form

$$H = T + V(z) + U(z) + eFz. \quad (2)$$

Here $T = [-\hbar^2/(2m)] \partial^2/\partial z^2$ is the kinetic energy operator, and e is the magnitude of the electronic charge.. For convenience we also define the impurity-free Hamiltonian

$$H_1 = T + V(z) + eFz \quad (3)$$

and the field- and impurity-free Hamiltonian

$$H_0 = T + V(z). \quad (4)$$

Unfortunately, Eqs. (1)-(2) are too complicated to be solved exactly by analytical methods. Instead we consider this equation in the single-band tight-binding (SBTB) approximation, whereby the following two changes are made:

First, we require that the electron wave function be describable at all times as a linear combination of suitably chosen basis functions associated with a single band of index j of the field- and impurity-free Hamiltonian H_0 . Specifically, we employ as a basis the set of orthonormal Wannier functions, $\phi_j(z - na)$, so that

$$\psi(z, t) = \sum_{n=-\infty}^{\infty} f_n(t) \phi_j(z - na), \quad (5)$$

where

$$\int_{-\infty}^{\infty} dz \phi_j(z - na) \phi_j(z - ma) = \delta_{nm}. \quad (6)$$

The Wannier function $\phi_j(z - na)$ should be viewed as a localized function, of width on the order of $a/2$, centered about site n , or about $z = na$. The quantity $|f_n(t)|^2$ has the physical interpretation of being the probability for the electron to be found in the state $\phi_j(z - na)$ at time t . We will call $f_n(t)$ the probability amplitude for site n .

Second, we replace H by that portion of its spectral representation corresponding to the single band j , i.e.,

$$H \rightarrow \sum_{m,n=-\infty}^{\infty} \langle m, j | H | n, j \rangle |m, j\rangle \langle n, j|, \quad (7)$$

where $|n, j\rangle = \phi_j(z - na)$. Henceforth we shall suppress the band index.

Substituting (5) and (7) into (3), one directly obtains

$$i \hbar \dot{f}_n(t) = \sum_{m=-\infty}^{\infty} \langle n | H | m \rangle f_m(t). \quad (8)$$

That is, the single differential equation (1) is replaced by the infinite set of coupled first order differential equations (8) for the functions $f_n(t)$. The matrix element $\langle n | H | m \rangle$ appearing in (8) can be rewritten as follows:

$$\langle n | H | m \rangle = \langle 0 | H_1 | m - n \rangle + neFa \delta_{nn} + U_{nm}, \quad (9)$$

where we have defined $U_{nm} \equiv \langle n | U | m \rangle$. In view of the localized character of both the Wannier functions and $U(z)$, it is reasonable to adopt the approximation

$$U_{nm} \equiv U_{0,0} \delta_{m,0} \delta_{n,0}. \quad (10)$$

Indeed, utilizing Wannier functions constructed as described in Ref. 5, and impurities such as those described in Ref. 15, we find that $U_{0,0}$ is typically two orders of magnitude greater than the next largest matrix element, $U_{0,1}$. Thus the approximation (10) is well justified.

Combining equations (8)-(10), we arrive at the system of differential equations

$$i \hbar \dot{f}_n(t) = neFa f_n(t) + U_{0,0} \delta_{n,0} f_0(t) + \sum_{m=-\infty}^{\infty} \langle 0 | H_1 | m \rangle f_{m+n}(t). \quad (11)$$

Equation (11) will be solved without further approximations. By contrast, (3) and (4) are intractable.

It should be noted that for the case $U_{0,0} \equiv 0$, i.e., with no impurity, (11) has been solved in Ref. 16 [see Eq. (12) of that work]. The seemingly innocuous term, $U_{0,0} \delta_{n,0} f_0(t)$, complicates the problem considerably, but as shown below, (11) is exactly solvable by analytical methods.

The form of (11) can be simplified if we express the time, t , in terms of a new dimensionless variable, τ ,

$$t \equiv \frac{\hbar}{eFa} \tau. \quad (12)$$

Eq. (11) then reduces to

$$i\dot{f}_n(\tau) = n f_n(\tau) + \frac{1}{eFa} \left[U_{0,0} \delta_{n,0} f_0(\tau) + \sum_{m=-\infty}^{\infty} \langle 0 | H_1 | m \rangle f_{m+n}(\tau) \right], \quad (13)$$

where now the dot notation signifies a derivative with respect to τ .

B. Transformation to an Equivalent Integral Equation

The treatment to be given in this section is patterned after the method given in Ref.

16. We define an auxiliary function,

$$\Psi(\varphi, \tau) = (2\pi)^{-1/2} \sum_{n=-\infty}^{\infty} f_n(\tau) e^{in\varphi}. \quad (14)$$

The Fourier coefficient $f_n(\tau)$ is expressible in terms of Ψ by the inverse relation

$$f_n(\tau) = (2\pi)^{-1/2} \int_0^{2\pi} d\varphi e^{-in\varphi} \Psi(\varphi, \tau). \quad (15)$$

Multiplying (13) by $e^{in\varphi}$ and summing over all integers n one finds that the auxiliary function $\Psi(\varphi, \tau)$ satisfies the first order inhomogeneous partial differential equation

$$i \frac{\partial \Psi}{\partial \tau} = -i \frac{\partial \Psi}{\partial \varphi} + \frac{1}{eFa} \left[(2\pi)^{-1/2} U_{0,0} f_0(\tau) + V(\varphi) \Psi \right], \quad (16)$$

where

$$V(\varphi) = \sum_{n=-\infty}^{\infty} e^{-in\varphi} \langle 0 | H_1 | n \rangle. \quad (17)$$

Now (16) is of the so-called Lagrange form and can be solved exactly.^{16,17} The final result is

$$\Psi(\varphi, \tau) = \Psi(\varphi - \tau, 0) e^{-i[\tilde{V}(\varphi) - \tilde{V}(\varphi - \tau)]} - i(2\pi)^{-1/2} \xi e^{-i\tilde{V}(\varphi)} \int_{\varphi - \tau}^{\varphi} d\theta f_0(\tau - \varphi + \theta) e^{i\tilde{V}(\theta)}, \quad (18)$$

where

$$\xi = \frac{U_{0,0}}{eFa} \quad (19)$$

and

$$\tilde{V}(\varphi) = \frac{1}{eFa} \int_0^\varphi d\theta V(\theta). \quad (20)$$

Consider an arbitrary normalized initial wave function, i.e., the $f_n(0)$ are arbitrary except that

$$\sum_{n=-\infty}^{\infty} |f_n(0)|^2 = 1.$$

From (14), we then have the initial auxiliary function,

$$\Psi(\varphi, 0) = (2\pi)^{-1/2} \sum_{n=-\infty}^{\infty} f_n(0) e^{in\varphi}. \quad (21)$$

By substituting (21) into (18) and then inverting via (15), we obtain an expression for $f_n(\tau)$ as

$$\begin{aligned} f_n(\tau) = & \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} f_m(0) \int_0^{2\pi} d\varphi \exp \{ -i[(n-m)\varphi + \tilde{V}(\varphi) - \tilde{V}(\varphi - \tau) + m\tau] \} \\ & - \frac{i}{2\pi} \xi \int_0^\tau dT f_0(\tau - T) \int_0^{2\pi} d\varphi \exp \{ -i[n\varphi + \tilde{V}(\varphi) - \tilde{V}(\varphi - T)] \}. \end{aligned} \quad (22)$$

We remark that if we set $U_{0,0}$, or equivalently, ξ , to zero this expression correctly reduces to Eq. (42) of Ref. 16. (Note that in Ref. 16 the actual time t appears, whereas here we use the dimensionless time τ . The two quantities are related by $\tau = \omega_0 t$, with $\hbar \omega_0 = eFa$.)

For convenience, we define the quantities

$$f_n^{(0)}(\tau) \equiv \frac{1}{2\pi} \int_0^{2\pi} d\varphi \exp \{ -i[n\varphi + \tilde{V}(\varphi) - \tilde{V}(\varphi - \tau)] \} \quad (23)$$

and

$$g_n(\tau) \equiv \sum_{m=-\infty}^{\infty} f_m(0) e^{-im\tau} f_{n-m}^{(0)}(\tau). \quad (24)$$

Note that (23) is equivalent to Eq. (43) of Ref. 16. Thus, $f_n^{(0)}(\tau)$ denotes the site probability amplitude of an electron in a periodic potential in the presence of an electric field but in the absence of any impurities. It is expressed as an integral involving specified quantities and should therefore be viewed as a known quantity. It has been studied in detail in Ref. 16. Similarly, since $g_n(\tau)$ only involves specified quantities, it should also be viewed as a known quantity. We can now rewrite (22) as

$$f_n(\tau) = g_n(\tau) - i\xi \int_0^{\tau} dT f_0(\tau - T) f_n^{(0)}(T). \quad (25)$$

For $n = 0$, (25) is an integral equation with a convolution form:

$$f_0(\tau) = g_0(\tau) - i\xi \int_0^{\tau} dT f_0(\tau - T) f_0^{(0)}(T). \quad (26)$$

We now seek the solution, $f_0(\tau)$, of (26). For all other values of n , Eq. (25) provides an expression for $f_n(\tau)$, expressed as an integral (*not* an integral equation) of $f_n^{(0)}$ and f_0 . Once the $f_n(\tau)$ have been found for all n , we can obtain the full electron wave function $\psi(z, t)$ via (5).

C. Solution of the Integral Equation (26)

We use some elementary properties of Laplace transforms to solve the integral equation (26), in which $g_0(\tau)$ and $f_0^{(0)}(\tau)$ are known functions, and $f_0(\tau)$ is to be determined. Let $F_0(s)$, $G_0(s)$, and $F_0^{(0)}(s)$ denote the Laplace transforms of $f_0(\tau)$, $g_0(\tau)$, and $f_0^{(0)}(\tau)$, respectively,

$$F_0(s) = \int_0^{\infty} d\tau e^{-s\tau} f_0(\tau) \equiv \mathcal{L}\{f_0\}. \quad (27)$$

Taking the Laplace transform of both sides of (26) and recalling that the Laplace transform of the convolution integral is simply the product of the Laplace transforms of the individual functions in the convolution integral, we obtain

$$F_0(s) = G_0(s) - i\xi F_0(s) F_0^{(0)}(s), \quad (28)$$

or

$$F_0(s) = \frac{G_0(s)}{1 + i\xi F_0^{(0)}(s)}. \quad (29)$$

Performing the inverse transformation yields

$$f_0(\tau) = \mathcal{L}^{-1} \left\{ \frac{G_0(s)}{1 + i\xi F_0^{(0)}(s)} \right\}. \quad (30)$$

Before we can evaluate the right-hand side of (30), we must first obtain $f_0^{(0)}(\tau)$ and $g_0(\tau)$ and compute their Laplace transforms $F_0^{(0)}(s)$ and $G_0(s)$, respectively. To simplify this task, we specialize to the physically most relevant case of retaining only nearest-neighbor overlap integrals, i.e.,

$$\langle 0 | H_1 | n \rangle = iV_1 (\delta_{n,1} - \delta_{n,-1}), \quad (31)$$

so that (17) and (20) reduce to

$$V(\varphi) = 2V_1 \sin \varphi \quad (32)$$

and

$$\tilde{V}(\varphi) = \frac{2V_1}{eFa} (1 - \cos \varphi), \quad (33)$$

respectively. Now $f_n^{(0)}(\tau)$, given by (23), is easily shown to reduce to

$$f_n^{(0)}(\tau) = (-1)^n e^{-in\tau/2} J_n \left(\frac{4V_1}{eFa} \sin \frac{\tau}{2} \right), \quad (34)$$

where $J_n(x)$ is the Bessel function of order n . (See Eqs. (46)-(50) of Ref. 16.) By substituting (34) into (24), we find that

$$g_n(\tau) = \sum_{m=-\infty}^{\infty} f_m(0) (-1)^{n-m} e^{-i(n+m)\tau/2} J_{n-m} \left(\frac{4V_1}{eFa} \sin \tau/2 \right). \quad (35)$$

We are considering first the case where $n = 0$, so (34) and (35) become

$$f_0^{(0)}(\tau) = J_0\left(2\beta \sin \frac{\tau}{2}\right) \quad (36)$$

and

$$g_0(\tau) = \sum_{m=-\infty}^{\infty} f_m(0) (-1)^m e^{-im\tau/2} J_m(2\beta \sin \tau/2), \quad (37)$$

where we have defined the dimensionless parameter

$$\beta = \frac{2V_1}{eFa}. \quad (38)$$

To obtain the Laplace transform of (36), we first note that $f_0^{(0)}(\tau)$ is a periodic function of period 2π . (Recall that $J_0(x)$ is an even function of its argument.) We can therefore write

$$\begin{aligned} F_0^{(0)}(s) &= \sum_{n=0}^{\infty} \int_{2n\pi}^{2(n+1)\pi} d\tau e^{-s\tau} f_0^{(0)}(\tau) \\ &= \left(\sum_{n=0}^{\infty} e^{-2n\pi s} \right) \int_0^{2\pi} d\tau e^{-s\tau} f_0^{(0)}(\tau), \end{aligned}$$

so that

$$F_0^{(0)}(s) = \frac{1}{1 - e^{-2\pi s}} \int_0^{2\pi} d\tau e^{-s\tau} J_0(2\beta \sin \tau/2). \quad (39)$$

Upon evaluating the integral¹⁸ in (39), we obtain

$$F_0^{(0)}(s) = \frac{\pi}{\sinh \pi s} J_{is}(\beta) J_{-is}(\beta). \quad (40)$$

In the following it will be more convenient to employ the variable x defined by

$$s = ix, \quad (41)$$

so that

$$F_0^{(0)}(s) = -\frac{i}{x} K_0(x; \beta), \quad (42)$$

where we have defined a new function

$$K_0(x; \beta) = \frac{\pi x}{\sin \pi x} J_x(\beta) J_{-x}(\beta). \quad (43)$$

To similarly obtain the Laplace transform $G_0(s)$, note that $g_0(\tau)$ is a periodic function of period 2π . Also, the result for each term in the sum over m will depend on whether m is positive or negative. By analogy to Eqs. (36)-(43), we find that

$$G_0(s) = -\frac{i}{x} I_0(x; \beta), \quad (44)$$

where we have defined

$$I_0(x; \beta) = \sum_{m=-\infty}^{\infty} f_m(0) (-i)^m \frac{\pi x}{\sin \pi x} [J_{|m| \pm x}(\beta) J_{\mp x}(\beta)]_m. \quad (45)$$

In (45) and henceforth, the subscript m outside the square brackets denotes that the upper sign corresponds to $m \geq 0$, while the lower sign corresponds to $m < 0$.

Note that we can write I_0 in terms of K_0 as

$$I_0(x; \beta) = K_0(x; \beta) \tilde{I}_0(x; \beta), \quad (46)$$

where

$$\tilde{I}_0(x; \beta) = \sum_{m=-\infty}^{\infty} f_m(0) (-i)^m \left[\frac{J_{|m| \pm x}(\beta)}{J_{\pm x}(\beta)} \right]_m. \quad (47)$$

Finally we can write,

$$F_0(x) = \frac{-i K_0(x; \beta) \tilde{I}_0(x; \beta)}{x + \xi K_0(x; \beta)}. \quad (48)$$

Let us examine some properties of $F_0(x)$ which we will exploit in the next section to perform the inverse Laplace transform to obtain $f_0(\tau)$. Specifically, we show that $F_0(x)$ has simple poles coinciding with the zeros of

$$h(x; \xi, \beta) = x + \xi K_0(x; \beta), \quad (49)$$

the denominator of (48).

In terms of the variable x we can obtain a more convenient representation of K_0 using the formula¹⁹

$$K_0(x; \beta) = \frac{\pi x}{\sin \pi x} J_x(\beta) J_{-x}(\beta) = x \sum_{l=-\infty}^{\infty} \frac{J_l^2(\beta)}{x-l}. \quad (43')$$

This expansion is valid since K_0 , defined by (43), is analytic for all finite x except for an infinite set of simple poles, corresponding to the zeros $(0, \pm 1, \pm 2, \dots)$ of $\sin \pi x$. The form of (43') displays explicitly the location of the poles of K_0 and the corresponding residues. If we consider real, positive values of x , inspecting (43') we see that K_0 diverges to $+\infty$ ($-\infty$) as x approaches the point l from the right (left). In fact,

$$K_0(x \rightarrow l; \beta) \rightarrow \frac{l J_l^2(\beta)}{x-l}.$$

Similarly, for real, negative values of x , K_0 diverges to $+\infty$ ($-\infty$) as x approaches the point l from the left (right). For a given, fixed value of β the quantity $J_l^2(\beta)$ decreases to zero extremely rapidly for sufficiently large l . For example, for $\beta = 1$, $l = 10$, $l J_l^2(\beta) = 6.92 \times 10^{-19}$. This suggests that the singularity of K_0 should become evident only *extremely* close to l . This is confirmed by Fig. 3, in which $K_0(x; \beta)$ for the choice $\beta = 1$ is plotted as a function of x .

To obtain the zeros of $h(x; \xi, \beta)$ [Eq. (49)] it suffices to find the roots of the equation

$$K_0(x; \beta) = -\frac{x}{\xi}. \quad (50)$$

In Fig. 3, the straight lines $-x/\xi$ for $\xi = 0.01, 0.1$, and 1 are shown along with $K_0(x; \beta)$. For a given value of ξ , the line $-x/\xi$ intersects once with $K_0(x; \beta)$ in the open interval $(l, l+1)$ where l is any integer, and therefore $h(x; \xi, \beta)$ has one simple zero within the interval $(l, l+1)$. It follows that $F_0(x)$, Eq. (48), has simple poles coinciding with the zeros of $h(x; \xi, \beta) = x + \xi K_0(x; \beta)$. We now show that $F_0(x)$ has no other poles.

The numerator of (48) has poles at $x = l$, but at those poles the function F_0 itself reduces to the finite value

$$F_0(x=l) = -\frac{i}{\xi} \sum_{m=-\infty}^{\infty} f_m(0) (-i)^m \left[\frac{J_{|m| \pm l}(\beta)}{J_{\pm l}(\beta)} \right]_m. \quad (51)$$

In the special case where β is a zero of J_l or J_{-l} , $F_0(x=l)$ reduces to zero. This can be seen by evaluating (45) when β is a zero of J_l or J_{-l} . Hence, the only poles of the function $F_0(x)$ are at the zeros of $h(x; \xi, \beta)$ [Eq. (49)], and for all other finite x it is analytic and single-valued.

Fig. 3 $K_0(x; \beta=1)$ (solid curves) and $-x/\xi$ (broken lines, each labeled with the corresponding value of ξ) versus x .

D. Laplace Inversion of $F_0(s)$

1. Introduction

To obtain $f_0(\tau)$ we utilize the fact that this function can be expressed as a contour integral,²⁰

$$f_0(\tau) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} ds e^{s\tau} F_0(s), \quad (52)$$

where c is to be chosen so that the integrand is analytic for all $\text{Re } s > c$. As noted at the close of the previous section, the function F_0 is analytic and single-valued in s except at its poles, to be denoted $s_l = i x_l$, which coincide with the zeros of $h(x; \xi, \beta)$ [Eq. (49)]. The corresponding residues of F_0 are denoted r_l . That is, we will seek to write $F_0(s)$ in a form which shows explicitly the poles and residues, i.e.,

$$F_0(s) = \sum_l \frac{r_l}{s - s_l}.$$

Since all of the poles of F_0 lie on the real x axis (i.e. imaginary s axis), $F_0(s)$ is analytic for all $\text{Re } s > 0$, and we choose c to be any real positive number. The integral in (52) can be evaluated by closing the contour with a semicircle of infinite radius in the left-hand s -plane, and using Cauchy's formula. After verifying that the integrand does indeed vanish on the semicircle as the radius approaches ∞ , we have

$$f_0(\tau) = \sum_{l=-\infty}^{\infty} r_l e^{i x_l \tau}. \quad (53)$$

If the frequencies x_l in (53) are mutually incommensurate [$x_l/x_{l'}$ is an irrational number for all $l, l' (\neq l)$], then $f_0(\tau)$ has the characteristic signature of an "almost-periodic" function.¹⁰ Though the function is not periodic, i.e., there is no period τ_0 such that $f_0(\tau + \tau_0) = f_0(\tau)$ for all τ , yet one can find "quasi-periods" τ_i , ($i = 1, 2, 3, \dots$) such that $f_0(\tau + \tau_i)$ approximates $f_0(\tau)$ to within any desired degree of accuracy for all τ .

Our task thus reduces to calculating the infinite set of poles x_l and residues r_l and then evaluating the series (53). The first part will be achieved using a combination of analytical and numerical methods. The evaluation of (53) will be performed by numerical methods only. As shown below, typically the r_l decrease *very* rapidly with increasing l . Therefore, in practice it is sufficient to retain only a dozen or so terms in the series for most relevant values of ξ .

In the special case with no impurity potential, i.e. $\xi = 0$, from (48) one obtains

$$F_0(x) = -i K_0(x; \beta) \tilde{I}_0(x; \beta).$$

The poles are seen from (43') to be $x_l = l$, and the residues are $r_l = J_l^2(\beta) \tilde{I}_0(l; \beta)$, so that (53) reduces to

$$f_0^{(0)}(\tau) = \sum_{l=-\infty}^{\infty} J_l^2(\beta) \tilde{I}_0(l; \beta) e^{il\tau}. \quad (54)$$

The series (54) is a Fourier series describing a function periodic in τ . In terms of the real time variable the period is $\tau_B = h / (eFa)$, which is the period of Bloch oscillations. As we will show in Sec. III, there are generally only small modifications in the values of the poles x_l and residues r_l which arise when ξ is nonzero.

2. Determination of the Poles x_l

As mentioned in Sec II C, to obtain the zeros of $h(x; \xi, \beta)$ it suffices to find the roots of the equation

$$K_0(x; \beta) = -\frac{x}{\xi}. \quad (50)$$

In Fig. 3, the straight lines $-x/\xi$ for $\xi = 0.01, 0.1$, and 1 are shown along with $K_0(x; \beta)$, with $\beta = 1$. Clearly, (50) possesses an infinite number of roots, and these very rapidly approach the set of points l , where l is an integer.

It should be appreciated that it is essential to avoid overlooking any poles x_l . Most root-finding algorithms rely on one's ability to bracket the root, i.e., to know *a priori* that the desired root lies within a specified interval. We have shown that all of the x_l are on the real x axis, and that the interval $(l, l + 1)$ brackets precisely one root x_l . The roots of (50) may therefore be found by a simple bisection algorithm.²¹ However, because $K_0(x; \beta)$ has poles at $x = l$, care must be taken in evaluating $K_0(x; \beta)$ in the vicinity of $x = l$. A detailed procedure for obtaining the values of x_l is given in the Appendix.

3. Determination of the Residues r_l

We seek an expression for $F_0(x)$ which allows us to read explicitly the residues r_l at the poles x_l . To do so let us expand the function $h(x; \xi, \beta) = x + \xi K_0(x; \beta)$ in a Taylor series about its zero x_l :

$$h(x; \xi, \beta) \approx (x - x_l) \left[1 + \xi \frac{\partial K_0(x; \beta)}{\partial x} \Big|_{x \rightarrow x_l} \right]. \quad (55)$$

With (55) inserted into (48), we have

$$F_0(x) = \frac{-i K_0(x; \beta) \tilde{I}_0(x; \beta)}{(x - x_l) \left[1 + \xi \frac{\partial K_0(x; \beta)}{\partial x} \Big|_{x \rightarrow x_l} \right]}. \quad (56a)$$

In order to have the residue r_l shown explicitly, we want (56) to show the poles in terms of the variable s . Now for s approaching s_l (56a) becomes

$$F_0(s \rightarrow s_l) = \frac{K_0(x_l; \beta) \tilde{I}_0(x_l; \beta)}{1 + \xi \frac{\partial K_0(x; \beta)}{\partial x} \Big|_{x_l}} \frac{1}{s - s_l}, \quad (56b)$$

and the residue r_l is seen to be

$$r_l = \frac{K_0(x_l; \beta) \tilde{I}_0(x_l; \beta)}{1 + \xi \frac{\partial K_0(x; \beta)}{\partial x} \Big|_{x_l}}. \quad (57)$$

Since x_l is the root of (50), we can replace $K_0(x_l; \beta)$ in (57) with $-x_l/\xi$, so that (57) reduces to

$$r_l = -\frac{x_l}{\xi} \frac{\tilde{I}_0(x_l; \beta)}{1 + \xi \left. \frac{\partial K_0(x; \beta)}{\partial x} \right|_{x_l}}. \quad (58)$$

Some care must be taken in evaluating (58), since the derivative in the denominator can be quite large, particularly for large l , and straightforward evaluation may result in a floating point overflow. A procedure for a reliable evaluation of (58) is provided in the Appendix.

E. Solution of (25) for $n \neq 0$

Taking the Laplace transform of (25) for $n \neq 0$, we have

$$F_n(s) = G_n(s) - i\xi F_0(s) F_n^{(0)}(s). \quad (59)$$

If we recall (48), we obtain

$$F_n(x) = \frac{G_n(x)[x + \xi K_0(x; \beta)] - \xi K_0(x; \beta) \tilde{I}_0(x; \beta) F_n^{(0)}(x)}{x + \xi K_0(x; \beta)}. \quad (60)$$

From inspection of (60), it is clear that $F_n(x)$ will have simple poles at the zeros of $h(x; \xi, \beta) = x + \xi K_0(x; \beta)$, as before. We will now show that the inverse Laplace transform of (60) can be written as

$$f_n(\tau) = \sum_{l=-\infty}^{\infty} r_{nl} e^{ix_l \tau}, \quad (61)$$

where the $\{x_l\}$ are identical to those appearing in (53) and the residues r_{nl} are easily calculated once we employ the results for r_{0l} , from (58). In Eq. (61), the quantity $x_l \tau$ may also be written $x_l (eFa/\hbar) t$. From here we see that the energy spectrum, in real energy units, is given by $E_l = x_l (eFa)$. Thus, finding the poles, x_l , is equivalent to finding the energy spectrum. It follows that the $\{x_l\}$ must be, and in fact are, independent of site number n . That is, the energy spectrum is a characteristic of the system, and does not depend on the site

number. Note also that if for all l , $x_l = l$, the spectrum would be the equally spaced WSL. The degree to which the x_l depart from integer values determines the degree of distortion of the WSL.

We expand the denominator of (60) in a Taylor series about its zero x_l as in (55) and obtain $F_n(s)$ for s approaching s_l ,

$$F_n(x) \Big|_{s \rightarrow s_l} = \frac{-i\xi K_0(x_l; \beta) \tilde{I}_0(x_l; \beta) F_n^{(0)}(x_l)}{1 + \xi \left. \frac{\partial K_0(x; \beta)}{\partial x} \right|_{x_l}} \frac{1}{s - s_l}. \quad (62)$$

The residues r_{nl} can now be seen to be

$$r_{nl} = \frac{-i\xi K_0(x_l; \beta) \tilde{I}_0(x_l; \beta) F_n^{(0)}(x_l)}{1 + \xi \left. \frac{\partial K_0(x; \beta)}{\partial x} \right|_{x_l}}. \quad (63)$$

As before, we can replace $K_0(x_l; \beta)$ with $-x_l/\xi$. To further simplify (63), we must compute $F_n^{(0)}(x_l)$. The Laplace transform of (34) is very similar to a single term of (37). By analogy to (36)-(43), we find that

$$F_n^{(0)}(x) = i^{|n|} - 1 \frac{\pi}{\sin \pi x} \left[J_{|n| \pm x}(\beta) J_{\mp x}(\beta) \right]_n. \quad (64)$$

[Recall that the subscript n outside the square brackets denotes that the upper (lower) sign should be taken if $n \geq 0$ ($n < 0$).]

Note that we can write $F_n^{(0)}(x)$ in terms of $K_0(x; \beta)$ as

$$F_n^{(0)}(x) = K_0(x; \beta) \tilde{K}_0(x; \beta), \quad (65)$$

where

$$\tilde{K}_0(x; \beta) = i^{|n|} - 1 \frac{J_{|n| \pm x}(\beta)}{J_{\pm x}(\beta)} \Big|_n. \quad (66)$$

Substituting (65) and (66) into (63), we finally obtain

$$r_{nl} = -\frac{x_l}{\xi} \frac{\tilde{I}_0(x_l; \beta)}{1 + \xi \left. \frac{\partial K_0(x; \beta)}{\partial x} \right|_{x_l}} (-i)^{|n|} \left[\frac{J_{|n| \pm x}(\beta)}{J_{\pm x}(\beta)} \right]_n. \quad (67)$$

Note that r_{nl} is equal to the residue r_l found in (58) multiplied by an n -dependent factor, and that (67) correctly reduces to (58) for $n = 0$.

We have now completely specified the probability amplitudes $f_n(\tau)$ for all sites n . In the next section we compute numerical values for $f_n(\tau)$ as a function of τ and explore the eigenfrequency spectrum and dynamical behavior of the electron.

III. RESULTS AND DISCUSSION

In the theoretical treatment of Sec. II, all of the effects of the impurity strength, lattice potential, and electric field are cast in terms of the two dimensionless parameters, $\beta = 2V_1 / (eFa)$ and $\xi = U_{00} / (eFa)$. In addition, variations in the initial wave function are specified by the values of the initial site probability amplitudes $\{f_n(0)\}$. In this section we explore the dependence of the eigenfrequency spectrum, $\{x_l\}$, and the associated dynamical behavior, on β , ξ , and $\{f_n(0)\}$. We compare our results with the impurity-free case in which the spectrum is an equally spaced Wannier-Stark ladder (WSL) and the dynamical behavior is strictly periodic Bloch oscillations.

A. Eigenfrequency Spectrum

We first consider the eigenfrequency spectrum, $\{x_l\}$, which of course is independent of site number n and the initial probability amplitudes $\{f_n(0)\}$. We begin by examining Fig. 3 more closely. In this figure, β has a fixed value of unity, and we show several impurity strengths, $\xi = 0.01, 0.1$, and 1 . These values of β and ξ are reasonable for a wave function occupying the lowest miniband of a GaAs/Al_xGa_{1-x}As superlattice with well (GaAs layer) and barrier (AlGaAs layer) widths $w \sim 80$ Å and $b \sim 20$ Å, respectively, $eFa \sim 5$ meV, and with a weak impurity introduced in the unit cell centered around $z = 0$. For example, with 30% Al ($x = 0.3$) in the AlGaAs layers, the impurity might consist of up to $\sim 0.5\%$ Al contamination in the $n = 0$ quantum well, which is nominally a GaAs layer. The conduction band profile of this imperfect superlattice may be reasonably modeled by the potential energy functions shown in Fig. 2.

As seen in Fig. 3, for very small values of ξ (~ 0.01), the intersections, x_l , of the line $-x / \xi$ with $K_0(x; \beta = 1)$ are very nearly equal to the integers, i.e., $x_l \approx l$. As ξ is increased

to 0.1, we see that x_{-1} , x_0 , and x_1 depart slightly from integer values, but all other x_l appear to be very nearly equal to integers. With a still greater value of $\xi = 1$, we can see that x_{-1} , x_0 , and x_1 depart significantly from integer values, x_{-2} is slightly displaced from the value of $l = -2$, while all the other x_l are nearly equal to integers. The computed values of several of the x_l corresponding to the β and ξ used in Fig. 3 are given in Table I. The trends observed in the figure are confirmed in Table I.

Recall that for ξ identically zero (i.e., no impurity), the eigenvalue spectrum is the WSL, $E_l = leFa$. In these dimensionless units, this is equivalent to $x_l = l$. Hence Fig. 3 and Table I imply that the presence of an impurity at site $n = 0$ serves to distort the WSL near $l = 0$, and the degree of the distortion increases with increasing impurity strength.

Fig. 4 shows $K_0(x; \beta = 5)$ along with the lines $-x/\xi$ for $\xi = 0.01, 0.1$, and 1. The computed values of several of the x_l corresponding to the same β and ξ are given in Table II. These three cases correspond to the same electric field and impurity strength as in Fig. 3 and Table I, but with a matrix element $V_1 = \langle 0 | H | 1 \rangle$ which is five times greater than that corresponding to Fig. 3. This matrix element is proportional to the energy width of the occupied tight-binding band. Whereas $\beta = 1$ is a reasonable value representing the lowest energy miniband of a GaAs/Al_xGa_{1-x}As superlattice with $w \sim 80$ Å, $b \sim 20$ Å, and $eFa \sim 5$ meV, the choice $\beta = 5$ is more reasonable for a wider first or second excited miniband of the same superlattice. Alternatively, $\beta = 5$ would be reasonable for the lowest miniband of a superlattice with narrower barriers, say $b \sim 5$ Å.

Fig. 4 and Table II, as in Fig. 3 and Table I, show that the WSL is increasingly distorted with increasing ξ . In addition, by comparing Fig. 4 (Table II) with Fig. 3 (Table I), we see that for a fixed value of ξ , the distortion of the WSL is more pronounced with increasing β .

Table I Roots x_l for index l for a system with $\beta = 1.0$ and $\xi = 0.01$ (left), $\xi = 0.1$ (center), and $\xi = 1.0$ (right).

l	x_l	l	x_l	l	x_l
-15	-15.00000000	-15	-15.00000000	-15	-15.00000000
-14	-14.00000000	-14	-14.00000000	-14	-14.00000000
-13	-13.00000000	-13	-13.00000000	-13	-13.00000000
-12	-12.00000000	-12	-12.00000000	-12	-12.00000000
-11	-11.00000000	-11	-11.00000000	-11	-11.00000000
-10	-10.00000000	-10	-10.00000000	-10	-10.00000000
-9	-9.00000000	-9	-9.00000000	-9	-9.00000000
-8	-8.00000000	-8	-8.00000000	-8	-8.00000000
-7	-7.00000000	-7	-7.00000000	-7	-7.00000000
-6	-6.00000000	-6	-6.00000000	-6	-6.00000000
-5	-5.00000000	-5	-5.00000001	-5	-5.00000008
-4	-4.00000006	-4	-4.00000063	-4	-4.00000828
-3	-3.00000384	-3	-3.00003969	-3	-3.00059450
-2	-2.00013276	-2	-2.00139763	-2	-2.02893302
-1	-1.00194955	-1	-1.02073338	-1	-1.37867149
0	-0.00585514	0	-0.05841785	0	-0.47258058
1	0.99807653	1	0.98187767	1	0.88955445
2	1.99986870	2	1.99874907	2	1.99152060
3	2.99999619	3	2.99996304	3	2.99971783
4	3.99999994	4	3.99999940	4	3.99999513
5	5.00000000	5	4.99999999	5	4.99999995
6	6.00000000	6	6.00000000	6	6.00000000
7	7.00000000	7	7.00000000	7	7.00000000
8	8.00000000	8	8.00000000	8	8.00000000
9	9.00000000	9	9.00000000	9	9.00000000
10	10.00000000	10	10.00000000	10	10.00000000
11	11.00000000	11	11.00000000	11	11.00000000
12	12.00000000	12	12.00000000	12	12.00000000
13	13.00000000	13	13.00000000	13	13.00000000
14	14.00000000	14	14.00000000	14	14.00000000
15	15.00000000	15	15.00000000	15	15.00000000

Fig. 4 $K_0(x; \beta=1)$ (solid curves) and $-x/\xi$ (broken lines, each labeled with the corresponding value of ξ) versus x .

According to Eq. (61), if the x_l 's are given by integer values, as in the impurity-free case, the probability amplitudes $f_n(\tau)$ are time-periodic with the Bloch period. (Recall that $\tau = \omega_0 t = 2\pi t/\tau_B$.) In cases where an impurity distorts the WSL, the behavior described by (61) is termed "almost-periodic."¹⁰ Specifically, the probability amplitude $f_n(\tau)$ is an infinite sum of sinusoidal components with mutually incommensurate frequencies $[x_l/x_{l'}]$ is an irrational number for all $l, l' (\neq l)$, with each component weighted by the corresponding residue r_{nl} .

Table II Roots x_l for index l for a system with $b = 5$ and $x = 0.01$ (left), $x = 0.1$ (center), and $x = 1.0$ (right).

l	x_l	l	x_l	l	x_l
-15	-15.00000000	-15	-15.00000000	-15	-15.00000000
-14	-14.00000000	-14	-14.00000000	-14	-14.00000000
-13	-13.00000000	-13	-13.00000000	-13	-13.00000000
-12	-12.00000000	-12	-12.00000000	-12	-12.00000001
-11	-11.00000000	-11	-11.00000001	-11	-11.00000014
-10	-10.00000002	-10	-10.00000022	-10	-10.00000244
-9	-9.00000031	-9	-9.00000309	-9	-9.00003521
-8	-8.00000339	-8	-8.00003443	-8	-8.00040439
-7	-7.00002855	-7	-7.00029090	-7	-7.00358777
-6	-6.00017220	-6	-6.00176443	-6	-6.02332703
-5	-5.00068396	-5	-5.00702578	-5	-5.09415606
-4	-4.00153322	-4	-4.01556464	-4	-4.17593617
-3	-3.00133041	-3	-3.01324633	-3	-3.12381439
-2	-2.00002168	-2	-2.00021654	-2	-2.00213794
-1	-1.00107333	-1	-1.01075430	-1	-1.10829288
0	-0.00031541	0	-0.00315379	0	-0.03127537
1	0.99892717	1	0.98929563	1	0.89655248
2	1.99997831	2	1.99978288	2	1.99780352
3	2.99866838	3	2.98663319	3	2.86449205
4	3.99847197	4	3.98495411	4	3.87250894
5	4.99932006	5	4.99337707	5	4.94783094
6	5.99982872	6	5.99832730	6	5.98646401
7	6.99997157	7	6.99972085	7	6.99763822
8	7.99999662	8	7.99996667	8	7.99970856
9	8.99999970	9	8.99999699	9	8.99997314
10	9.99999998	10	9.99999979	10	9.99999807
11	11.00000000	11	10.99999999	11	10.99999989
12	12.00000000	12	12.00000000	12	11.99999999
13	13.00000000	13	13.00000000	13	13.00000000
14	14.00000000	14	14.00000000	14	14.00000000
15	15.00000000	15	15.00000000	15	15.00000000

Hence, if the W-S ladder is significantly distorted (large ξ, β) and in addition, the distorted portion of the spectrum is heavily weighted (large r_{nl}), then the dynamical behavior will reflect many different "competing" frequencies, and the wave function will not reproduce its original form within any reasonable measurable time scale. If, however, the WSL is not significantly distorted, or the essentially undistorted portion of the spectrum is heavily weighted, then the dynamical behavior closely resembles time-periodic Bloch oscillations for several periods before the almost-periodic behavior becomes evident. Thus it is not *only* the degree of distortion of the WSL which determines the degree of departure from Bloch oscillatory behavior, but also, and perhaps more importantly, the values of the residues which weight each frequency component. These issues are explored more fully below.

B. Dynamical Behavior

1. Introduction

Unlike the frequency spectrum $\{x_l\}$, the site probability amplitudes $f_n(\tau)$, given by (61), and the position expectation value depend on the initial site probability amplitudes, $\{f_n(0)\}$, through the residues of Eq. (67), with $\tilde{I}_0(x; \beta)$ given by (47). The position expectation value, $z(\tau)$, is defined by

$$\begin{aligned} z(\tau) &\equiv \int_{-\infty}^{\infty} dz \psi^*(z, \tau) z \psi(z, \tau) \\ &= \sum_{n, n'=-\infty}^{\infty} f_n^*(\tau) f_{n'}(\tau) \langle n | z | n' \rangle \\ &= \sum_{n, n'=-\infty}^{\infty} f_n^*(\tau) f_{n'}(\tau) [na \delta_{n, n'} + \langle 0 | z | n - n' \rangle]. \end{aligned} \quad (68a)$$

The matrix element $\langle 0 | z | n - n' \rangle$ can be written as

$$\langle 0 | z | n - n' \rangle = \langle 0 | z - (n - n')a/2 | n - n' \rangle + (n - n')a/2 \langle 0 | n - n' \rangle.$$

The second term is identically zero, since the Wannier functions are orthonormal. If the Wannier function $|n\rangle$ has even parity about $z = na$, as is the case for the $l = 0$ miniband (see Fig. 1 of Ref. 5), then the first term on the right side is also zero, since the integrand is odd about $z = (n - n')/2$. Thus, the position expectation value is seen to be

$$= a \sum_{n=-\infty}^{\infty} n |f_n(\tau)|^2. \quad (68b)$$

If the Wannier function $|n\rangle$ does not have even parity about $z = na$, as is the case in other minibands (see Fig. 1 of Ref. 5), then the off-diagonal matrix elements $\langle 0 | z | n - n' \rangle$ are non-zero. However, in experiments electrons are typically excited from the valence band into the lowest miniband of the conduction band, so (68b) is sufficient for the cases of greatest interest.

We can write $z(\tau)$ as an almost-periodic function by substituting the expression (61) for $f_n(\tau)$ into (68b) to yield

$$z(\tau) = a \sum_{l, l'=-\infty}^{\infty} W_{ll'} e^{i\omega_{ll'}\tau}, \quad (69)$$

where $W_{ll'}$ is defined by

$$W_{ll'} = W_{l'l} = \sum_{n=-\infty}^{\infty} n r_{nl} r_{nl'} \quad (70)$$

and $\omega_{ll'} = x_l - x_{l'}$. Here again, if the x_l are integers, as in the impurity-free case, then (69) describes a strictly periodic Bloch oscillation.

2. Single Wannier Function Initial State

We first consider the simple case where initially only the $n = 0$ site is occupied, i.e., $f_n(0) = \delta_{n,0}$. With this initial state, in the absence of any impurity, the electron wave packet undergoes a special type of Bloch oscillation, which we have termed a "Bloch breathing

mode,"⁵ which has the character of a time-periodic coherent breathing mode with period τ_B . For the Bloch breathing mode, the position expectation value is identically zero, independent of β .

Fig. 5 shows $z(\tau)/a$ as a function of τ for $\xi = 0$ (no impurity), 0.01, 0.1, and 1, with $\beta = 1$ (panel a) and $\beta = 5$ (panel b). For $\xi \neq 0$, the curves in panel a (b) were computed from (70) utilizing the poles x_l given in Table I (II) and the residues given by (67).

There are two key trends to note in Fig. 5. First, for a given value of β , as ξ is increased from zero, $z(\tau)$ departs more and more from the impurity-free value of zero. This is in accord with our earlier finding that as ξ is increased, the eigenvalue spectrum becomes increasingly distorted away from the uniformly spaced WSL of the impurity-free system.

Secondly, as β is increased from 1 to 5, the $z(\tau)$ curves for small values of ξ approach the impurity-free line $z(\tau) = 0$. In fact, in the case of $\beta = 5$, the curve for $\xi = 0.01$ is so close to zero that it is barely visible on the scale of the figure. This result is perhaps surprising, in that Table II, for $\beta = 5$, shows a *more* distorted eigenvalue spectrum than Table I, for $\beta = 1$. Therefore, one might expect the $\beta = 5$ curves to depart *more* from the impurity-free behavior than the $\beta = 1$ curves. However, as indicated above it is not only the frequencies, but also the *weights* of the different frequency components that determines the dynamical behavior.

Shown in Table III (IV) are the absolute value of the weights $W_{ll'}$ for selected values of l, l' for $\xi = 0.01$ and $\beta = 1$ (5). The absolute value of the elements of $W_{ll'}$ vary quite smoothly with l, l' , so Tables III and IV give good representations of the full matrices. For $\beta = 1$, we see that even though the eigenvalue spectrum is not very distorted from the WSL, the weights $W_{ll'}$ drop off extremely rapidly with increasing $|l|, |l'|$. Hence, the distorted few around $l, l' = 0$ are given much greater weight than the eigenvalues with large $|l|$, which are nearly equal to integers. By contrast, although the $\beta = 5$ spectrum is more distorted, the weights $W_{ll'}$ drop off much more slowly than in the case of $\beta = 1$. In fact, for $\beta = 5$ the .

Fig. 5 Position expectation value $z(\tau)/a$ versus τ for an electron initially occupying only site $n=0$, with (a) $\beta=1$ and (b) $\beta=5$, and $\xi=0$ (short dash), 0.01 (dash-triple-dot), 0.1 (long dash), and 1.0 (solid).

Table III Weights $W_{ll'}$ for selected l and l' for a system with $\beta = 1$ and $\xi = 0.1$. The l values run across the top; the l' values are listed down the left side.

	-15	-10	-5	0	5	10	15
-15	0.3E-66	0.4E-52	0.2E-40	0.3E-33	0.5E-40	0.4E-52	0.3E-66
-10	0.4E-52	0.1E-34	0.1E-22	0.4E-19	0.7E-26	0.5E-38	0.4E-52
-5	0.2E-40	0.1E-22	0.1E-7	0.1E-5	0.2E-13	0.7E-26	0.5E-40
0	0.3E-33	0.4E-19	0.1E-5	0.1E0	0.1E-5	0.4E-19	0.3E-33
5	0.5E-40	0.7E-26	0.2E-13	0.1E-5	0.1E-7	0.7E-23	0.2E-40
10	0.4E-52	0.5E-38	0.7E-26	0.4E-19	0.7E-23	0.3E-35	0.4E-52
15	0.3E-66	0.4E-52	0.5E-40	0.3E-33	0.2E-40	0.4E-52	0.3E-66

Table IV Weights $W_{ll'}$ for selected l and l' for a system with $\beta = 5$ and $\xi = 0.1$. The l values run across the top; the l' values are listed down the left side.

	-15	-10	-5	0	5	10	15
-15	0.7E-15	0.2E-10	0.3E-8	0.4E-10	0.2E-12	0.2E-17	0.6E-25
-10	0.2E-10	0.5E-6	0.9E-4	0.5E-4	0.2E-5	0.2E-10	0.8E-18
-5	0.3E-8	0.9E-4	0.2E-1	0.1E-1	0.1E-1	0.1E-5	0.9E-13
0	0.4E-10	0.5E-4	0.1E-1	0.8E-2	0.1E-1	0.5E-4	0.3E-10
5	0.2E-12	0.2E-5	0.1E-1	0.1E-1	0.2E-1	0.9E-4	0.2E-8
10	0.2E-17	0.2E-10	0.1E-5	0.5E-4	0.9E-4	0.5E-6	0.1E-10
15	0.6E-25	0.8E-18	0.9E-13	0.3E-10	0.2E-8	0.1E-10	0.3E-15

weights for $l, l' = \pm 15$ are around the same order of magnitude as those for $l, l' = \pm 5$ in the case of $\beta = 1$. Thus, a substantial portion of the essentially *undistorted* ladder spectrum is significantly weighted in the $\beta = 5$ case. The overall effect is that the $z(\tau)$ curve for $\beta = 5$ resembles the perfect WSL impurity-free case much more closely.

3. Multiple Wannier Function Initial State

We now turn to an initial wave function consisting of a linear combination of Wannier functions. As an example we take three equally weighted Wannier functions at sites $n = -1, 0$, and 1 . Specifically, $f_n(0) = 3^{-1/2}(\delta_{n,-1} + \delta_{n,0} + \delta_{n,1})$. (The choice of equal coefficients is nonessential. Qualitatively similar results are obtained for different linear combinations of Wannier functions.)

Fig. 6 shows $z(\tau)/a$ as a function of τ for this initial wave function, with $\xi = 0, 0.1$, and 1 and with $\beta = 1$ (panel a) and $\beta = 5$ (panel b). The solid lines correspond to $\xi = 0$, the impurity-free case, which is strictly periodic with period τ_B in real time units. The amplitude of the Bloch oscillation for this initial state is $A = (4/3)\beta a$ (see Appendix A of Ref. 5). From Fig. 6 we see that for fixed β , as ξ is increased, the departure from the Bloch oscillatory behavior becomes evident earlier. In addition, for fixed ξ , as β is increased, $z(\tau)$ follows the Bloch oscillation closely for a longer time before departing noticeably.

These trends can be understood both from a mathematical and an intuitive physical perspective. Mathematically, the frequencies $\omega_{ll'}$ are differences between the x_l shown in Tables I and II, and the weights $W_{ll'}$ are identical to those in Tables III and IV except for an overall factor which is independent of l and l' , arising from $\tilde{I}_0(x;\beta)$ [see Eq. (47)]. Therefore, we should expect the same trends as seen in Fig. 5, and this expectation is borne out in Fig. 6. From a more physical perspective, it is clear that in the absence of any impurity, Bloch oscillations would occur. However, if a very weak impurity is introduced into the

Fig. 6 $z(\tau)/a$ versus τ for an electron wave function initially occupying sites $n = -1, 0$, and 1 in a system with $\beta = 1$ (panel a) and $\beta = 5$ (panel b), and with $\xi = 0$ (solid), 0.1 (short dash), and 1.0 (dash-triple-dot).

system at site $n = 0$, that impurity disturbs the wave function very slightly whenever it, the wave function, overlaps significantly with the impurity site. Each Bloch period, as the wave function repeatedly passes through the vicinity of $n = 0$, it becomes more and more disturbed until finally the behavior no longer resembles a Bloch oscillation. If a large impurity is introduced, it may sufficiently disturb the initial wave function as to cause it to deviate substantially from the Bloch oscillation immediately. This explains the trend as ξ is varied.

As mentioned above, increasing β for fixed ξ is equivalent to increasing the tight-binding band width. The Wannier functions associated with a wider band are typically less spatially localized than those associated with a narrow band.⁵ For example, whereas narrow-band Wannier functions may be localized to a single quantum well, those associated with a wider band may extend over not only its central site, but its nearest-neighbor, and possibly even next-nearest-neighbor sites, as well. Thus, for the same initial probability amplitudes $f_n(0) = 3^{-1/2}(\delta_{n,-1} + \delta_{n,0} + \delta_{n,1})$, the wave function itself for the wider band is likely to be much more diffuse than that for the narrower band. Hence, the impurity impacts a smaller portion of the wave function, and therefore has less effect when β is large.

This interpretation of the trend which occurs as β is varied is supported by Fig. 7, which shows $z(\tau)$ versus τ for $\xi = 0.1$, $\beta = 1$, and for initial wave function including 3, 7, and 11 equally weighted contiguous Wannier functions, i.e.,

$$f_n(0) = m^{-1/2} \sum_{i=-(m-1)/2}^{(m-1)/2} \delta_{n,i},$$

where $m = 3, 7, 11$. Each curve is normalized by the corresponding Bloch oscillation amplitude, so that if it were a perfect Bloch oscillation it would show a periodic behavior between zero and unity. Fig. 7 indicates that the greater the number of included Wannier functions, or the more diffuse the wave packet, the less it is disturbed by the impurity, and the more slowly the dynamical behavior departs from the impurity-free case. This is in accord with the interpretation of the β trend exhibited by Fig. 6.

Fig. 7 Scaled position expectation value, $z(\tau)/(2Aa)$, versus τ for a system with $\beta = 1$, $\xi = 0.1$, and an electron wave function initially occupying three sites (solid), seven sites (long dash), and eleven sites (short dash). Here A is the amplitude of the corresponding Bloch oscillation (i.e., when $\xi = 0$).

4. Varying Electric Field Strength

Thus far, we have considered the effect of varying ξ while keeping β fixed, which is equivalent to varying the impurity strength, and the effect of varying β while keeping ξ fixed, which is equivalent to varying the energy band width. A third consideration would be to vary β and ξ together, which is equivalent to varying the electric field strength. Recall that $\beta = 2V_1 / (eFa)$ and $\xi = U_{00} / (eFa)$. If V_1 , U_{00} , and a are all fixed, and we vary F , then both β and ξ are modified by the same factor.

Fig. 8 shows $z(\tau)/a$ versus τ for different values of electric field, so that $\beta = \xi = 1, 0.5$, and 0.25 (panel a), and $\beta = \xi = 0.1$ and 0.01 (panel b). Note that β and ξ decrease as F is increased. The initial probability amplitudes are given by

Fig. 8 Position expectation value $z(\tau)/a$ versus τ for an electron wave function initially occupying sites $n = -1, 0$, and 1 in a system with (a) $\beta = \xi = 1.0$ (solid), 0.5 (dash-triple-dot), and 0.25 (short dash), and (b) $\beta = \xi = 0.1$ (solid) and 0.01 (dashed). The arrows at the right mark the upper limit of the Bloch oscillation which occurs in the corresponding impurity-free system with the designated value of β .

$f_n(0) = 3^{-1/2} (\delta_{n,-1} + \delta_{n,0} + \delta_{n,1})$. In the impurity-free case, the amplitude of the Bloch oscillation is proportional to β , or to $W/(eFa)$, where W is the band width. The upper limit (twice the amplitude) of the Bloch oscillations for the different values of β are marked by arrows on the right ordinate of Fig. 8.

Although one cannot identify a single "amplitude" for each of the curves in Fig. 8, it is clear that the electronic motion is bounded so that it oscillates between approximately zero and approximately the upper limit of the corresponding Bloch oscillation. Thus, in an aperiodic lattice, the electric field plays a similar role as in a periodic lattice. It serves to localize the wave function and confine the motion to a small region of space. The stronger the electric field, the smaller the spatial extent of the motion.

Another important point illustrated by Fig. 8 is the meaning of the term "weak" impurity. There are three relevant energies in the problem. One is U_{00} , which is a measure of the impurity strength. The second is V_1 , the magnitude of which is proportional to the band width, and is in that sense a measure of how tightly or loosely the Wannier functions are bound to their central site. The third is eFa , which is the potential energy difference from one site to the next due to the electric field. As shown above, if $\xi \ll \beta$, or more specifically, if $|U_{00}| \ll |V_1|$, then the departure from Bloch oscillatory behavior develops rather slowly. Under those conditions, the impurity may be considered "weak" in the sense that its ability to disturb the dynamical behavior, and cause it to depart from Bloch oscillations is not very great. In Fig. 8, we see that a separate condition can also allow slow departure from Bloch oscillations. When $\beta = \xi = 0.01$, the position expectation value appears to be periodic. In that case, U_{00} is *not* much smaller than V_1 , but it *is* much smaller than eFa . Thus if *either* $|U_{00}| \ll |V_1|$ *or* $|U_{00}| \ll eFa$, the impurity may be considered weak, because the dynamical behavior will have the *appearance* initially of Bloch oscillations, which then give way to almost-periodic oscillations.

Although we have shown results only for positive V_1 , we have found that the same general conclusions hold for negative values of V_1 , but the inequalities are more extreme. For example, for positive V_1 , as many as ten Bloch-like oscillations may be observed before almost-periodic behavior becomes evident when V_1 is approximately *ten* times U_{00} , whereas for negative V_1 , the magnitude of V_1 may have to be as much as *twenty-five* times U_{00} in order for the same number of Bloch-like oscillations to be observed before they give way to almost-periodic oscillations. As discussed elsewhere,¹⁵ a negative V_1 is typically obtained from Wannier functions associated with the lowest miniband of a superlattice, whereas a positive V_1 would typically be obtained from Wannier functions associated with a first excited miniband. Even though the band widths of two such bands may be the same, the Wannier functions associated with the lowest miniband are likely to be more localized than those corresponding to the first excited miniband. Therefore, in cases with negative V_1 , with Wannier functions which tend to be more localized, the wave function is more greatly influenced by an impurity. Therefore, it takes a particularly wide lowest miniband, or a particularly large magnitude of V_1 , in order for Bloch-like oscillations to occur initially, rather than almost-periodic oscillations setting in essentially immediately.

C. Relationship to Experiments

In recent DFWM experiments performed by Feldmann *et al.*^{10,11} and Leo *et al.*¹² on GaAs/Al_xGa_{1-x}As superlattices, only a few (1-5) Bloch oscillations were detected before the signal had completely decayed. Feldmann *et al.*^{10,11} attributed the rapid dephasing to scattering and electron transitions from one miniband to the next. However, Bouchard and Luban⁵ showed that in the same class of superlattices, with the field strengths employed in the experiments, interband transitions are entirely negligible for times on the order of 25 Bloch periods. Hence, some other mechanism must be responsible for the rapid dephasing.

The DFWM technique probes the extent to which the original wave form is maintained. Thus, if the electrons in the sample lose coherence so that the original wave function is not recovered after each Bloch time τ_B , then the expected photon echo peaks measured in the experiment decay over time.²² We speculate that almost-periodic oscillations due to scattering from weak impurities, such as those modeled in Fig. 1(b)-(e), for example, could give rise to the dephasing phenomena observed in the experiments. Such impurities could be generated from aluminum contamination in a nominally GaAs layer, incorrect aluminum concentration in an AlGaAs layer, an interface which is not atomically abrupt, etc. If the impurity is weak, i.e., if $U_{00} \ll V_1$ or $U_{00} \ll eFa$, then one might observe, in a DFWM experiment, a few Bloch oscillations which would steadily decay over time, as they give way to almost-periodic oscillations, as seen for example in Fig. 6(b). This possibility is explored more thoroughly in a forthcoming article¹⁵ which focuses specifically on electrons in aperiodic semiconductor superlattices subject to an electric field.

IV. SUMMARY

We have solved the time-dependent Schrödinger equation in the single-band tight-binding approximation for independent electrons in a Koster-Slater⁸ single-impurity aperiodic lattice subject to a uniform electric field. We showed that the eigenvalue spectrum consists of discrete energy levels having nonuniform spacing. Specifically, a small number of levels E_l for l near zero differ significantly from the corresponding Wannier-Stark ladder level $leFa$, but for $l > 10$, the energy eigenvalues agree very closely with the Wannier-Stark ladder. The stronger the impurity, the more distorted is the Stark ladder. The corresponding dynamical behavior is almost-periodic, i.e., it can be described by an infinite sum of sinusoidal components with mutually incommensurate frequencies.

If the impurity is strong and the distorted portion of the spectrum is heavily weighted, the dynamical behavior evidences many different "competing" frequencies, and the wave function never reproduces its original form on any measurable time scale. If, however, the impurity is weak *and* the undistorted portion of the spectrum is heavily weighted, i.e., if *either* $U_{00} \ll V_1$ *or* $U_{00} \ll eFa$, then the electron wave function will undergo oscillations which closely resemble Bloch oscillations for several periods before the almost-periodic behavior is clearly evident. We speculate that such short-lived Bloch oscillation-like behavior associated with small impurities could give rise to the rapid dephasing seen in recent degenerate four-wave mixing experiments¹¹⁻¹³ in GaAs/Al_xGa_{1-x}As superlattices.

APPENDIX: DETAILED PROCEDURE FOR COMPUTING POLES AND RESIDUES

In order to numerically evaluate the poles x_l and residues r_l , we consider another useful form for $K_0(x; \beta)$, which has been given in the text by (43) and (43'). Using the fact that $J_{-n}^2(\beta) = J_n^2(\beta)$, we have

$$\begin{aligned} K_0(x; \beta) &= J_0^2(\beta) + x \sum_{n=1}^{\infty} J_n^2(\beta) \left(\frac{1}{x-n} + \frac{1}{x+n} \right) \\ &= J_0^2(\beta) + 2 \sum_{n=1}^{\infty} J_n^2(\beta) + 2 \sum_{n=1}^{\infty} J_n^2(\beta) \frac{n^2}{x^2 - n^2} \\ &= 1 + 2 \sum_{n=1}^{\infty} J_n^2(\beta) \frac{n^2}{x^2 - n^2}. \end{aligned} \quad (\text{A1})$$

To obtain the last form (A1) we have used a limiting case of Neumann's addition theorem,²³ whereby

$$J_0^2(\beta) + 2 \sum_{n=1}^{\infty} J_n^2(\beta) = 1, \quad (\text{A2})$$

independent of β .

Now, consider in particular the vicinity of $x = l$. Using (A1) we can write

$$K_0(x; \beta) = \frac{\alpha_l}{x^2 - l^2} + k_l(x; \beta), \quad (l = 1, 2, 3, \dots) \quad (\text{A3})$$

where

$$\alpha_l = 2l^2 J_l^2(\beta) \quad (\text{A4})$$

and

$$k_l(x; \beta) = 1 + \sum_{\substack{n=1 \\ (n \neq l)}}^{\infty} \frac{\alpha_n}{x^2 - n^2}. \quad (\text{A5})$$

Note that for x near l (A5) is finite, but the first term of (A3) can be quite large. Now define

$$h_l \equiv \frac{\alpha_l}{x^2 - l^2} + k_l(x; \beta) + \frac{x}{\xi}, \quad (\text{A6})$$

which equals $h(x; \xi, \beta)$, but which we will use only in the interval $(l, l + 1]$. We seek the zero x_l of h_l . The first term of (A6) has a pole at $x = l$. As long as we keep our search restricted to the interval $(l, l + 1]$, we can multiply (A6) by $x^2 - l^2$ and search for the zeros of a new function

$$\tilde{h}_l = (x^2 - l^2) h_l = \alpha_l + (x^2 - l^2) \left(k_l(x; \beta) + \frac{x}{\xi} \right) \quad (\text{A7})$$

without introducing any new unwanted zeros. (A7) has the advantage that it has no terms which grow in the vicinity of $x = l$, so it can easily be evaluated without floating overflow difficulties. It is with (A7) that we proceed with a straightforward bisection algorithm to find the zero x_l .

To obtain the residues,

$$r_l = -\frac{x_l}{\xi} \frac{\tilde{I}_0(x_l; \beta)}{1 + \xi \left. \frac{\partial K_0(x; \beta)}{\partial x} \right|_{x_l}}, \quad (\text{A8})$$

we similarly make use of (A3). We can compute the partial derivative in (A8) as follows:

$$\left. \frac{\partial K_0}{\partial x} \right|_{x_l} = \frac{-2\alpha_l x_l}{(x_l^2 - l^2)^2} + \left. \frac{\partial k_l}{\partial x} \right|_{x_l}, \quad (\text{A9})$$

where

$$\frac{\partial k_l}{\partial x} = -2x \sum_{\substack{m=1 \\ (m \neq l)}}^{\infty} \frac{\alpha_m}{(x^2 - m^2)^2}. \quad (\text{A10})$$

Since at $x = x_l$ the function $h_l = 0$, we can solve (A6) for $x_l^2 - l^2$ and write (A9) as

$$\left. \frac{\partial K_0}{\partial x} \right|_{x_l} = \frac{-2x_l}{\xi^2 \alpha_l} (x_l + \xi k_l(x_l; \beta))^2 + \left. \frac{\partial k_l}{\partial x} \right|_{x_l}. \quad (\text{A11})$$

By inserting (A11) into (A8) and simplifying, we arrive at

$$r_l = \frac{\frac{1}{2} \alpha_l}{\left(x_l + \xi k_l(x_l; \beta)\right)^2 - \frac{\alpha_l \xi}{2x_l} \left(\xi \frac{\partial k_l}{\partial x_l} + 1\right)}. \quad (\text{A12})$$

Now notice from (A10) that $\partial k_l / \partial x_l$ has the opposite sign of x_l . Then we can finally write

$$r_l = \frac{\frac{1}{2} \alpha_l}{\left(x_l + \xi k_l(x_l; \beta)\right)^2 + \frac{\alpha_l \xi}{2|x_l|} \left(\xi \left|\frac{\partial k_l}{\partial x_l}\right| - \text{sgn } x_l\right)}. \quad (\text{A13})$$

It is (A13), together with (A10), that we actually evaluate to obtain r_l .

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PAPER IV

**QUANTUM DYNAMICAL PHENOMENA OF ELECTRONS
IN APERIODIC SEMICONDUCTOR SUPERLATTICES**

**Quantum dynamical phenomena of electrons
in aperiodic semiconductor superlattices**

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ABSTRACT

We solve the time-dependent Schrödinger equation for independent electrons in aperiodic semiconductor superlattices, with a superimposed uniform electric field, F , using high-accuracy numerical methods. Specific examples include the slight modification of the material composition in one or more superlattice layers and the translation of an interface by as little as a single monolayer in an otherwise periodic superlattice. This theoretical treatment allows one to model deviations from periodicity which can arise either intentionally or unintentionally during the superlattice growth process.

We find that, depending on the form of the aperiodicity, the miniband structure of the corresponding periodic superlattice, the value of F , and the initial wave function, a number of dynamical phenomena can occur. These include "almost-periodic" oscillations with frequency and amplitude similar to those of the Bloch oscillations which would occur in the corresponding periodic superlattice, high-frequency low-amplitude intra-well oscillations, and unbounded acceleration.

In many cases, electrons occupying a miniband with a large energy width initially exhibit many (> 10) oscillations with frequency and amplitude in close agreement with the Bloch oscillations which would occur in the corresponding periodic lattice, before the almost-periodic behavior becomes clearly evident. We propose that experiments based on electrons injected into large miniband-width superlattices may provide the opportunity to observe Bloch oscillations of significantly longer lifetime than have been seen before.

I. INTRODUCTION

The question of whether independent electrons in a periodic potential subject to a uniform electric field (henceforth referred to as "Wannier-Stark electrons") undergo Bloch oscillations^{1,2} has been a matter of great controversy³ for over six decades. The controversy largely has centered around the use of various uncontrolled approximations, the impact of which it is difficult to assess. Interest in this problem has intensified in recent years with the advent of epitaxial crystal growth techniques and the development of semiconductor superlattices of high mobility. In such systems, the superlattice constant, a , and electric field strength, F , can be on the order of 100 Å and 1 kV/cm, respectively. Thus the Bloch period, $\tau_B = h/(eFa)$, where h is Planck's constant and e is the magnitude of the electron charge, can be sufficiently small (~ 1 psec) that several oscillations may be completed in less than estimated⁴ electron scattering times. In addition, it has been proposed^{5,6} that electrons undergoing Bloch oscillations in superlattices could serve as a source of terahertz radiation.

Recent work by Bouchard and Luban^{6,7} verified that Bloch oscillations are a *bona fide* component of the exact dynamics of Wannier-Stark electrons in a certain class of ideal GaAs/Al_xGa_{1-x}As superlattices. They solved the time-dependent Schrödinger equation (TDSE) by high-accuracy numerical methods, utilizing the complete Hamiltonian for independent electrons in a one-dimensional periodic lattice potential plus a uniform electric field. However, recent degenerate four-wave mixing (DFWM) experiments by J. Feldmann *et al.*⁸ and K. Leo *et al.*⁹ on superlattices in this same class, although providing evidence in support of Bloch oscillations, also showed rapid dephasing of the signal, indicating that the Bloch oscillatory behavior was short-lived (~ 1 -4 oscillations before complete signal decay).

Feldmann *et al.*⁸ attributed the rapid dephasing to loss of coherence due to scattering and electronic interband transitions. However, Bouchard and Luban⁶ showed that in superlattices like those used in the DFWM experiments, Bloch oscillations are robust and long-lived with negligible interband transitions for times as long as $25 \tau_B$. Thus, interband transitions do not appear likely to be responsible for the observed rapid dephasing. Other mechanisms for coherence loss must play a role, such as scattering from static impurities, or from phonons, from other electrons, or from holes.

In this article we explore the role of static impurities on the dynamical behavior of Wannier-Stark electrons in GaAs/Al_xGa_{1-x}As superlattices. An infinite number of possible impurity configurations exists. We consider here a special class of aperiodic potentials involving the systematic modification of one or more entire layers of the superlattice. Fig. 1(a) shows the Kronig-Penney potential which is commonly used to model the conduction band of a periodic superlattice, along with some examples of aperiodic potentials we will consider. In Fig. 1(b), the impurity consists of a small amount (<1%) of aluminum uniformly contaminating a nominally GaAs layer. Fig. 1(c) shows an inexact aluminum concentration in an AlGaAs layer (differing from the nominal Al concentration by <1%). In Fig. 1(d), an interface between GaAs and AlGaAs layers is translated by as little as a single atomic monolayer.

Luban and Luscombe,¹⁰ and more recently Bouchard, Luban, and Luscombe¹¹ have shown that in the single-band tight-binding (SBTB) approximation, for cases in which only a single unit cell is modified, as in Fig. 1, the dynamical behavior can be described as "almost-periodic"¹² in the time variable. Specifically, the probability amplitude for the electron to occupy any particular state of the occupied band can be written as an infinite series of terms of the form $\exp(i\omega_j t)$, where the frequencies ω_j , ($j = 0, \pm 1, \pm 2, \dots$) are mutually incommensurate [$\omega_j / \omega_{j'}$ is an irrational number for all $j, j' (\neq j)$], and their values depend on

Fig. 1 (a) Model periodic superlattice potential, $V(z)$, as a function of z . Schematic model for the total superlattice potential, $V(z) = V(z) + U(z)$, and aperiodic potential, $U(z)$, for Al contamination in a single well (b), inexact Al composition in a barrier (c), and the translation of a single interface (d).

F , the superlattice potential, and the form of the aperiodicity. Thus, the dynamical behavior is not characterized by a single frequency, as is the case when no aperiodicity is present. Rather, it exhibits many different "competing" frequencies, so that on the time scale of any realistic measurement, the exact form of the wave function is not repeated.

In this article, we solve the TDSE for an electron in an aperiodic superlattice, subject to a uniform electric field, using high-accuracy numerical methods. We employ the Hamiltonian

$$H = T + V(z) + eFz, \quad (1)$$

where

$$V(z) = V(z) + U(z), \quad (2)$$

T is the kinetic energy operator, $V(z) = V(z + a)$ is the periodic superlattice potential [as in Fig. 1(a)] with lattice constant a , $U(z)$ is the aperiodic potential [as in Figs. 1(b)-(d)], and eFz is the scalar potential energy associated with the electric field, with e the magnitude of the electron charge. With this approach, we can explore not only the regime in which the single-impurity SBTB^{10,11} treatment is likely to provide a good approximation, but we can also examine an enormous variety of situations for which no analytical calculations are known to exist. These include systems with multiple aperiodic unit cells, cases in which more than one band is occupied, and cases in which the inter-band transition rate is high.

We find that a rich variety of dynamical phenomena can occur, including not only almost-periodic oscillations, but also high-frequency small-amplitude intra-well oscillations, and unbounded acceleration. The precise blend of these basic dynamical components depends on the form of the impurity potential, $U(z)$, the miniband structure of the superlattice potential, $V(z)$, the strength of the electric field, F , and the detailed form of the initial wave function, $\psi(z, 0)$.

Of particular interest for experiments is the regime dominated by almost-periodic oscillations, which is characterized by the following conditions: (1) The energy gap, E_g , separating any initially occupied miniband of the periodic superlattice Hamiltonian,

$H_0 = T + V(z)$, from the next higher band must be large compared to eFa , and (2) the energy width, W , of the occupied miniband must be comparable to or greater than twice eFa . In this regime, in the single-impurity case, if the initial wave function occupies a relatively narrow miniband (~ 10 - 20 meV), the dynamical behavior is almost-periodic. (With multiple impurities, and impurities with width greater than a single unit cell, the behavior is qualitatively similar, although without corresponding analytical results we cannot confirm that the behavior can be characterized as almost-periodic.) We conjecture that such almost-periodic oscillations could give rise to the rapid dephasing observed in DFWM experiments.^{8,9} By contrast, if the initial wave function occupies a relatively wide miniband (~ 100 - 200 meV), we find that the wave function typically undergoes oscillations which are very nearly time-periodic with the Bloch period for several (~ 5 - 10) periods before giving way to almost-periodic oscillations.

In recent experiments,^{8,9} electrons were selectively injected into the narrow (~ 20 meV) lowest miniband of the superlattice. We conjecture that if the experiments were instead performed on a superlattice which supports relatively wide minibands, for example, a short-period superlattice (e.g., $a \sim 50$, where the band width can be ~ 150 meV), Bloch oscillations should be detected for several periods before the dephasing becomes evident. Such an experiment may provide information regarding the cause of the rapid dephasing seen in recent experiments, and perhaps more importantly, it may provide an opportunity to observe Bloch oscillations for significantly longer times than have been seen before.^{8,9}

In the following section, the elements of our theory are presented. In Sec. III we present an overview of our results and discuss the conditions under which each dynamical phenomenon is dominant. In Sec. IV we focus on the regime of greatest interest for experiments, in which and almost-periodic oscillations are dominant. A summary and conclusions are given in Sec. V.

II. THEORY

A. Numerical Method

A detailed discussion of our numerical technique has been presented elsewhere.⁷ Here we will give only a brief outline. To solve the TDSE, $i\hbar \partial\psi/\partial t = H\psi$, we utilize a modified Cayley method,^{13,14} in which the wave function evolves in time according to

$$\psi(t + \Delta t) = U_C(\Delta t) \psi(t)$$

where the strictly unitary Cayley operator^{13,14}

$$U_C(\Delta t) = \left(1 + \frac{i\Delta t}{2\hbar} H\right)^{-1} \left(1 - \frac{i\Delta t}{2\hbar} H\right), \quad (3)$$

is used as an approximation to the exact evolution operator, $U(\Delta t) = \exp(-iH\Delta t/\hbar)$, with an error which is only of order $(\Delta t)^3$. Thus if we choose Δt to be sufficiently small, the error introduced in using the Cayley operator (3) rather than the exact evolution operator is exceedingly small and accumulates over time extremely slowly.

To monitor the dynamical evolution of the system we obtain a sequence of snapshots of the wave function, at uniformly spaced times, $t_k = k \Delta t$, ($k = 0, 1, 2, \dots$). The system is described at the time t_k by a column vector, $\Psi(t_k)$, whose elements are the instantaneous values of the electron wave function for the uniformly spaced set of spatial mesh points $z_n = n \Delta z$, ($n = 1, \dots, N$). The choices $\Delta z = a/200$ and $\Delta t = \tau_B/2000$ generally ensure accurate results for time intervals up to $50\tau_B$. We employ "hard wall" boundary conditions by requiring that the wave function vanish at the ends of the system. We then choose N sufficiently large that the wave function never comes close to the boundaries for the duration of the simulation. (Typically N was chosen in the range 5×10^3 to 10^5 .)

B. Superlattice and Impurity Models

We consider electrons in a GaAs/Al_xGa_{1-x}As superlattice which we model in the effective mass approximation by a one-dimensional square-well/square-barrier potential of the form $V(z) = 0, (|z| \leq w/2); = V_0, (w/2 < z < b + w/2); V(z+a) = V(z)$, where V_0 depends on Al concentration x . Here b and w are the barrier (Al_xGa_{1-x}As) and well (GaAs) widths, respectively, and the superlattice period is $a = b + w$. The effective mass is piecewise constant with a value depending on the Al concentration x in each layer. We remark that we utilize the kinetic energy operator of the form $T = -(\hbar^2/2) (\partial/\partial z) \{ [1/m^*(z)] \partial/\partial z \}$, so as to accommodate a position-dependent effective mass, $m^*(z)$.

The impurity potential, $U(z)$, we model analogously to the superlattice potential. The magnitude of $U(z)$ depends on the Al concentration at z (see Fig. 1). In addition, we modify $m^*(z)$ to correspond to the total Al concentration implied by the total aperiodic superlattice potential $\mathcal{V}(z) = V(z) + U(z)$.

C. Matrix Elements for Comparison to the SBTB Theory

The regime likely to be of greatest interest for experiments is the almost-periodic oscillation regime. Where appropriate we will utilize results from a single-impurity SBTB model^{10,11} of the TDSE to guide our investigation of this regime. There are two matrix elements important for making contact with that theory, one associated with the periodic superlattice in a uniform electric field, described by the Hamiltonian, $H_1 = T + V(z) + eFz$, the other associated with the aperiodicity, $U(z)$.

We utilize the set of Wannier functions, $\{|nJ\rangle\}$, to compute the matrix elements. These are defined by

$$|nJ\rangle = \phi_1(z - na) = \frac{a}{2\pi} \int_{-\pi/a}^{\pi/a} dq e^{-inqa} \Phi_{1,q}(z), \quad (4)$$

where $\Phi_{l,q}(z)$ is the Bloch eigenfunction associated with the band l and wave vector q of the periodic superlattice Hamiltonian

$$H_0 = T + V(z) \quad (5)$$

and normalized according to

$$\int_0^a dz |\Phi_{l,q}(z)|^2 = 1.$$

The function $|n,l\rangle$ is localized about the n th site with a width on the order of $a/2$, and the set of functions $|n,l\rangle$, including $l = 0, 1, 2, \dots$ and $n = -\infty, \dots, \infty$, form a complete orthonormal set. Details regarding the calculation of the Wannier functions, as well as plots of several of them as a function of z , are given in Ref. 7. Here we only point out a few salient features.

First, for a given superlattice, the Wannier function $|n',l\rangle$ is obtained from $|n,l\rangle$ by a spatial translation through a distance $(n' - n)a$. Secondly, Wannier functions associated with different bands l and l' of the same superlattice are *very* dissimilar, since the Bloch eigenfunctions of different minibands are so dissimilar. (See Fig. 1 of Ref. 7.) Finally, Wannier functions associated with the same band index l and the same site n in *different* superlattices can be quite dissimilar, again through the Bloch eigenfunctions of the different periodic potentials. (See Fig. 1 of Ref. 7.)

One matrix element important for the SBTB model^{10,11} is defined by

$$V_1 = \langle 0,0 | H_0 | 1,0 \rangle, \quad (6)$$

where H_0 is the field- and impurity-free Hamiltonian defined in (3). In a tight-binding system, the magnitude of V_1 is proportional to the energy width of band $l = 0$. It can also be thought of as a measure of how localized the Wannier functions are about their central site. Typically if the magnitude of V_1 is small, then the Wannier functions are highly localized, to within one or two unit cells of their respective centers. If V_1 is larger, then the Wannier functions are more diffuse and can extend over many unit cells. From V_1 we define the dimensionless parameter

$$\beta = \frac{2V_1}{eFa}, \quad (7)$$

which is equal to the parameter β in Ref. 11.

The other relevant matrix element is defined for the special class of single-impurity potentials, i.e., where the aperiodic potential $U(z)$ is nonzero only within the single unit cell about $n=0$:

$$U_{00} = \langle 0,0 | U | 0,0 \rangle. \quad (8)$$

U_{00} is a measure of the strength of the impurity. As shown below and in Ref. 11, if the impurity-free system sustains Bloch oscillations, then for small values of U_{00} , the electron wave function still exhibits oscillations which closely approximate Bloch oscillations for several periods before giving way to almost-periodic oscillations. If U_{00} is large, however, the wave function is sufficiently disturbed by the aperiodicity that the almost-periodic oscillations are evident within the first one or two oscillations. From U_{00} we define the dimensionless parameter

$$\xi = \frac{U_{00}}{eFa}, \quad (9)$$

which is identical to the parameter ξ in Ref. 11.

By computing the values of β and ξ , we use the SBTB theory to predict the qualitative dynamical behavior of the wave function in a superlattice with a single impurity at site $n=0$. However, the agreement will not be exact, because in the numerical calculations we employ the complete Hamiltonian, H , given by (1), rather than a truncated version as in the SBTB theory.^{10,11} In addition, in the numerical work we utilize a position-dependent effective mass function, whereas in the SBTB treatment a uniform mass was used. However, in cases where the initial wave function can be represented by a linear combination of Wannier functions of a single band characterized by the inequality $E_g \gg eFa$, the SBTB model should provide reasonable qualitative predictions of the dynamical behavior.⁷ It should be stressed, however, that in cases where the aperiodic potential extends over more than one unit cell, the initial wave

function occupies more than one band, or the band gaps are not large compared to eFa , our numerical approach allows us to obtain results for which no known analytical theory is available.

D. Initial Wave Function

In aperiodic superlattices, as well as periodic superlattices,⁷ the dependence of the electron dynamics on the initial wave function can be characterized by two properties: (i) the number of contiguous superlattice sites with significant initial electron probability density, and (ii) the properties of the minibands initially occupied.

As discussed in Ref. 7, in the case of a periodic superlattice, the *specific* shape of the initial wave function, e.g. whether it is a Gaussian, or a Lorentzian, or any other localized function, is *not so important*. What is important is how many contiguous wells are occupied, and the properties of the minibands which are occupied.

Because each of the complete orthonormal set of Wannier functions $\{|n,l\rangle\}$ associated with a given superlattice can easily be identified with a particular miniband and a particular lattice site, they make up an ideal basis from which to construct initial states. In this work, we choose as initial states specific linear combinations of Wannier functions, thereby specifying which bands are occupied and which sites have large probability density initially. Thus, we can systematically explore the dependence of the dynamics on the properties of each miniband and on the site occupancy.

III. OVERVIEW OF RESULTS

A. Results and Discussion

In periodic semiconductor superlattices subject to an electric field [$U(z) = 0$], the miniband structure of the periodic superlattice Hamiltonian, $H_0 = T + V(z)$, strongly influences which of the basic dynamical elements, Bloch oscillations, intra-well oscillations, or acceleration, will dominate the dynamical behavior.⁷ If the energy gap, E_g , separating any occupied miniband from the next higher miniband is large compared to eFa , then the interband transition rate^{2,7} is low and Bloch oscillations are the dominant phenomenon. The amplitude of the oscillation is proportional to $(a/2)[W/(eFa)]$, where W is the energy width of the occupied miniband.⁷ If more than one miniband is occupied, then high-frequency small-amplitude intra-well oscillations can also occur, whose amplitude increases with increasing field strength and whose frequency is determined approximately by the difference between the mean energy of each of the occupied minibands.⁷ If eFa is comparable to or greater than the band gap separating an occupied miniband from the next higher miniband, the interband transition rate is high and acceleration can dominate the dynamical behavior.⁷

As we will presently show, in aperiodic superlattices, the observed dynamical phenomena include almost-periodic oscillations, intra-well oscillations, and acceleration. The conditions for each of these dynamical phenomena to occur are analogous to those described above for the periodic superlattice. If the energy gap, E_g , separating an occupied miniband from the next higher miniband is large compared to eFa , the dominant phenomenon is almost-periodic oscillations, which we will show are closely related to the Bloch oscillations which would occur in the absence of any aperiodicity. If more than one miniband is occupied, intra-well oscillations occur, and if $E_g \lesssim eFa$, acceleration effects dominate.

We first consider a superlattice with well and barrier widths $w = 95\text{\AA}$ and $b = 25\text{\AA}$, respectively ($a = 120\text{\AA}$), and with Al concentration $x = 0.3$. For this system the potential barrier height is $V_0 = 243\text{ meV}$, and the effective mass has the value $0.067 m_e$ in the GaAs layers and $0.092 m_e$ in the $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ layers, where m_e is the bare electron mass. The energy minima and maxima for several of the lowest minibands of this superlattice are shown in Table I, along with the associated band widths and gaps. We choose a field strength of $F = 2.5\text{ kV/cm}$, so that $eFa = 3\text{ meV}$, and $\tau_B = 1.38\text{ psec}$. Note that the band gaps shown in Table I are large compared to eFa , so that the inter-band transition rate can be expected to be very low.^{2,7}

We choose an initial state designed to provide a reasonable approximation to the probability distribution of electrons in experiments where electrons are selectively injected into a particular miniband, often by means of a tuned laser excitation from the valence band. We assume that the initial probability density is large in the quantum wells and small in the barriers, and that there is roughly equal probability in each of several contiguous wells. Such initial

TABLE I Minimum and maximum energy values, E_{min} and E_{max} , respectively, for miniband of band index l of a GaAs/ $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ superlattice with $w = 95\text{ \AA}$ and $b = 25\text{ \AA}$. Also included are the miniband width, W , and the energy gap, E_g , between minibands. All energy values are given in meV.

l	E_{min}	E_{max}	W	E_g
0	27	38	11	72
1	110	153	43	93
2	246	346	100	85
3	431	619	188	

states are conveniently described by a linear combination of Wannier functions, $|n,l\rangle$, associated with a particular band l . As an example, we choose an equally weighted sum of Wannier functions associated with six contiguous wells for the lowest ($l = 0$) miniband, $\psi(z,0) = 6^{-1/2} \sum_n |n,0\rangle$, $n = -2, \dots, 3$. (Similar results are obtained for other linear combinations of two or more contiguous wells. For an initial state consisting of only a single Wannier function, the dynamical behavior is qualitatively quite similar, but the electron motion typically is localized to a smaller region of space.) This superlattice and initial state are identical to those of Ref. 7, Fig. 2, which shows that (in the absence of any impurities) Bloch oscillations occur with no observable deterioration for as many as 25 Bloch periods.

We introduce a single impure layer, by adding 0.5% Al in the $n = 0$ GaAs well. The impurity potential is $U(z) = 4.05 \text{ meV}$ ($|z| \leq w/2$); 0 ($|z| > w/2$), and the effective mass in the region $|z| \leq w/2$ is $0.0674 m_e$. The total potential energy function, $V(z) + U(z) + eFz$, is shown in Fig. 2(a) along with the initial probability density, $|\psi(z,0)|^2$.

The subsequent dynamical behavior, as obtained from the numerical solution of the exact TDSE, is shown in Fig. 2(b), where the probability density, $|\psi(z,t)|^2$, is plotted as a function of z and t/τ_B . The impurity serves to disturb the wave function such that the Bloch oscillations are supplanted by a behavior which is no longer time-periodic.

Fig. 3 shows the position expectation value, $z(t) = \langle \psi(z,t) | z | \psi(z,t) \rangle$, as obtained from the numerical solution of the TDSE [i.e. corresponding to the probability density shown in Fig. 2(b)] and from the SBTB^{10,11} approximation to the TDSE. For the SBTB calculation we have used $\beta = -1.75$ and $\xi = 1.17$, computed from Eq. (7) and (9), respectively. The agreement between the two curves is quite good. The differences can be attributed to the fact that in the numerical calculation the effective mass differs from layer to layer, whereas in the SBTB calculation, a single effective mass is utilized. Thus, the exact dynamical behavior can reasonably be described by the SBTB model, and according to the

Fig. 2 (a) Total potential energy, $V(z) + U(z) + eFz$, (right ordinate) for a GaAs/ $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ superlattice with $w = 95 \text{ \AA}$, $b = 25 \text{ \AA}$, and $F = 2.5 \text{ kV/cm}$, and 0.5% Al contamination in the GaAs layer centered at $z = 0$. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to six contiguous Wannier functions of the lowest field-free miniband. (b) Probability density as a function of z and t/τ_B for the initial state and potential energy shown in (a).

Fig. 3 Position expectation value, $z(t)$, versus t/τ_B as computed from the numerical solution of the exact TDSE (solid curve) and from the SBTB model (dashed curve). For the SBTB calculation we have used $\beta = -1.75$ and $\xi = 1.17$.

results of that model,^{10,11} can be termed almost-periodic.¹² A detailed discussion of the physics of this phenomenon has been provided in Ref. 11.

We now consider a different initial wave function, the dynamical behavior of which can no longer be described by the SBTB model. It consists of six equally weighted Wannier functions centered at contiguous sites from each of the two lowest minibands, $\psi(z,0) = (1/\sqrt{12}) \sum_n (|n,0\rangle + |n,1\rangle)$, where the sum extends over $n = -2, \dots, 3$. We use the same superlattice, impurity, and electric field strength as in Fig. 2.

The probability density as a function of z and t/τ_B is shown in Fig. 4, in which three distinct dynamical phenomena are identifiable. First, a localized portion of the wave packet (yellow in color) between approximately $z = 50$ and $z = -30$ nm exhibits almost-periodic oscillations very similar to those seen in Fig. 2(b). This dynamical phenomenon can be attributed to the portion of the wave function associated with the lowest miniband. Secondly, a portion of the wave function (dark green) appears to be undergoing Bloch oscillations between approximately $z = 0$ and $z = 200$ nm, with a second weaker component in the opposite

Fig. 4 Probability density as a function of z and t/τ_B for the superlattice, impurity and field strength used in Fig. 2. The initial wave function consists of six contiguous Wannier functions from the lowest two minibands.

direction, between $z = 0$ and -200 nm. As shown below, this behavior is associated with the $l = 1$ miniband. The third phenomenon is the high-frequency small-amplitude intra-well oscillations which are evident only in the orange and yellow regions near $z = 0$.

The intra-well oscillations are a manifestation of the coupling between the two bound states of a single well, due to the electric field.⁷ The frequency of the oscillations is given by $\nu = (E_1 - E_0)/h$, where E_1 and E_0 are energy values approximately in the middle of the $l = 1$ and $l = 0$ minibands, respectively. The amplitude of the oscillations is only large enough to be observable when more than one bound band is occupied at time $t = 0$. This phenomenon is discussed in greater detail in Ref. 7.

We now consider an initial state consisting of six contiguous wells associated with the $l = 1$ miniband, $\psi(z, 0) = 6^{-1/2} \sum_n |n, 1\rangle$, $n = -2, \dots, 3$. Fig. 5 shows the probability density as a function of z and t/τ_B . We can clearly identify the large amplitude Bloch oscillation-like behavior in Fig. 4 with the virtually identical behavior in Fig. 5.

Fig. 5 Probability density as a function of z and t/τ_B for the superlattice, impurity and field strength used in Fig. 2. The initial wave function consists of six contiguous Wannier functions from the first excited miniband.

Close examination of these oscillations reveals that they are *not* exactly periodic Bloch oscillations, as they of course cannot be, since the impure superlattice is not periodic. They are actually almost-periodic oscillations, as in Fig. 2. However, in the present case of an electron in the $l = 1$ miniband, the impurity appears to have a much less dramatic effect on the electron dynamics than it does in the $l = 0$ miniband. As shown below, we find it to be common that the dynamical behavior of electrons in an excited miniband more closely resembles Bloch oscillations for a longer time than electrons in the lowest miniband in the same system.

In an experiment, for example a DFWM experiment^{8,9} or an experiment to detect the radiation emitted from an oscillating electron,¹⁵ we conjecture that, in the absence of other effects, such as electron-phonon interactions, which were not included in the independent-electron Hamiltonian we have employed, the behavior in Fig. 5 would be observed initially

as Bloch oscillations for several (~ 5 -10) periods, which then give way to almost-periodic oscillations. In contrast, the behavior in Fig. 2 would be observed as at most one or two Bloch oscillations which almost immediately become almost-periodic. In principle, then, one could perform experiments based on electrons injected into the first excited miniband of the superlattice in order to observe more Bloch-like oscillations than have been seen before.

It is likely, however, that in experiments based on electrons injected into an excited miniband, electron-phonon interactions actually are very important, and may allow a very fast relaxation channel from the first excited to the lowest miniband, with scattering times less than a single Bloch period even at very low temperatures.¹⁵ Hence, an experiment based on electrons in the first excited miniband is probably unfeasible for the detection of the oscillations of the type shown in Fig. 5.

However, as demonstrated in Ref. 11, the feature of the first excited miniband which is important for allowing the Bloch-like oscillations to occur before the almost-periodic oscillations are evident is the relatively large band width (or equivalently, the relatively diffuse Wannier functions associated with the wide miniband). One can engineer a superlattice band structure such that a wave function undergoes Bloch-like oscillations, as in Fig. 5, with the wave function in the *lowest* miniband, so that phonon relaxation is not an important factor. Experiments performed on such systems may provide the opportunity to detect more Bloch-like oscillations than have been detected in the experiments which have been reported to date,^{8,9,16} which have all been based on superlattices with a relatively narrow lowest miniband. These issues are discussed further in Sec. IV.

In all of the above results, the band gap, E_g , separating any occupied miniband from the next higher miniband was large compared to eFa , and therefore the interband transition rate was low.^{2,7} In fact, projections of the wave function onto Wannier functions of each miniband confirm that in all three cases (Fig. 2, 3, and 4), the probability initially associated with each

miniband did not change by more than 0.1% for the entire duration of the simulation. Now we consider a case in which the band gap is comparable to, or even less than, eFa .

We consider a superlattice with $w = 200 \text{ \AA}$, $b = 5 \text{ \AA}$ ($a = 205 \text{ \AA}$), and $x = 0.2$. The barrier height is $V_0 = 162 \text{ meV}$, and the effective mass is $0.067 m_e$ in the GaAs layers and $0.084 m_e$ in the $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers. The energy minima, maxima, widths, and gaps associated with the lowest four minibands are given in Table. II. We will again introduce 0.5% Al contamination in the $n = 0$ nominally GaAs layer, so that $U(z) = 4.05 \text{ meV}$ ($|z| \leq w/2$); 0 ($|z| > w/2$), and the effective mass for $|z| \leq w/2$ is $0.0674 m_e$. We employ an electric field strength $F = 3.9 \text{ kV/cm}$ ($eFa = 8 \text{ meV}$ and $\tau_B = 0.52 \text{ psec}$).

We choose an initial wave function again consisting of six continuous Wannier functions of the lowest miniband of this superlattice $\psi(z,0) = 6^{-1/2} \sum_n |n,0\rangle$, $n = -2, \dots, 3$. (Qualitatively similar results are obtained for any linear combination of Wannier functions of the same miniband.) Note that these Wannier functions are different from those used above, because we are considering a different superlattice. To compare Wannier functions of the two

TABLE II Minimum and maximum energy values, E_{min} and E_{max} , respectively, for miniband of band index l of a GaAs/ $\text{Al}_{0.2}\text{Ga}_{0.8}\text{As}$ superlattice with $w = 200 \text{ \AA}$ and $b = 5 \text{ \AA}$. Also included are the miniband widths, W , and the energy gap, E_g , between minibands. All energy values are given in meV.

l	E_{min}	E_{max}	W	E_g
0	3	13	10	7
1	20	53	33	28
2	61	119	58	69
3	127	212	85	

superlattices, see Fig. 1 of Ref. 7. The initial probability density and total potential energy are shown in Fig. 6(a).

Fig. 6(b) shows the probability density as a function of z and t/τ_B . Although a portion of the wave function appears to undergo almost-periodic oscillations near $z = 0$, a significant probability density is simply accelerated anti-parallel to the electric field. The leading edge of the accelerating portion is traveling in approximately a parabolic trajectory, analogous to a classical uniformly accelerated particle. It is as if the energy barriers were essentially transparent to the accelerating portion of the wave function.

Fig. 7 shows the projection of the wave function on Wannier functions of each band, $P_l(t)$, defined by

$$P_l(t) = \sum_{n=-\infty}^{\infty} |\langle \psi(t) | n, l \rangle|^2, \quad (10)$$

for each of the four lowest minibands. Also shown is the sum of $P_l(t)$ for $l = 0, \dots, 3$. At time $t = 0$, Fig. 7 shows that the wave function is occupying the lowest miniband only, since it was constructed explicitly of Wannier functions of that miniband. However, as time goes on, the wave function is seen to cascade from the $l = 0$ miniband to the $l = 1$, then the $l = 2$, the $l = 3$, and so on. Thus, in a superlattice in which $E_g \lesssim eFa$, Zener tunneling² occurs from one energy band to the next, and the real-space manifestation of this is the unbounded acceleration of a portion of the wave packet anti-parallel to the electric field.⁷

B. Summary

In Sec. III, we have provided the solution of the exact TDSE based on the complete Hamiltonian, H , for an electron in a superlattice with an aperiodic potential term which is nonzero only in a single unit cell, in the presence of a uniform electric field. We have shown

Fig. 6 (a) Total potential energy, $V(z) + U(z) + eFz$, (right ordinate) for a GaAs/ $\text{Al}_{0.2}\text{Ga}_{0.7}\text{As}$ superlattice with $w = 200 \text{ \AA}$, $b = 5 \text{ \AA}$, and $F = 3.9 \text{ kV/cm}$, and 0.5% Al contamination in the GaAs layer centered at $z = 0$. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to six contiguous Wannier functions of the lowest field-free miniband. (b) Probability density as a function of z and t/τ_B for the initial state and potential energy shown in (a).

Fig. 7 Projection, $P_l(t)$, of the wave function onto Wannier functions of bands $l = 0$ (open circles), $l = 1$ (open diamonds), $l = 2$ (open triangles), $l = 3$ (open squares), and the sum of the projections onto bands $l = 0, 1, 2$, and 3 (solid circles), as a function of t/τ_B .

that a number of different dynamical phenomena can occur, including almost-periodic oscillations, intra-well oscillations, and acceleration.

The conditions for each of these phenomena to occur are analogous to those for the periodic superlattice in an electric field.⁷ When $E_g \gg eFa$, the Zener tunneling rate is low.^{2,7} In a periodic superlattice under this condition, the electron exhibits time-periodic Bloch oscillations. In the presence of an impurity, however, the strictly periodic Bloch oscillations are replaced by almost-periodic oscillations. If the influence of the aperiodic term in the Hamiltonian is weak, for example, when the wave function occupies a miniband with a large energy width, the wave packet undergoes oscillations which closely resemble Bloch oscillations for several periods before the almost-periodic behavior is clearly evident. In both periodic and aperiodic superlattices, if more than one band is initially occupied, intra-well

oscillations can also occur. If however, $E_g \lesssim eFa$, then the Zener tunneling rate can be very high, and a large portion of the wave packet undergoes unbounded acceleration anti-parallel to the field.

In recent experiments,^{8,9,16} anywhere from one to eight Bloch oscillations were observed in semiconductor superlattices under the condition $E_g \gg eFa$. In each experiment, however, the detected signal was not exactly time-periodic, but decayed rather rapidly over time. We suggest that static impurities could give rise to the decaying Bloch oscillation-like signal observed in experiments. The regime where almost-periodic oscillations are the dominant dynamical phenomenon, and where Bloch oscillations are dominant in the corresponding periodic superlattice, is of greatest interest to relate to experiments on Bloch oscillations, and is therefore worthy of a more thorough investigation. This is the subject of the next section.

IV. ALMOST-PERIODIC OSCILLATION REGIME

In this section, we consider a superlattice and field strength in which Bloch oscillations would be the dominant phenomenon if no aperiodicity were present ($E_g \gg eFa$ and $W > 2eFa$ for any occupied miniband)⁷. We will consider different types of impurities, and initial states associated with different minibands of the superlattice. Recall that β and ξ , which are the two important quantities for determining the dynamical behavior in the SBTB model,^{10,11} depend on the particular periodic potential, $V(z)$, the associated Wannier functions, $|n,l\rangle$, and the particular aperiodic potential, $U(z)$, via Eq. (6)-(9). Thus, as we modify the aperiodic potential, or choose initial states associated with different bands (and therefore consider Wannier functions associated with different l), we will examine how the values of β and ξ , and the corresponding dynamical behavior, are affected. We will then relate our findings to the experimental detection of these dynamical phenomena.

A. Single Impurity

We begin by presenting a more comprehensive picture of our results for a single impurity, for which we have the SBTB theory^{10,11} as a guide. According to that theory, an electron in a single-impurity aperiodic lattice potential subject to a uniform electric field exhibits almost-periodic¹² oscillations. If the impurity is weak ($\xi \ll \beta$ or $\xi \ll 1$), the dynamical behavior looks much like Bloch oscillations for several periods before the almost-periodic behavior is strongly evident.

We summarize here our findings for two very different systems. One is the superlattice used for Figs. 2-4, in which $w = 95 \text{ \AA}$, $b = 25 \text{ \AA}$, and $x = 0.3$, with a fixed field strength $F = 2.5 \text{ kV/cm}$ ($eFa = 3 \text{ meV}$). We will henceforth refer to this system as the 120- $\text{\AA}$ superlattice, since $a = 120 \text{ \AA}$. The second is a superlattice with $w = 40 \text{ \AA}$, $b = 10 \text{ \AA}$, and $x = 0.3$, with a

fixed field strength $F = 16$ kV/cm ($eFa = 8$ meV). We will consider initial states constructed from Wannier functions of either the lowest or the first excited miniband. Specifically, we choose the form $\psi(z,0) = 6^{-1/2} \sum_n |n,0\rangle$, where the sum is taken over sites $n = -2, \dots, 3$, (Qualitatively similar results are obtained for different linear combinations of Wannier functions of the same band.) This system we will refer to as the 50-Å superlattice. The energy minima and maxima of the lowest three minibands are shown in Table III, along with the corresponding energy widths and gap. Note that the width of the lowest miniband of the 50-Å superlattice is substantially larger than that of the 120-Å superlattice (see Table I), and also that the band gap separating the lowest from the first excited miniband is large compared to $eFa = 8$ meV.

TABLE III Minimum and maximum energy values, E_{min} and E_{max} , respectively, for the miniband of band index l of a GaAs/Al_{0.3}Ga_{0.7}As superlattice with $w = 40$ Å and $b = 10$ Å. Also included are the miniband widths, W , and the energy gap, E_g , between minibands. All energy values are given in meV.

l	E_{min}	E_{max}	W	E_g
0	43	201	158	111
1	312	817	505	151
2	968	1871	903	

We will consider initial states constructed from Wannier functions of either the lowest ($l = 0$) miniband of each superlattice. Specifically, we choose the form $\psi(z,0) = 6^{-1/2} \sum_n |n,0\rangle$, where the sum is over sites $n = -2, \dots, 3$. (Qualitatively similar results are obtained for different linear combinations of Wannier functions of the same band.)

Utilizing Wannier functions of the lowest ($l = 0$) miniband of the 120-Å superlattice, we find that $\beta = -1.76$. Table IV shows the values of ξ for the different types of impurities

Table IV Values of ξ for single impurities in the 120-Å superlattice, in the form of: (left) an inexact barrier composition with a difference δx between nominal and actual Al concentration, (center) an interface translated by a distance δz , and (right) a nominally GaAs well contaminated by an amount δx of Al.

barrier		interface		well	
δx	ξ	δz	ξ	δx	ξ
0.10 %	4.91E-3	-5.0 Å	1.41	0.10 %	0.258
0.25 %	1.23E-2	-2.5 Å	0.484	0.25 %	0.646
0.50 %	2.46E-2	2.5 Å	-0.506	0.50 %	1.29
1.00 %	4.91E-2	5.0 Å	-0.742	1.00 %	2.58

Table V Values of ξ for single impurities in the 50-Å superlattice, in the form of: (left) an inexact barrier composition with a difference δx between nominal and actual Al concentration, (center) an interface translated by a distance δz , and (right) a nominally GaAs well contaminated by an amount δx of Al.

barrier		interface		well	
δx	ξ	δz	ξ	δx	ξ
0.10 %	5.75E-3	-5.0 Å	2.09	0.10 %	0.0843
0.25 %	1.44E-2	-2.5 Å	0.935	0.25 %	0.211
0.50 %	2.87E-2	2.5 Å	-0.692	0.50 %	0.422
1.00 %	5.75E-2	5.0 Å	-1.17	1.00 %	0.843

shown in Fig. 1(b)-(d). Similarly, for the 50-Å superlattice, we obtain $\beta = -9.12$, and the analogous values of ξ are given in Table V.

First consider an impurity of the type shown in Fig. 1(c), in which a single barrier (AlGaAs) layer has an inexact composition, with a difference δx between the actual Al concentration and the nominal Al concentration. According to Table IV, for $\delta x \leq 1.0\%$, ξ is quite small compared to unity. This is understandable, since the Wannier function $|0,0\rangle$ for the 120-Å superlattice is sharply peaked in the quantum well and is very low in the region of the barrier (see Fig. 1(a) of Ref. 7). The overlap of $|0,0\rangle$ with $U(z)$ is small, and therefore ξ is small. According to the SBTB model, since $\xi \ll 1$, we should expect the dynamical behavior to resemble Bloch oscillations for several periods before the almost-periodic behavior becomes strongly evident.

For the same type of impurity we see that the values of ξ for the 50-Å superlattice (Table V) are substantially larger than the corresponding values for the 120-Å superlattice. This is because the Wannier function $|0,0\rangle$ for the 50-Å superlattice is considerably more diffuse than that for the 120-Å superlattice, so that the overlap with $U(z)$ is greater. However, for the 50-Å system, ξ is small compared to unity and very small compared to the magnitude of β , so we should expect the behavior to closely resemble Bloch oscillations for several periods in this case as well, before the almost-periodic behavior is evident.

Fig. 8 shows $z(t)$ as a function of t/τ_B for $\delta x = 1.0\%$, as computed from the numerical solution of the exact TDSE. The solid (dashed) curve corresponds to the 120-Å (50-Å) superlattice. According to Fig. 8, even for a barrier composition in error by as much as 1.0%, the dynamical behavior is very nearly Bloch oscillatory for as many as ten Bloch periods. However, the amplitude of both curves is seen to decay slightly with time, and the period is not exactly τ_B .

Fig. 8 Position expectation value $z(t)$ as a function of t/τ_B in the case of an inexact barrier Al concentration differing from the nominal Al concentration by $\delta x = 1.0\%$. The solid (dashed) curve corresponds to the 120-Å (50-Å) superlattice.

These results are easily understood intuitively. Since the electron probability density is very low in the barrier regions at all times (see Fig. 2-4 and Ref. 7), an impurity in a barrier has little ability to disturb the oscillatory behavior of the electron wave function. Hence, the departure from Bloch oscillations develops quite slowly. For values of δx less than 1.0%, the departure from Bloch oscillations is even less noticeable than that shown in Fig. 8.

Now consider an impurity of the type shown in Fig. 1(d), in which a single interface is translated by a distance δz . Shown in Table III (IV) are the values of ξ associated with the 120-Å (50-Å) superlattice for δz corresponding to approximately ± 1 and ± 2 monolayers, where we take a monolayer thickness to be approximately 2.5 Å, to use a round figure. None of the values of ξ for either superlattice is small compared to unity for any of the listed values

of δz . This is because $U(z)$ has such a large value, 243 meV, so that the matrix element U_{00} is large. In addition, none of the values of ξ in Table IV, corresponding to the 120-Å superlattice, is small compared to the magnitude of $\beta = -1.76$. This suggests that for wave functions in this system the almost-periodic behavior will be evident almost immediately. In Table V, for the 50-Å superlattice, however, we see that for $\delta z = \pm 2.5$ Å, the magnitude of ξ is considerably smaller than the magnitude of $\beta = -9.12$, so that we expect the wave function to undergo perhaps a few oscillations of approximately the Bloch frequency, before giving way to almost-periodic oscillations.

Shown in Fig. 9 are $z(t)$ as a function of t/τ_B for $\delta z = 2.5$ Å. The solid (dashed) curve corresponds to a wave function associated with the 120-Å (50-Å) superlattice. As anticipated the curve for the 120-Å superlattice exhibits almost-periodic oscillations almost

Fig. 9 Position expectation value $z(t)$ as a function of t/τ_B in the case of a single interface translated by a distance $\delta z = 2.5$ Å. The solid (dashed) curve corresponds to the 120-Å (50-Å) superlattice.

immediately. In contrast, the curve associated with the 50-Å superlattice has the appearance of Bloch oscillations with a decaying amplitude for about 5 periods, then almost-periodic oscillations.

The third type of impurity we will consider here which is restricted to a single unit cell is shown in Fig. 1(b), where a single semiconductor layer which is nominally GaAs is contaminated with a small concentration δx of Al. Tables IV and V indicate that this type of impurity is stronger than an error in barrier composition but weaker than an translation of an interface. For all values of δx considered, the value of ξ associated with the 50-Å superlattice is slightly smaller than that for the 120-Å superlattice. This is because the Wannier function $|0,0\rangle$ associated with the 50-Å system extends over a greater spatial range than that corresponding to the 120-Å system, and it therefore has a smaller overlap with the impurity in the quantum well region. None of the values of ξ for either superlattice are very small compared to unity. For the 120-Å system, only the values of ξ corresponding to $\delta x = 0.1\%$ and 0.25% are somewhat small compared to the magnitude of β . Thus for electrons in the 120-Å superlattice, we could expect almost-periodic oscillations immediately for $\delta x = 0.5\%$ and 1.0% , but several Bloch-like oscillations which then give way to almost-periodic oscillations for $\delta x = 0.1\%$ and 0.25% . In Table V, however, ξ is fairly small compared to β for all values of δx shown, so several Bloch-like oscillations should be exhibited by electrons in the 50-Å superlattice before the almost-periodic behavior becomes evident.

The results for $z(t)$ as a function of t/τ_B are shown in Fig. 10. Panel (a), (b), and (c) correspond to Al contamination of $\delta x = 0.1\%$, 0.5% , and 1.0% , respectively, and in each panel, the solid (dashed) curve corresponds to wave functions in the 120-Å (50-Å) superlattice. As the amount of Al contamination is increased, the almost-periodic oscillations becomes evident earlier in both curves. However, even in the case of 1.0% contamination, where the wave function in the 120-Å superlattice exhibits almost-periodic oscillations virtually

Fig. 10 Position expectation value $z(t)$ as a function of t/τ_B in the case of a single well contaminated by (a) 0.1 %, (b) 0.5 %, and (c) 1.0% Al. In each panel, the solid (dashed) curve corresponds to 120-Å (50Å) superlattice.

immediately, in the 50-Å superlattice the wave function exhibits Bloch-like oscillations with decaying amplitude for a few periods before the definite onset of almost-periodic oscillations.

One very important point should be made about Figs. 6-8. In all of the impurity cases considered, the wave function associated with the wide-miniband (50-Å) superlattice exhibits oscillations which resemble Bloch oscillations for many more periods than the wave function associated with the narrow-miniband (120-Å) superlattice.

Recent experiments^{8,9,16} to observe Bloch oscillations have been based on electrons selectively injected into the lowest miniband of the superlattice conduction band, with a miniband width on the order of 10 meV. In those experiments,^{8,9,16} anywhere from one to eight oscillations were observed, but in all three experiments,^{8,9,16} the detected signal was strongly damped over time. The DFWM technique^{8,9} probes the extent to which the original form of the wave function is maintained. Thus, if the electrons in the sample lose coherence so that the original wave function is not recovered after each Bloch time, τ_B , then the expected photon echo peaks,¹⁷ which are measured in the experiment, will decay over time. In the measurements of the electromagnetic transients,¹⁶ the detected radiation field strength should essentially map out the position expectation value of the wave packet.¹⁸ Hence, if the electron wave packet undergoes Bloch-like oscillations with decreasing amplitude which then give way to almost-periodic oscillations, as, for example, in the solid curve of Fig. 10(a), it is likely that the detected signal in either type of experiment would appear to be damped Bloch oscillations for several periods.

We conjecture that almost-periodic oscillations (or approximate Bloch oscillations) associated with aperiodicity in the sample could give rise to the signal decay observed in experiments.^{8,9,16} Such aperiodicity could arise, for example, from imperfections introduced either intentionally or unintentionally during crystal growth. We further propose that experiments based on superlattices with a *wide* lowest miniband may provide an opportunity to observe

Bloch-like oscillations for significantly longer times than have been seen before, in narrow-miniband superlattices.

In the next section, we explore other aperiodicity configurations which extend over several unit cells of the superlattice. We will show that although we cannot confirm that the dynamical behavior is specifically almost-periodic, because we do not have an analytical theory describing multiple impurities which is analogous to the single-impurity SBTB theory,^{10,11} qualitatively the dynamical behavior is very similar to the single-impurity case. In particular, wave functions in a wide-miniband superlattice are typically less disturbed by the aperiodicity than in a narrow-miniband superlattice and therefore our above proposal for an experiment in a wide-miniband superlattice remains pertinent.

B. Multiple Impurities

In the previous section, we showed that in cases where the aperiodic potential is nonzero in only a single unit cell, electrons in wide-miniband superlattices would exhibit behavior much more closely resembling Bloch oscillations than electrons in narrow-miniband superlattices. However, a single-impurity system is a very special case, which we have used as a starting point because of the availability of analytical work on the single-impurity problem.^{10,11} It is more likely, rather, that there are weak impurities distributed randomly throughout the system. In a future article,¹⁹ we will explore the effects of a large number of randomly distributed impurities. Here we restrict our investigation to systematic impurities of the type discussed in the previous section, but placed in more than one unit cell.

Since the amplitude of the motion of electrons in the 50-Å superlattice is so large, the likelihood of encountering additional impurities is enhanced relative to an electron in the lowest miniband. The question arises whether the additional impurities encountered by the wave function in the 50-Å superlattice disturb it enough that the dynamical behavior is actually *less*

like Bloch oscillations than the wave function in the 120-Å superlattice, in contrast to our earlier findings for a single impurity.

As an example, we consider the same 50-Å superlattice ($w = 40$ Å, $b = 10$ Å, $x = 0.3$) and field strengths ($F = 16$ kV/cm) as used for Figs. 8-10. Now we introduce *two* 0.5% Al contaminated wells, one at site $n = 0$ ($|z| \leq w/2$) and one at site $n = -10$ ($|z - 500 \text{ Å}| \leq w/2$). We choose initial states of the form $\psi(z, 0) = 6^{-1/2} \sum_n |n, 0\rangle$, where the sum extends over sites $n = -2, \dots, 3$. (Other linear combinations of contiguous Wannier functions of a single band l give qualitatively similar results.) From Figs. 8-10, we see that the motion in the 50-Å superlattice is such that electrons are likely to scatter from the contaminated well centered about $z = 500$ Å in addition to the one centered about $z = 0$.

Shown in Fig. 11 are $z(t)$ versus t/τ_B for this two-impurity system (solid curve) and for the corresponding system with a single impurity at $n = 0$ (dashed curve), which was also shown in Fig. 10(b). Without an analytical theory for multiple impurities, we cannot confirm whether the dynamical behavior is specifically almost-periodic,¹² but the amplitude and frequency of the oscillations in Fig. 11 are qualitatively similar to the almost-periodic oscillations in Fig. 10(b). If we compare directly the results of Fig. 11 with those of Fig. 10(b), we see that the second impurity does indeed disturb the wave function, but only slightly.

An important point to note in Fig. 11 is that even though the scattering from the additional impurity results in the Bloch-like oscillations giving way to almost-periodic oscillations much sooner than with only a single impurity, the 50-Å superlattice still supports more damped Bloch oscillations than the 120-Å superlattice. Thus, even in the presence of multiple impurities, it appears likely that experiments based on wide-miniband superlattices should still provide the opportunity to observe more Bloch-like oscillations than have been observed in narrow-miniband superlattices.

Fig. 11 Position expectation value $z(t)$ as a function of t/τ_B in the case of two impurities: the $n = 0$ well and the $n = 7$ well each contaminated by 0.5% Al. The solid (dashed) curve corresponds to the 120-Å (50-Å) superlattice.

C. Impurity Length Scale

Thus far we have only considered impurities of a fixed length scale: a barrier with an inexact composition for its entire width, a GaAs quantum well with Al contamination for its entire width, and an interface translated by an entire monolayer, where the barrier width, well width, and monolayer thickness are fixed. Here we examine the sensitivity of the dynamical behavior to the length scale of the impurity. We show that the spatial width of the impurity relative to the relevant lengths of the superlattice, such as w , b , and a , strongly influences whether almost-periodic oscillations are evident immediately, or whether some number of Bloch-like oscillations are exhibited before the almost-periodic behavior is evident. In addition, we show that the conclusions drawn above regarding the behavior of electrons in

wide-miniband superlattices compared to narrow-miniband superlattices are not unique to the types and sizes of impurities considered above. They are valid for other types of impurities and length scales as well.

We choose an impurity in the form of a Gaussian distribution of Al differing from the nominal Al concentration by

$$\delta x = x_0 \exp \left[-(\ln 2)(z - z_0)^2 / \sigma^2 \right], \quad (11)$$

where x_0 is the maximum error in Al concentration, z_0 is the center of the distribution, and σ is the full width at half maximum (FWHM). [In Eq. (11), $\ln$ signifies the natural logarithm, not a product of the indices l and n .] By using such a form for the impurity, we can conveniently vary the FWHM, σ , to examine the dependence of the dynamical behavior on the length scale of the impurity.

We will again use both the 120-Å ($w = 95$ Å, $b = 25$ Å, and $x = 0.3$) and 50-Å ($w = 40$ Å, $b = 10$ Å, $x = 0.3$) superlattices. We choose $F = 2.5$ kV/cm and 16 kV/cm, respectively, and an initial wave function of the form $\psi(z, 0) = 6^{-1/2} \sum_n |n, 0\rangle$, $n = -2, \dots, 3$. Fig. 12 shows the initial probability density for the $l = 0$ initial wave function along with the total potential energy $V(z) + U(z) + eFz$ in the case of the 120-Å superlattice for an impurity of the form (11) with $x_0 = 0.5\%$, $z_0 = 0$, and $\sigma = 50$ Å. The slight bowing of the potential energy due to the Gaussian distribution of Al contamination is evident in Fig. 12.

Fig. 13 shows $z(t)$ versus t/τ_B for wave functions in the 120-Å superlattice (solid curves of each panel) and in the 50-Å superlattice (dashed curves), for an impurity with $x_0 = 0.5\%$, $z_0 = 0$, and for several different choices of σ . In Fig. 13(a), corresponding to $\sigma = 5$ Å, the impurity is fairly small, so that electrons in either superlattice undergo Bloch-like oscillations, for four or so oscillations in the 120-Å superlattice, and for at least ten in the 50-Å superlattice, before giving way to almost-periodic oscillations. In panel (b) and (c), for which $\sigma = 50$ Å and 250 Å, respectively, the impurity is sufficiently large that the wave function in the 120-Å superlattice exhibits almost-periodic oscillations immediately, while that in the 50-Å

Fig. 12 (a) Total potential energy, $V(z) + U(z) + eFz$, (right ordinate) for a GaAs/ $\text{Al}_{0.3}\text{Ga}_{0.7}\text{As}$ superlattice with $w = 95 \text{ \AA}$, $b = 25 \text{ \AA}$, and $F = 2.5 \text{ kV/cm}$, and a Gaussian impurity with $x_0 = 0.5\%$, $z_0 = 0$, and $\sigma = 50 \text{ \AA}$. Initial electron probability density, $|\psi(z, 0)|^2$ (left ordinate), corresponding to a six contiguous Wannier functions of the lowest field-free miniband.

superlattice exhibits Bloch-like oscillations which give way to almost-periodic oscillations earlier as σ is increased. In panels (d) and (e), corresponding to $\sigma = 500 \text{ \AA}$ and $1000 \text{ \AA}$, respectively, the Bloch-like oscillations for many periods are recovered, i.e., the "lifetime" of the Bloch-like oscillations *increases* with increasing σ . This is because as σ becomes large, the potential energy $U(z)$ due to the impurity varies so slowly with position that the force, $-dU/dz$, to disturb the Bloch oscillation-like behavior becomes quite small.

It should be noted, however, that the results in Fig. 13, as in all of the other data presented above, support the conclusion that electrons in wide-miniband superlattices undergo

(a)

(b)

(c)

Fig. 13 Position expectation value $z(t)$ as a function of t/τ_B for wave functions in the 120-Å superlattice (solid curve in each panel) and in the 50-Å superlattice (dashed curve), for a Gaussian impurity with $x_0 = 0.5\%$, $z_0 = 0$, and $\sigma = 5$ Å (a), 50 Å (b), 250 Å (c), 500 Å (d), and 1000 Å (e).

Bloch-like oscillations of a longer lifetime before giving way to almost-periodic oscillations than electrons in narrow-miniband superlattices.

V. SUMMARY

In this article, we have presented the dynamical evolution of the electron wave function in aperiodic semiconductor superlattices, subject to a uniform external electric field. Our approach consists of solving the TDSE based on the complete Hamiltonian, H , using high-accuracy numerical methods. The electron wave function can exhibit a rich blend of different dynamical phenomena, including almost-periodic oscillations,^{10,11} intra-well oscillations, and unbounded acceleration. The precise mixture of these basic dynamical elements depends on the miniband structure of the periodic potential, the form, strength, and length scale of the impurity, the electric field strength, and the detailed form of the initial wave function.

When the band gap, E_g , above any occupied miniband is large compared to eFa , we find that almost-periodic oscillations dominate the dynamical behavior. If more than one band is occupied, still with $E_g \gg eFa$, intra-well oscillations also occur. If, by contrast, $E_g \lesssim eFa$, a large portion of the wave function is accelerated anti-parallel to the electric field.

In recent experiments^{8,9,16} based on electrons injected into the narrow (~ 10 meV) lowest miniband of a superlattice, one to eight Bloch oscillations were observed, but the detected signal decayed rapidly with time. We speculate that static impurities could give rise to the observed^{8,9,16} damped Bloch oscillatory behavior.

Our results indicate that electrons occupying a wide miniband of an aperiodic superlattice typically undergo oscillations which closely resemble Bloch oscillations for many more periods before giving way to almost-periodic oscillations than electrons in a narrow miniband. We propose that an experiment based on superlattices with a wide lowest miniband, such as the 50-Å superlattice studied here, may elucidate the question as to the nature of the rapid signal decay observed in previous experiments.^{8,9,16} More importantly, it may provide

an opportunity to observe Bloch-like oscillations for many more periods than have ever been seen before.

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GENERAL CONCLUSION

In this work we have determined the exact time-dependent behavior of independent electrons in both periodic and aperiodic semiconductor superlattices subject to a uniform electric field. We have done so by solving the time-dependent Schrödinger equation based on the complete independent-electron Hamiltonian, with no approximations, using high-accuracy numerical techniques. In addition, we have undertaken a complementary investigation by analytical methods, based on a single-band tight-binding approach to the time-dependent Schrödinger equation. In situations where the electron wave packet is effectively confined to a single band, the single-band tight-binding treatment provides a reasonable approximation to the exact problem and is valuable for the interpretation of the exact numerical results. In cases where the single-band approximation is not valid, however, our numerical approach provides a unique opportunity to explore the rich variety of dynamical phenomena which are inaccessible to investigation by analytical methods.

We have shown that long-lived Bloch oscillations are a real component of the exact dynamics of Wannier-Stark electrons in periodic superlattices of the GaAs/Al_xGa_{1-x}As system. They are the dominant phenomenon when the following conditions are satisfied: (1) The band gap, E_g , separating any occupied miniband from the next higher miniband is large compared to eFa , so that the inter-band transition rate is very low,²⁻⁶ and (2) the width, W , of the occupied miniband is comparable to or greater than twice eFa . We find that the amplitude and frequency of the Bloch oscillations are such that the emitted terahertz radiation is detectable by existing methods. Indeed, very recent experiments by Waschke *et al.*⁶⁴ directly detected coherent radiation originating from Bloch oscillations in such superlattices.

Under different conditions, however, the electron wave function can exhibit other dynamical phenomena which can coexist with, or even mask, the Bloch oscillations. These

include intra-well oscillations and acceleration, the quantum analogs of the classical behavior of a charged particle in a periodic potential and a uniform electric field. The acceleration is a manifestation of the cascade of probability from one miniband to the next. This occurs when E_g is comparable to or less than eFa , so that the interband transition rate is high.²⁻⁶ The intra-well oscillations are a manifestation of the coupling between bound states of a single quantum well due to the electric field. These oscillations have an observable amplitude only when the inter-band transition rate is low and when more than one bound band is occupied initially.

In aperiodic superlattices, a similarly rich variety of dynamical phenomena occur. The conditions for acceleration and intra-well oscillations are essentially the same as in ideal superlattices. Under conditions where Bloch oscillations are dominant in ideal superlattices, electrons in superlattices with a single impurity undergo almost-periodic⁶⁶ oscillations with amplitude and frequency on the same order of magnitude as the Bloch oscillations. In superlattices with multiple impurities, we have no analytical theory with which to compare the numerical results, but the dynamical behavior is qualitatively similar to the almost-periodic oscillations seen in superlattices with a single impurity.

There are a number of important remarks to be made about the behavior of electrons in aperiodic superlattices. First, almost-periodic oscillations due to weak impurities could give rise to the signal decay observed in recent degenerate four-wave mixing experiments.⁵⁹⁻⁶¹ Secondly, since the frequency and amplitude are comparable to those of Bloch oscillations in the corresponding periodic superlattice, the radiation should also be in the terahertz regime. However, it would not be a single frequency, nor would it have constant amplitude. In fact, the radiation detected by Waschke *et al.*⁶⁴ was not periodic. Rather, the observed amplitude decayed over time. The number of oscillations observed in the experiment increased as the field was increased, consistent with the behavior of electrons in aperiodic superlattices of the type considered here.

Thirdly, according to our calculations, an electron wave packet in a first excited miniband of an aperiodic superlattice typically executes oscillations which closely resemble Bloch oscillations for many more periods than a wave packet in the lowest miniband of the same system before giving way to almost-periodic oscillations. Thus, if it is static impurities which are causing the signal decay in both the degenerate four-wave mixing experiments⁵⁹⁻⁶¹ and the radiation experiments,⁶⁴ then experiments based on electrons injected into an *excited* miniband may afford the opportunity to elucidate the nature of the signal decay. In addition, and more importantly, such an experiment may provide the opportunity to observe many more Bloch-like oscillations than have been seen before, a goal which has been sought for over six decades.

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