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Simulational Studies of Epitaxial Semiconductor Superlattices: Quantum Dynamical Phenomena in ac and dc Electric Fields

by

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To my wife Sandra

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ABSTRACT

Using high-accuracy numerical methods we investigate the dynamics of independent electrons in both ideal and realistic superlattices subject to arbitrary ac and/or dc electric fields. For a variety of superlattice potentials, optically excited initial wave packets, and combinations of ac and dc electric fields, we numerically solve the time-dependent Schrödinger equation. In the case of ideal periodic superlattice potentials, we investigate a long list of dynamical phenomena involving multiple miniband transitions and time-dependent electric fields. These include acceleration effects associated with interminiband transitions in strong fields, Zener resonances between minibands, dynamic localization with ac fields, increased single-miniband transport with an auxiliary resonant ac field, and enhanced or suppressed interminiband probability exchange using an auxiliary ac field. For all of the cases studied, the resulting time-dependent wave function is analyzed by projecting the data onto convenient orthonormal bases. This allows a detailed comparison with approximate analytic treatments.

In an effort to explain the rapid decay of experimentally measured Bloch oscillation (BO) signals we incorporate a one-dimensional representation of interface roughness (IR) into our superlattice potential. We show that as a result of IR, the electron dynamics can be characterized in terms of many discrete, incommensurate frequencies near the Bloch frequency. The interference effects associated with these frequencies cause a substantial decrease in amplitude of the signal after several Bloch periods. We suggest that this is an important source of coherence loss in BO signals at low temperature and low carrier density. We also propose an experimental method that should significantly reduce the effects of IR by exciting electrons to only a single layer of the superlattice. This is accomplished by doping

the central GaAs layer with a very small amount $(< 1\%)$ of In, thus reducing the energy gap for this layer. Thus, a laser excitation pulse tuned somewhat below the nominal electron-hole excitation energy, will only excite a few Wannier-Stark eigenstates associated with this In-doped layer. Our numerical simulations show that the THz signal from electrons optically excited using this novel procedure is nearly free from all inhomogeneous broadening associated with IR.

CHAPTER 1. GENERAL INTRODUCTION

I. Dissertation Organization

Section II of this introductory chapter provides relevant background material that brings the reader to date on the recent advances in ultra-fast semiconductor physics, and more specifically, information relating to electron dynamics in superlattices subject to both ac and dc electric fields. This is followed by a short section summarizing the content of the four main chapters of the dissertation. These four chapters, Chapters 2–5, are focused articles exploring interesting dynamical behavior of excited carriers for specific superlattice models and electric fields. Each of these articles either has been published in or will be submitted to a refereed scientific journal. For this reason, each chapter's general layout has the form of an independent article with abstract, body, appendices, and references. A general conclusion is given as the last chapter of the thesis, which is followed by a list of references cited in this introduction and the general conclusion, and finally the acknowledgements.

II. Background Theory and Literature Review

A. Introduction

Recent advances in epitaxial growth techniques, such as Molecular Beam Epitaxy (MBE), are allowing scientists to develop a new generation of ultra-small, ultra-fast semiconductor quantum devices. The potential for such heterostructural devices to replace conventional semiconductor electronics has sparked a world-wide interest in their development. As the name implies, the physical properties of these devices can only be adequately explained in the context of quantum mechanics. The quantum regime becomes important in semiconductor transport as at least one spatial dimension of the heterostructure becomes comparable to the de Broglie wavelength of the electron carriers.

An important class of semiconductor quantum devices is based on a structure known as a "superlattice." A superlattice consists of a periodic array of thin alternating semiconductor layers, where the important feature of the participating semiconductors is their differing energy band gaps. An important class of superlattices is fabricated using alternate layers of GaAs and $Al_xGa_{1-x}As$, where x indicates the fraction of Al. The energy band gaps in the different layers of $GaAs/Al_xGa_{1-x}As$ superlattices differ by $1.247x$ eV. The alternating size of the energy gap between the valence and conduction bands gives rise to a "lattice" potential of barriers and wells along the growth axis (see Fig. 1). Thus, excited carriers moving perpendicular to the plane of the layers are affected by the periodic potential. Similar to the bulk lattice, the "super"-lattice forms its own "mini"-bands. If one treats the underlying atomic potential in the effective-mass approximation, the dynamics of excited carriers are predominantly determined by the miniband structure of the superlattice. Because the period of the superlattice is typically two orders of magnitude larger than the lattice period of the bulk semiconductor constituents, the energy scale of the superlattice minibands is also two orders of magnitude smaller, typically in the tens of meV range.

A static electric field, F, applied normal to the layers of a superlattice, disrupts the periodicity of the superlattice. In the case of a weak field, when interminiband coupling can be ignored, the effect of the electric field is to split each of the energy minibands into a set of uniformly spaced energy levels known as a Wannier-Stark (WS) ladder. The spacing of these energy levels in the conduction band of a superlattice is eFa where a is the period of the superlattice and e is the magnitude of the electron charge. Electron dynamics induced by the external electric field, such as Bloch oscillations, will have characteristic times, τ, on the order of $h/(eFa)$. Thus, for typical electric field strengths $(\sim 10\ \text{kV/cm})$ and superlattice

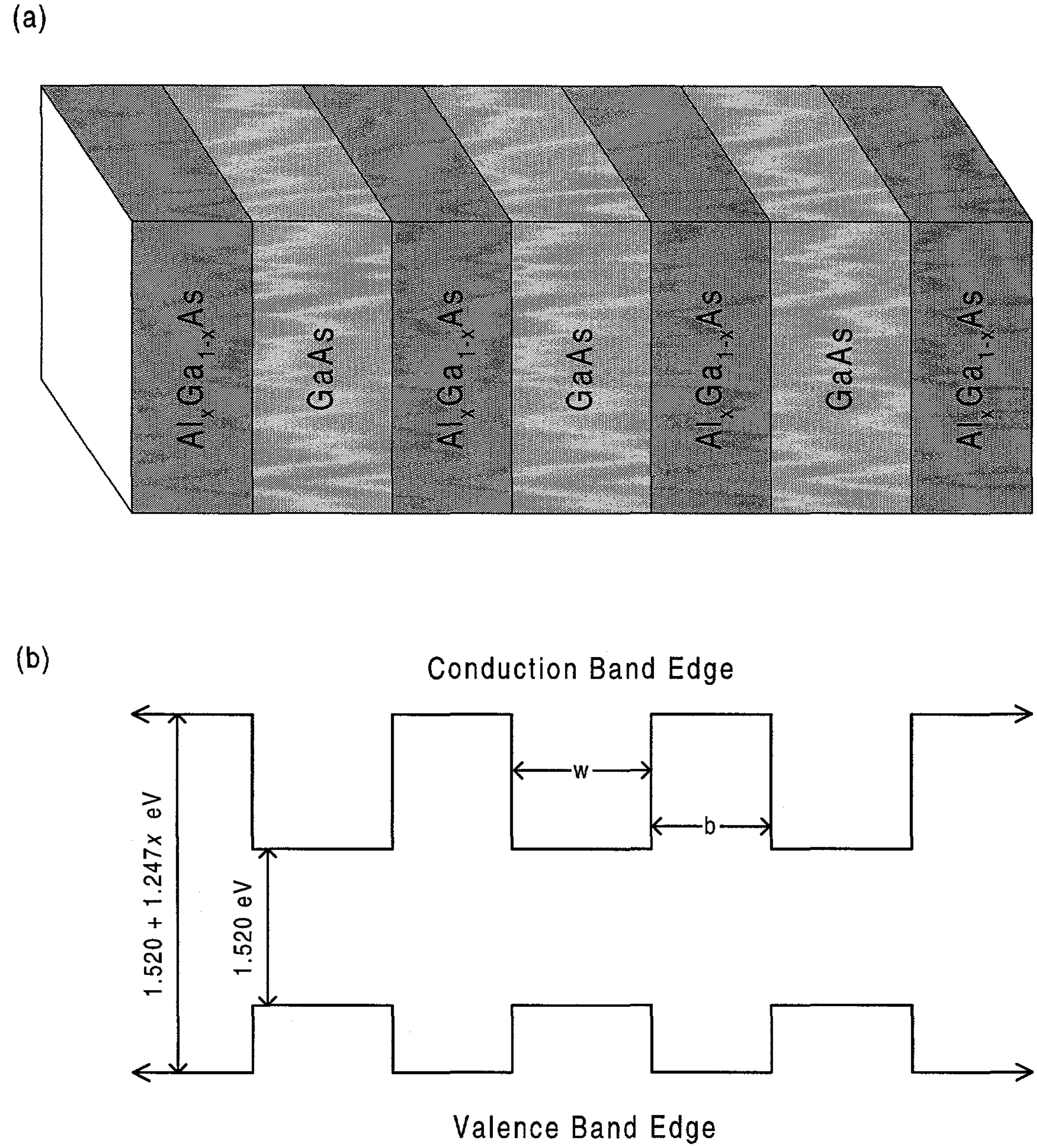

Fig. 1 (a) A schematic diagram showing a semiconductor superlattice made from GaAs and $Al_xGa_{1-x}As$ layers. (b) Valence and conduction energy band edges are shown, indicating differing band gaps for each semiconductor layer. Electrons or holes excited to these carrier bands feel a periodic potential of wells of width w, and barriers of width b.

periods $(\sim 10\ \text{nm})$ used in quantum device designs, the electron dynamics can have sub-picosecond time scales.

Because these ultra-fast times for the electron dynamics are shorter than typical momentum and phase relaxation times for an electron in a semiconductor, the phase information for the wave function describing the electron is retained. This is known as coherent or ballistic transport. In this ultra-fast, ultra-small regime of quantum device physics more traditional approaches based on semiclassical statistical approaches, such as the semiconductor Bloch equations, cannot accurately describe important aspects of the dynamics. For this reason the single-particle time-dependent Schrödinger equation (TDSE) should be employed, and understandably it provides more accurate results.

The extra complications involved in solving a fully quantum-mechanical treatment of electron transport in semiconductor superlattices cause analytic approaches to be very limited, *e.g.* single-band tight-binding models. We have found that solving the time-dependent Schrödinger equation using high-accuracy numerical methods enables one to observe a regime of complex quantum behavior where approximate analytic solutions provide no help. After obtaining quantitative data describing the "true" time-dependent behavior of the electron wave packet, we are able to then work backward, already knowing the results, to make a qualitative analysis of the underlying physics. Also, the quantitative numerical results provide important insights as to which approximations can safely be assumed in an analytic approach, without eliminating important aspects of the dynamics.

Accurate numerical simulations serve not only as an investigatory tool for theoretical understanding, but they also aid experimentalists in the design of working devices. Typically, there are hundreds of different design parameters involved in the engineering of a semiconductor quantum device, each of which subtlety affects the physics involved in the operation of the device. Because of the speed and ease of searching the parameter space using numerical simulations, such efforts can guide experimentalists toward a desired effect quickly and easily. This assistance provided by numerical modeling allows the experimentalist to design working devices more efficiently, circumventing the costly trial-and-error methods commonly used.

Using such a combination of numerical and analytic methods, we have investigated many important issues, all closely related to current experimental work investigating carrier dynamics in superlattices. The first two articles, Chapters 2 and 3 of this thesis, explore a variety of different dynamics possible in ideal periodic superlattices subject to an external electric field. In the first of these two articles, we explore the regime of strong dc electric fields, where we observe acceleration effects associated with massive interminiband transitions. The second article examines four types of dynamical phenomena, which occur for very special choices of superlattice parameters and electric fields. By numerically solving for the time-dependent dynamics, we examine Zener resonances in dc fields, dynamic localization in ac fields, resonant delocalization in a combined ac-dc fields, and the suppression or enhancement of interminiband transitions in strong ac-dc fields. The following two articles, Chapters 4 and 5 of this thesis, deal with the inevitable aperiodicity present in epitaxially grown superlattices and their effect on Bloch oscillations. Chapter 4 suggests that this aperiodicity is responsible for the rapid decay of BO signals observed in experimental samples, and Chapter 5 gives one possible novel solution to the aperiodicity involving localized carrier excitation. Section III of this introduction provides a more detailed summary of these articles.

B. Theoretical studies

1. Bloch oscillations

Although the theoretical development of Bloch oscillations[1] is more than six decades old, only recently has the successful observation of BO[2] within semiconductor superlattices allowed experimental verification and support for theoretical treatments. The early theoretical work involving BO was orphaned from experimental studies, because any possibility of observing such coherent dynamics in bulk semiconductors was superseded by scattering events with characteristics times far shorter than the Bloch period.

Although there have been many conflicting theoretical treatments of BO,[3] the basic one-electron effective-mass model presented in a rather terse article by Houston[4] in 1940 accurately describes the electron dynamics. In that paper Houston formulates the equations of motion for an electron in a periodic field subject to a uniform electric field using the Bloch eigenstates of the field-free superlattice. Alternate derivations of the Houston equations are also given by Krieger and Iafrate[5] and more recently by Rotvig *et al.*[6] Although there have been criticisms[7] of this approach in the past, it is now generally agreed that Houston's approach is valid. In Chapter 3 of this work, we demonstrate the validity of the Houston equations by comparing their numerical solutions with those obtained by solving the TDSE. The Houston equations provide a good conceptual understanding even in the case of high field strengths where multiple minibands are needed to describe the dynamics.

An electron in a superlattice subject to an arbitrary time-dependent electric field is described in the single-particle effective-mass approximation by the following Hamiltonian, $H = T + V(z) + eF(t)z$, where $T = (-\hbar^2/2)\partial/\partial z([1/m^*(z)]\partial/\partial z)$ is the conventional kinetic energy operator, $V(z)$ is the superlattice potential with period a, $F(t)$ is the uniform electric field applied along the growth axis z, and e is the magnitude of the electric charge. One obtains Houston's equations, or the equations of motion in Bloch space, by expanding the time-dependent wave function $\psi(z,t)$ using the complete set of orthonormal Bloch eigenstates of the field-free Hamiltonian, and then substituting this expansion into the TDSE (see Appendix of Chapter 2). The result can be written as

$$i\hbar\frac{\partial}{\partial t}g_l(k,t) = E_l(k)g_l(k,t) + ieF(t)\frac{\partial}{\partial k}g_l(k,t) + eF(t)\sum_{l'}X_{l,l'}(k)g_{l'}(k,t), \qquad (1)$$

where $g_l(k,t)$ are the coefficients of the Bloch eigenstate expansion, $E_l(k)$ the miniband energy eigenvalues, and $X_{l,l'}(k)$ the interminiband coupling operator between minibands l and l' for the wave vector k. One may identify two separate coupling terms in Eq. (1), each proportional to the electric field $F(t)$. The first is an intraband coupling term, $ieF(t)\partial g_l(k,t)/\partial k$, producing a displacement through Bloch space of the wave packet within its own miniband. The second is the interminiband coupling term, $eF(t)\sum_{l'} X_{l,l'}(k) g_{l'}(k,t)$, which allows the "vertical" exchange of probability among minibands at the same value of the wave vector k.

When the electric field strength is weak enough that the interminiband coupling term can be ignored, Eq. (1) can be used to produce a single-miniband equation of motion for the wave packet in Bloch space:

$$\frac{\partial}{\partial t}\left|g(k,t)\right|^2 = \frac{eF(t)}{\hbar}\frac{\partial}{\partial k}\left|g(k,t)\right|^2. \tag{2}$$

We can easily see from Eq. (2) that, in the single-miniband approximation, the Bloch space representation of the initial wave packet retains its original form and propagates with the same time dependence as the wave-vector,

$$k(t) = k(0) - \frac{e}{\hbar}\int_0^t F(t')\,dt'. \tag{3}$$

This is the same result obtained from a traditional semiclassical treatment of the problem, namely $d/dt(\hbar k) = -eF(t)$. In this single-band treatment, $g(k,t) = g(k+2\pi/a,t)$, so in the case of a static electric field the time evolution of the wave packet is periodic. This periodic motion then manifests itself as Bloch oscillations in configuration space. The Bloch oscillations will usually feature center of mass oscillations. However, for restricted forms of the initial wave packet, the Bloch oscillations can also oscillate symmetrically in a radiation-free symmetric breathing mode.[8]

2. *Zener tunneling*

As we will see in Chapter 2, this simplified single-band picture is greatly modified when multiple minibands participate significantly in the dynamics. There have been attempts to approximate the importance of coupling between minibands by calculating the amount of probability per unit time that "leaks" into the upper bands. This "Zener tunneling probability" was given by C. Zener in 1934.[9] Later, enhancements to Zener's model were made by Houston,[4] Wannier,[11] Keldysh,[12] Kane,[13] and others. Because the interest was typically concerned with the effect of Zener tunneling on BO, most of these treatments assume a low electric field value, so that the probability density slowly "leaks" to the higher bands. At higher fields where the Wannier-Stark ladders originating from many different minibands have become interwoven, one is really concerned with the "near crossing" of many energy levels. A modern paper by Lubin *et al.*[14] gives a good review of Zener's original treatment of crossing energy levels[10] with some added detail to the problem. As we will see in Chapter 3, this "crossing" problem is directly connected with the phenomenon of "Zener resonances."[6,16,25] Most theoretical studies involving crossing energy levels are limited to two-band models. He and Iafrate[15] have produced a multiple-band treatment of the *long-term* Zener tunneling probability using the Wigner-Weisskopf approximation. However, for our work we will be concerned with the short-time behavior, since with experimental measurements, dephasing of coherent carriers typically occurs after only a dozen or so Bloch periods.

Zener[9] approximated the transition probability per unit Bloch period to be:

$$\gamma = \exp\left(-\frac{m^* a^2}{4\hbar^2}\frac{E_g^2}{|eFa|}\right), \tag{4}$$

where E_g is the energy gap separating the two minibands. We have found that Zener's estimate of the tunneling probability provides an accurate estimate of the initial transition from the ground state miniband to the first excited miniband. For this reason, we use the value of γ to give a quantitative meaning to the terms "strong" or "weak" electric fields. However, we find that any subsequent transitions can only be determined by numerically solving the full multiple-miniband problem.

Another phenomena directly related to Zener tunneling occurs when a static electric field causes a WS eigenstate associated with a particular unit cell of the superlattice to be nearly degenerate with a neighboring WS eigenstate of an excited miniband. When this resonance condition is met, the amount of probability exchanged between minibands can reach 100% after many Bloch oscillations, even when γ is small. This increased transition rate for special values of the electric field is closely related to other resonant tunneling situations found in many semiconductor heterostructures. In 1995 we performed extensive studies of "Zener resonances" by examining the probability exchange among minibands by numerically solving the Houston equations (see Chapter 3). Concurrently, a group from the University of Copenhagen, Rotvig *et al.*,[16] published very similar numerical results demonstrating the phenomena. This same group has more recently published a second article[6] giving a good overview of Zener resonances. Their analytic treatment is given using a two-band tight-binding model. Hone and Zhao[25] provide a similar two-band treatment of Zener resonances.

3. Time-dependent electric fields

From examining Eq. (1), we see that a time-dependent field, as opposed to a static field, does not change the form of the Houston equations. There are still two important features, an intraband coupling term that causes the wave vector to propagate as,

$$k(t) = k(0) - (e/\hbar)\int_0^t F(t')\,dt', \tag{5}$$

and an interminiband coupling term that is proportional to the time dependent field. Although the infinite set of coupled equations due not significantly change with the extra complication of a time-dependent field, many new interesting dynamics emerge in superlattices subject to a periodic time-dependent electric field. One such phenomenon, known as "dynamic localization," was first described by Ignatov and Ramanov in 1976.[17] Employing classical kinetic equations, Ignatov and Ramanov found that the current in a superlattice of period a driven by a monochromatic laser field with amplitude F_{ac} and frequency ω became zero when the ratio $eF_{ac}a/(h\omega)$ was equal to a zero of the zero-order Bessel function J_0. Dunlap and Kenkre[18] examined "dynamic localization" in the context of a fully quantum-mechanical model. Using a single-band nearest-neighbor tight-binding model, they demonstrated that the mean-square-displacement of the initially localized particle remains finite at infinite times. They also investigated the influence of scattering, showing its negative influence on dynamic localization.[19] Xian-Geng Zhao,[20] also using a tight-binding model, provides more general results, allowing for ac-dc electric fields and non-nearest neighbor contributions. He shows that strictly speaking ac dynamic localization cannot exist when off-diagonal elements are considered. On the contrary he finds that with ac-dc fields, localization exists in all cases except when $eF_{dc}a\,/\,(\hbar\omega)$ is a rational number. All of these works were done assuming that interminiband coupling was weak, so that a single-band treatment was valid.

Later, Holthaus and Hone[21] rederived, using a different approach, the effect of dynamic localization with an ac field. Utilizing the time periodicity of the Hamiltonian they chose as a solution to the Schrödinger equation a set of Floquet wave functions with "quasienergies" as eigenvalues. These quasienergies are analogous to the quasimomentum, in that they are only defined up to integer multiples of the photon energy $\hbar\omega$. Using such a model they were able to equate the "dynamic localization" phenomena with "miniband collapse," where the quasienergy minibands approached zero-width when the ratio $eF_{ac}a/(h\omega)$ is equal to a zero of J_0. Zhao[22] also extended Holthaus's work by examining the effects of long-range intersite interactions on quasienergy band collapse. His results suggest that with experimental observation one should only see band suppression rather than band collapse. In addition, Zhao *et al.*[23] provide a detailed examination of band collapse within a two-band model subject to ac-dc electric fields. In this work,[23] they provide an exact analytic expression for the energy offset between the two WS ladders in the limit of weak coupling. Hone and Zhao[25] apply this expression to the important case of the Zener resonance crossing points, for it is this energy offset that provides the period of the long term probability exchange between the two minibands. They point out that Zener tunneling will be completely suppressed for a special set of superlattice and electric field parameters that cause both minibands to simultaneously collapse.

Holthaus and Hone[26] also performed studies of multiple quasienergy minibands, demonstrating resonance effects when the Stark-shifted quasiminiband energy difference is an integer multiple of the photon energy. They propose using these resonances to produce desired harmonics of the driving laser frequency.[27] Additional work by Holthaus *et al.*[28] shows, for superlattices subject to ac-dc field, that disorder disrupts the delocalization that occurs when the ac component of the electric field is tuned to the Bloch frequency. They suggest that systematic experimental control of the disorder in a superlattice can be used to study Anderson localization.

4. *Interface roughness*

Because of the importance of the semiconductor Bloch oscillator as viable source of tunable THz radiation, there has been considerable effort in the development of a usable device.[2] At this time, however, there are scattering mechanisms that dephase the coherent oscillations after only, typically, less than ten oscillations. Although of extremely good quality, the interface between each layer of an epitaxially grown superlattice is never perfectly planar. There can be small islands or depressions, usually of one or two monolayers, dispersed randomly along the interface surface. These small uncontrolled imperfections, or interface roughness (IR), disturb the perfect periodicity of the superlattice and thus the uniform spacing of the Wannier-Stark ladder. In addition, the rough surfaces should cause scattering parallel to the surface, thus disrupting vertical transport along the growth direction.

There have been many attempts to model the influence of IR in a variety of semiconductor heterostructures. Most of these focus on single and double barrier tunneling devices[29] and derive the modified transmission curves, typically using a transfer matrix formalism. Dharssi and Butcher,[30] using a simple IR model to calculate scattering rates for use in the Boltzmann transport equations, find that the IR limited mobility is less important than that of phonon scattering at room temperature. Unfortunately, however, any theoretical treatment of IR is very limited, because the exact nature of IR has not yet been fully revealed by experimental measurements.

There are many different experimental methods to examine heterostructure interfaces, each probing at a different length scale. X-ray diffraction, because it probes may layers simultaneously, returns an average effect that IR has on the periodicity of a superlattice, such measurements are useful in obtaining an estimate of the "width" of an interface in the growth direction.[31] High-resolution transmission electron microscope (HREM) images are often used to evaluate the quality of interfaces;[32] however, this method is typically limited by the visual interpretation of the images. Digital pattern recognition techniques used with HREM images have revealed compositional variation on length scales finer than those revealed using optical techniques.[33] The most common technique used to evaluate IR is photoluminescence,[34] where the measured exciton energy, being sensitive to the precise width of the heterostructure layer, probes the interface quality. When the photoluminescence spectra show a clear splitting in the exciton line, this is evident that there are broad monolayer regions causing variation in the widths to the superlattice layers. A broad exciton line indicates a smaller scale roughness since the exciton feels an averaged width variation. The width variation among the superlattice layers can then be estimated from by the energy change of the exciton line spectra. Using a time-resolved four-wave mixing technique, D. Birkedal *et al.*[35] found that split exciton peaks were dominant at short time delays and broad single lines at longer delay times, giving evidence of both large monolayer islands and smaller scale roughness.

C. Recent experimental advances

Over the past thirty years, advances in the field of ultra-fast optical spectroscopy have enabled experimentalists to examine the non-equilibrium, nonlinear, and transport properties of semiconductors and semiconductor nanostructures. The common availability of solid-state optical lasers capable of producing picosecond and femtosecond pulses is providing invaluable information on the non-equilibrium dynamics of excitations in semiconductor devices. As these laser sources become more compact and efficient, their usefulness as a repetitive source of excited carriers within practical devices also becomes possible.

The most common ultra-fast spectroscopy technique is the pump-probe method. In the degenerate form of pump-probe spectroscopy, a single laser pulse is divided into two pulses. The first pulse excites carriers in the semiconductor sample and the second time-delayed pulse can then probe any changes in the carrier distribution through some physical property of the sample (reflectivity, absorption, luminescence, etc.). If one requires probing at an energy value different from the excitation pulse, a second laser source is required for the probe pulse, this is known as non-degenerate pump-probe spectroscopy.

The field of ultra-fast spectroscopy is a fascinating, growing field stretching into many different areas of solid-state physics. An excellent review of current topics can be found in Ref. 36. Here I will outline three techniques that have been important in the measurement of Bloch oscillations in semiconductor superlattices. Following this discussion on the experimental observations of BO, I will summarize the status of experimental studies with time-dependent fields utilizing the Free Electron Laser (FEL).

Esaki and Tsu[37] first suggested that BO should be observable in semiconductor superlattices because the spatial period is typically two orders of magnitude larger than the crystalline period of bulk semiconductors. With the increased quality of modern MBE superlattice samples came the first unambiguous observation of WS ladders in superlattices.[38] Using degenerate four-wave mixing (FWM) pump-probe spectroscopy experiments, Bloch oscillations were first directly measured by Feldmann *et al.*[39] and Leo *et al.*[40] in 1992. Soon after this in 1992 Waschke *et al.*[41] measured directly the THz radiation emitted by the oscillating electric dipole moment associated with BO. A third detection method, transmittive electro-optical sampling (TEOS), was used by Dekorsy *et al.*;[42] this technique has produced BO with the longest duration to date. The authors of Ref. 42 have commented that TEOS and direct THz radiation measurements are sensitive to the dipole oscillation of the excited free electrons, while FWM experiments are dependent on electron-hole or excitonic effects. During simultaneous measurements, FWM signals decay more than twice as fast as TEOS signals. They have attributed this to the differing scattering conditions experienced by the holes involved in the FWM signal versus the scattering experienced by the free electrons alone. More recent experimental measurements,[43] using superlattices characterized by minibands that are narrow relative the exciton binding energy, have shown THz radiation to also be sensitive to excitonic effects.

Further experimental measurements of Bloch oscillations were done using FWM by Leisching *et al.*[44] examining a wide variety of superlattice parameters, electric field strengths, and excitation conditions. BO has been measured at room temperature using TEOS by Dekorsy *et al.*,[45] and Roskos *et al.*[46] measured THz radiation from electrons excited to higher minibands. A good review of the current progress of experimental BO can be found in Ref. 2.

With the advent of the FEL, direct experimental investigation of excited carriers' response to external time-dependent fields in the THz regime has become possible.[47-48]

Results from the UCSB Center for Free-Electron Laser Studies have measured what they describe as an "inverse Bloch oscillator,"[50] which is an increased mobility in a superlattice subject to a dc electric field when an auxiliary ac field is tuned to the Bloch frequency. This phenomenon is directly related to the theoretical calculations of Zhao[20] that show a delocalizing effect when $n\hbar\omega = eF_{dc}a \quad (n = 1, 2, \ldots)$. The Santa Barbara group has also found experimental evidence of ac dynamic localization.[49] Future results from the Santa Barbara facility and similar research centers should provide evidence for many new quantum phenomena in superlattices subject to time-dependent electric fields.

III. Summary of Dissertation

Recent experimental studies of quantum transport in semiconductor superlattices have provided valuable insight concerning previously inaccessible non-equilibrium dynamics. These experimental successes can be partially attributed to advances in epitaxial growth techniques that provide higher quality samples; the availability of ultra-fast optical laser sources for probing non-equilibrium dynamics; and the advent of the free-electron laser, which allows experimentalists to measure a superlattice system's response to high-frequency electric fields.

As a consequence of experimental advances, there has been a growing interest within the theoretical arena as well. There have been investigations of many different types of carrier dynamics possible in superlattices subject to an external uniform electric field. These analytic studies, which are typically limited to single and two-band tight-binding treatments of individual electrons, have successfully identified special system parameters that produce interesting, and perhaps useful, dynamics. We, however, use direct numerical methods to provide the time-dependent wave function for many of these interesting systems without having to use an approximate single-band approach. By first solving the TSDE using high-accuracy numerical methods, we are able to then extract, directly from the time-dependent wave function, detailed quantitative information on any quantity of physical interest, including the center-of-mass motion, or the width parameter (root-mean-squared deviation) of the wave function. Thus, we are able to accurately determine the time-dependent behavior of the wave packet for a variety of system parameters. For all of the articles in this thesis, our results are based on the time-dependent solution of the full quantum-mechanical Hamiltonian of independent electrons without resorting to single-band approximations.

It is often useful to project the time-dependent wave function on to several different orthonormal bases. This procedure yields new insightful features of the dynamics that would otherwise escape notice. We have found, as might be expected, that the Bloch functions of the field-free Hamiltonian provide a useful characterization of the dynamics in the presence of an electric field. For this reason, we have determined that solving the Houston equations[4] (the equations of motion of independent electrons subject to a uniform electric field in the Bloch space representation) provides a convenient and numerically efficient method to examine the complicated time dependence of interminiband transitions in the case of both weak and strong interminiband coupling. (These concepts are clarified in Chapter 2.) By confirming that the results arising from the Houston equations agree to high accuracy with the Bloch eigenstate projections obtained by numerical solutions of the TDSE, we verify both the accuracy of our numerical procedures applied to the TDSE, as well as the basic correctness of the Houston equations.

In Chapter 2 of this thesis, we investigate optically excited electrons in superlattices subject to intense dc electric fields, and we find that the electron dynamics are usefully described in terms of massive interminiband transitions. By numerically solving the TDSE for such systems, we find that the dominant behavior of the electron wave packet is to emit at regular intervals short bursts of highly localized packets which accelerate both parallel and anti-parallel to the electric field. We show, by projecting the time-dependent wave function onto the Bloch eigenstates of the field-free Hamiltonian, that the acceleration behavior is a direct consequence of electron transitions made, upward and downward (in energy), between minibands.

We find that we may describe the motion of the wave packet in the Bloch-space representation, using even for intense dc fields the accepted elementary picture, appropriate for weak fields. The electron wave packet, viewed as travelling through an extended zone scheme in Bloch space, proceeds at a fixed rate, $k(t) = k(0) - eFt/\hbar$. Our results show that even when the electric field gives rise to strong interminiband processes, the underlying equation remains in effect, since any exchange of probability between minibands occurs "vertically" at the same value of *k*. The precise amount of probability exchanged between successive energy minibands has a complex dependency on the strength of the electric field, the explicit form of the miniband energy, and on the specific details of the initial wave function, which makes any qualitative predictions impossible. However, from our accurate numerical methods we are able to precisely calculate quantitative values for the occupancy of each miniband as a function of time.

In Chapter 3 of this thesis, we investigate four interesting dynamical phenomena, which occur for very special choices of superlattice parameters and electric fields. The first topic, addressed in Sec. III of Chapter 3, is that of Zener resonances in dc electric fields. This effect can be qualitatively described as an enhanced exchanged of probability between two minibands caused by the near degeneracy of field-dependent energy levels ("Wannier-Stark levels" to be abbreviated as WS). Often, as with the particular superlattice used in our examination of Zener resonances, the coupling between resonant states is very weak. Therefore, the amount of probability exchanged between minibands will reach 100% only after many Bloch oscillations. So in this regard the phenomenon may seem only academic. However, for superlattices with narrow energy gaps, interminiband coupling is inherently stronger even in the case of moderate electric fields, and Zener resonances can influence the dynamics on a time scale equal to τ_B. But unfortunately, in the case of stronger coupling, the effect is often complicated by the involvement of many different minibands simultaneously. Complicated multiple miniband couplings can shroud the resonance effects that may exist between two specific minibands. Nonetheless, our investigations of how Zener resonances behave with weak interminiband coupling should assist in the more difficult case of strong coupling.

We study Zener resonances by numerically solving the Houston equations for the subset of minibands that participate in the dynamics. Although our studies are still in their preliminary stages, we can already make a few important comments. Firstly, we find that our numerically calculated resonance time periods, T_n (see Sec. III, Chapter 3), differ drastically from estimates made using the expression provided in Ref. 25, the expression being a first order estimate of an exact perturbation expression derived using a two-band tight-binding approach.[24] We also find that the T_n are not sensitive to the inclusion of higher minibands in the calculations. In other words, when we calculate T_n using only two minibands, we obtain approximately the same result as if we included higher minibands. We do find, however, that the higher minibands do slightly perturb the position of the resonance as a function of electric field strength. Of course, we also observe many secondary resonances associated with higher minibands that cannot be accounted for in a two-band approximate treatment. In certain cases, a simultaneous resonance condition is met involving three minibands.

In Sec. IV of Chapter 3, we examine ac dynamic localization. Although theories predicting dynamic localization are over twenty years old,[17] only recently has there been experimental evidence[49] of such an effect. The numerical results presented here are the first direct numerical solutions using the full single-electron Hamiltonian, providing a time-dependent wave function exhibiting dynamic localization. We confirm earlier predictions made using a single-band tight-binding theory,[18] that localization will occur in an ac electric field of the form, $F_{AC}(t) = F_{AC}\cos(\omega t)$, when the ratio $(eF_{ac}a)/(\hbar\omega)$ is a root of the Bessel function J_0. We have found that in the case of an ideal periodic superlattice, using a weak ac electric field strength so that interminiband transitions are negligible, that dynamic localization persists over many periods of the ac field. However, when using a more realistic model of a superlattice, one that incorporates the effects of interface roughness (see Chapter 4), we find that the perturbing effects associated with IR disrupt the dynamic localization after several periods of the ac field. Clearly, the effects of IR will play an important role in the experimental observation of dynamical localization effects. Furthermore, the effects of IR cannot realistically be incorporated within a tight-binding model.

Recent experimental results,[50] provided by the experimental group at Santa Barbara using the FEL, show an increase in the dc conductivity of a superlattice when an auxiliary ac field is tuned to the Bloch frequency or its higher harmonics. The authors of Ref. 50 have called this the "inverse Bloch oscillator effect." Qualitatively the effect can by explained as an increased mobility due to resonant coupling between adjacent WS energy levels mediated by photons associated with the ac field.

In Sec. V of Chapter 3, we have investigated in detail, by numerically solving the TDSE, electron dynamics in superlattices subject to the combined ac-dc electric field given by $F(t) = F_{dc} + F_{ac}\cos(\omega t + \phi)$. In particular we emphasize the special case when $eF_{dc}a = n\hbar\omega$, $(n = 1, 2, \ldots)$, because of its importance to the recent experimental studies[50] mentioned above. As we did with ac dynamic localization, we again chose field strengths so that interminiband transitions are negligible. From the analysis of the numerically calculated time-dependent wave function we have made some important observations for those cases when the resonance condition, $eF_{dc}a = n\hbar\omega$, $(n = 1, 2, \ldots)$, is met. First, the width parameter, $\rho(t) = [\langle z^2\rangle - \langle z\rangle^2]^{1/2}$, has been shown to have a time dependence with an average velocity proportional to $J_n(\beta)$, where $\beta = (eF_{ac}a)/(\hbar\omega)$. Second, the average velocity of the center-of-mass, behaves just as is predicted using a semiclassical approach, $\bar{v} \propto \sin(\beta\sin(\phi) - k_0 a + n\phi) J_n(n, \beta)$, where k_0 is the initial quasimomentum. This is to be expected, first, since we use an initial wave function, which has a representation in Bloch space that is narrow with respect to the width of the Brillouin zone, and, second, single-miniband approximations are also valid.

Because of its importance in regard to the experimental measurement of the "inverse Bloch oscillator effect," we investigate the effect of IR on the delocalization associated with the ac-dc resonance condition. We find, as expected, that the delocalizing effects are weakened by the perturbing effects of IR. However, there is still some spreading of the wave packet, indicating that the resonance condition is still partially satisfied. This would imply that the energy shifts in the WS ladder caused by IR are smaller than the energy width of the resonance condition.

In the final section of Chapter 3, we look at a very different superlattice system, whereas in Chapter 2, interminiband transitions dominate the dynamics. Here we use an auxiliary ac electric field to modulate the strength of the dc coupling between minibands. By changing the phase of the ac field, interminiband transitions may be either enhanced or suppressed. Our intention is to provide a means to control the real-space transport, by controlling the interminiband transitions. A method to conveniently control the transitions among minibands would definitely be useful in future quantum device designs. We have only found partial success using ac fields to modulate the interminiband transitions. This is because there is still a significant coupling that occurs at times when the total electric field is not zero. Perhaps more sophisticated time-dependent fields could provide better modulation of the coupling, resulting in complete control of the miniband occupancies.

The last two articles of this thesis, Chapters 4 and 5, provide an important analysis and a proposed solution to the problem of dephasing in the measured THz signals emitted by experimental Bloch oscillators.[2] We have determined that the small aperiodicities caused by imperfections in the superlattice interfaces perturb the uniform spacing of the Wannier-Stark energy levels. Because conventional optical excitation methods excite electrons within each layer of the superlattice with approximately equal probability, each of the Wannier-Stark eigenstates contributes equally to the dynamics. The quantum oscillations arising from many consecutive energy levels all have slightly different frequencies, which results in a large inhomogeneous broadening of the BO signal. We propose an excitation method, which circumvents the dephasing effects of the interface roughness. This is accomplished by doping the central GaAs layer with a very small amount of In, thus reducing the energy gap for this layer. Thus a laser excitation pulse tuned somewhat below the nominal electron-hole excitation energy will only excite a few Wannier-Stark eigenstates associated with this In-doped layer. Our numerical simulations show that the THz signal resulting from this novel excitation procedure is nearly free from all inhomogeneous broadening associated with IR.

CHAPTER 6. CONCLUSION

Our combined numerical and analytic approach to quantum transport in superlattices, using the single-particle time-dependent Schrödinger equation (TDSE), has provided valuable insight to a variety of important dynamical phenomena. Our results are important both for the development of novel quantum electronic devices and for the general advancement of quantum transport theory.

In the first article, Chapter 2, we analyze the complicated multiple miniband dynamics of excited electrons in a semiconductor superlattice subject to a strong electric field. In these high-field conditions portions of the electron wave packet accelerate, as would a free-particle, anti-parallel to the direction of the electric field. We also find that those portions of the wave packet that do not make a transition to higher minibands reverse their momentum and *decelerate* parallel to the electric field. By projecting the time-dependent wave function, obtained by numerically solving the TDSE, onto the Bloch eigenstates of the field-free superlattice, we are able to provide a quantitative description of the miniband occupancies as a function of time. This analysis shows that each portion of the wave packet, accelerating and decelerating in real space, corresponds to the occupancy of a specific miniband.

Chapter 3 gives a detailed analysis of a variety of superlattice dynamics involving interminiband transitions and time-dependent electric fields. The first topic investigated, Zener resonances, is a type of "resonant tunneling" that occurs in superlattices subject to dc electric fields. By numerically solving the multiple-miniband Houston equations, we provide a detailed, accurate analysis of Zener resonances. We have measured the period of the interminiband oscillations associated with Zener resonances, and we find that our results differ from previous estimates that use an approximate perturbation expression obtained from a single-band tight-binding treatment.[25]

Also in Chapter 3, we examine the effect of dynamic localization, both for pure ac electric fields and for combined ac-dc electric fields. In both cases we have verified that, for low field values, solving the full Hamiltonian gives similar results to single-band tight-binding models. We also provide quantitative measurements of the influence of interface roughness (IR), and we found that the special requirements for dynamic localization and resonant ac-field delocalization are disturbed by the perturbing effects of IR. In our investigation of resonant ac-field delocalization, we find that the average velocity of the center-of-mass motion of the wave packet is dependent on the phase of the ac field relative to position of the initial wave function in Bloch space. This dependency agrees with the expression derived from a semiclassical description of the dynamics.

Finally, in Chapter 3 we describe a novel use of an auxiliary ac electric field to suppress or enhance interminiband transitions in a thin barrier superlattice. We have found that such a method may be used to provide partial control over real-space transport by limiting the exchange of probability to the lowest two minibands.

The last two articles of this work, Chapters 4 and 5, provide an important analysis and a proposed solution for the problem of dephasing of the measured THz signals emitted by experimental Bloch oscillators. We have determined the small aperiodicities caused by imperfections in the superlattice interfaces perturb the uniform spacing of the Wannier-Stark energy levels. Because conventional optical excitation methods excite electrons within each layer of the superlattice with approximately equal probability, each perturbed Wannier-Stark eigenstate contributes equally to the dynamics. The quantum oscillations arising from the many consecutive energy levels each have slightly different frequencies, which gives rise to a large inhomogeneous broadening of the BO signal. We propose an excitation method that circumvents the dephasing effects of the interface roughness. This is accomplished by doping the central GaAs layer with a very small amount of In, thus reducing the energy gap for this layer. Thus a laser excitation pulse tuned somewhat below the nominal electron-hole excitation energy will only excite a few Wannier-Stark eigenstates associated with this In-doped layer. Our numerical simulations show that the THz signal resulting from this novel excitation procedure is nearly free from all inhomogeneous broadening associated with IR.

In this thesis, we have examined a few of the many possible systems, consisting of superlattice structures subject to external electric fields. Taking advantage of our numerical approach, we have gone well beyond previous theoretical approaches. Our methods allow for nearly unlimited combinations of realistic superlattice potentials and arbitrary time-dependent electric fields. In addition, because we are solving the full single-particle Hamiltonian, our results do not suffer from the inaccuracies associated with treatments based on single-miniband tight-binding methods. We believe that our approach to investigating the dynamics of photoexcited carriers in semiconductor heterostructures provides invaluable results for the experimental development of usable semiconductor quantum devices, as well as, allowing for the creative design of completely novel devices.

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