Saddle Points and Stark Ladders: Exact Calculations of Exciton Spectra in Superlattices

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Abstract

A new, exact method for calculating excitonic absorption in superlattices is described. It is used to obtain high resolution spectra showing the saddle point exciton feature near the top of the miniband. The evolution of this feature is followed through a series of structures with increasing miniband width. The Stark ladder of peaks produced by an axial electric field is investigated, and it is shown that for weak fields the line shapes are strongly modified by coupling to continuum states, taking the form of Fano resonances. The calculated spectra, when suitably broadened, are found to be in good agreement with experimental results.

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A proper treatment of excitonic effects is essential to the understanding of the optical properties of semiconductors. However, in superlattices it is difficult to calculate exciton spectra because of the anisotropic, quasi-three dimensional nature of the problem. In this letter, the first high resolution theoretical superlattice absorption spectra are presented, calculated using a new method which generalizes the Green’s function approach of Zimmermann [1] for quantum wells. Particular attention is paid to two aspects of the spectra which historically have been of considerable interest in the theory of semiconductors: the first is the saddle point exciton [2] associated with the top of the miniband, the second is the Fano resonance line shape [3] predicted for Stark ladder peaks when an electric field is applied to the superlattice.

A saddle point is a critical point in the bandstructure at which the sign of the mass is different along different directions. For superlattices, the important region is around the M1 critical point at the top of the miniband, where the axial dispersion has a negative effective mass, while the in-plane mass is positive. The associated excitonic feature is not a sharp peak, but a broad enhancement occurring on the low energy side of the saddle point. There has been a long theoretical interest in the behavior of this feature in bulk semiconductors [2], and some low resolution calculations exist for superlattices [4,5]. In this letter, high resolution spectra are used to trace the development of the superlattice saddle point exciton as the miniband width is increased. It is shown that weak structure occurs in the enhancement, corresponding to excited saddle point states.

A Stark ladder occurs as a result of the localization of miniband states when an axial electric field is applied to a superlattice. The localized states are peaked in individual wells and their energies form a ladder of equally spaced levels, with separation equal to the potential drop across a period of the structure. Transitions between different electron and hole states lead to a series of two dimensional like steps in the absorption spectrum, each with an associated exciton peak. For large fields, these are well defined and adequately treated by previous theoretical methods [4,6]. However, it is shown in this letter that, at low fields, the mixing between the excitons and the surrounding continuum states cannot be ignored.
and leads to Fano resonances \(^3\) with significant widths. The possibility of studying Fano resonances in low dimensional structures has been the subject of much theoretical discussion \(^4\). There has, however, been no clear experimental demonstration of the characteristic spectral line shapes, although time resolved measurements show the existence of homogeneous broadening due to resonant coupling \(^5\). The present calculations suggest that Fano line shapes should be observable in Stark ladder spectra from high quality superlattices.

The problem of the exciton in a superlattice has been considered theoretically by a number of authors, in treatments both with \(^6\) and without \(^7\) an electric field. A number of methods have been used, with good results for the energies of the bound and quasi-bound states. However, the only spectra which are obtained \(^8\) are fairly crude, with the continuum features approximated by sets of discrete peaks which need broadening to give realistic results. Though adequate for comparison with most experimental spectra, the broadening makes it impossible to resolve the detailed structure which is the subject of the present work.

The approach adopted here starts from an effective Hamiltonian which simplifies the exciton problem without loss of accuracy. This is solved numerically using a new method which avoids the problems with resolution. The spectra so obtained are essentially exact, and the calculation remains computationally feasible for arbitrarily small broadening.

The effective Hamiltonian is derived by restricting the states used in constructing the exciton to a single pair of minibands (one each for electron and hole) and diagonalizing the Coulomb interaction and electric field terms exactly within this basis \(^9\). This approximation is usually very good, because the miniband separations in typical superlattices tend to be large compared to the energy scale of the excitonic effects.

For the present purpose, the miniband states can best be described in a representation based on localized Wannier functions \(^10\). The wavefunction for optically active excitons can then be written in the form

\[
\Psi(z_e, z_h, r) = \sum_{n_e, n_h} \psi(n_e - n_h, r) f_e(z_e - n_e d) f_h(z_h - n_h d). \tag{1}
\]
Here, $\psi(n,r)$ is a superlattice scale envelope function which depends only on the separations of the electron and hole: $n = n_e - n_h$ (the number of periods of width $d$) along the growth axis and $r$ in the plane of the wells. $f_e(z_e)$ and $f_h(z_h)$ are the Wannier functions for the pair of minibands under consideration.

In the Wannier representation, the miniband kinetic energy takes the form of a tight-binding type hopping operator:

$$T_{\text{hop}} \psi(n,r) = \sum_i \Delta_i [\psi(n + i, r) + \psi(n - i, r)]$$

(2)

where the hopping terms $\Delta_i$ are obtained from a Fourier expansion of the miniband dispersion. The diagonal matrix element of the Coulomb interaction can be written as an effective potential

$$V_{\text{eff}}(n,r) = -\frac{1}{\varepsilon_r} \int \int dz_e dz_h \frac{|f_e(z_e)|^2 |f_h(z_h)|^2}{\sqrt{(z_e - z_h + nd)^2 + r^2}}.$$  

There are also small off-diagonal matrix elements, but these are always negligible in comparison with $V_{\text{eff}}$ and the superlattice coupling.

The envelope function satisfies a Schrödinger equation with Hamiltonian

$$H_{\text{eff}} = -\frac{1}{2m} \nabla_r^2 + T_{\text{hop}} + V_{\text{eff}}(n, r) + eFnd$$

(4)

with $m$ the in-plane reduced mass and $F$ the electric field. Eq. (4) is, despite its tight binding form, an essentially exact formulation of the superlattice exciton within a single pair of minibands, and is valid for minibands of any width.

Optical spectra are obtained from Eq. (4) using a generalization of the method developed by Zimmermann [1] for quantum well excitons. The absorption coefficient is calculated by taking the limit $r, r' \rightarrow 0$ of a Green’s function $G(n, r; n', r'; E)$ satisfying the inhomogeneous system of equations:

$$[H_{\text{eff}} - E] G(n, r; n', r'; E) = \delta_{n,n'} \delta(r - r')$$

(5)

The Green’s function is calculated from numerical solutions to Eq. (4) which satisfy the boundary conditions at $r \rightarrow 0$ or $r \rightarrow \infty$, using a straightforward generalization of the standard approach for the two point boundary value problem [12].
Figs. 1, 2 show the calculated zero field absorption spectra for a series of superlattices with miniband widths from 0 to 57 meV. The structures considered all have GaAs wells of width $L_w=30$ Å and Al$_{0.35}$Ga$_{0.65}$As barriers with widths $L_b$ ranging from 150 Å to 30 Å.

In Fig. 1 results are shown for the wider barrier structures, where the minibands are narrow, with widths smaller than, or comparable to, the exciton binding energy. As $L_b$ is reduced and the miniband width increases, the most obvious change is a reduction in the binding energy and oscillator strength of the main exciton peak. This corresponds to the ground state wavefunction spreading in the axial direction, becoming more three dimensional in form. Changes also occur in the higher exciton states. In the two dimensional limit, these are in-plane $s$ states ($2s, 3s \ldots$), but for finite miniband widths, an additional, more tightly bound state appears (labeled $a$ in Fig. 1), becoming progressively stronger as the width increases. Though it is not possible to describe this state with exact quantum numbers, its wavefunction contains large contributions from in-plane $1s$ states with electron and hole in adjacent periods, mixed in by the superlattice coupling.

The narrow minibands do not have enough width to support the main saddle point feature. There is, however, some excitonic structure apparent in the continuum. This consists of a series of oscillations just below the M1 critical point, which correspond to the higher states of the saddle point exciton. These oscillations broaden and weaken as the miniband becomes wider and the underlying density of states increases. Some evidence of this type of behavior can been seen in lower resolution calculations, but the oscillations seem to be too weak to be experimentally observable in presently available structures. Predictions of similar oscillations in bulk semiconductors have been made by Baslev.

Fig. 2 corresponds to the narrower barrier structures, where the minibands are wide, with widths larger than the exciton binding energy. As the miniband width increases, the main saddle point exciton feature develops: the shape of the continuum changes from dropping away at the band edge to rising gradually towards a broad peak some way below the M1 critical point. For wider minibands, the overall strength of the saddle point feature becomes greater, at the expense of the bound state, but its increasing width means that the height of
the enhancement decreases. The oscillations associated with the higher states remain, but they too become broader.

Figs. 3, 4 show Stark ladder spectra for a Ga$_{0.47}$In$_{0.53}$As-Al$_{0.24}$Ga$_{0.24}$In$_{0.52}$As superlattice with $L_w=39\,\text{Å}$ and $L_b=46\,\text{Å}$. This structure was chosen because of the availability of experimental absorption spectra [13], which provide a more direct basis for theoretical comparison than the more usual photocurrent spectra. A miniband width of 30 meV was used in the calculations, though this is considerably lower than the value of $\sim 40$ meV obtained from the Kronig-Penney model [13].

In Fig. 3, both experimental and broadened theoretical spectra are shown. The theoretical spectra generally reproduce the experimental results very well, especially on the low energy side. The additional experimental absorption at higher energies is mainly due to the light hole transitions, which are not included in the present theory. The poor agreement at $F=7\,\text{kV/cm}$ is probably a result of the electric field in the experiments being underestimated at low biases, due to incomplete depletion. A much better fit can be obtained by increasing the field in the calculation to $\sim 11\,\text{kV/cm}$.

At zero field, the band edge and saddle point exciton features are apparent, while at high field, the nearly fully localized two dimensional exciton (labeled $n=0$) has the correct position and strength. In between, the transitions to adjacent periods ($n=\pm 1$) can be seen moving to higher and lower energy ($\sim \pm eFd$). This behavior has been discussed by a number of authors [5,6] and further details can be found in the references.

Fig. 4 shows theoretical high resolution spectra for the same structure. The zero field spectrum is similar to those of Fig. 2, though it should be noted that the excitonic features are weaker at a comparable miniband width (see $L_b=40\,\text{Å}$ in Fig. 4), a result of the lower electron mass in GaInAs. For finite fields a considerable amount of structure is apparent in the spectra. At $F=14\,\text{kV/cm}$, the $n=0,\pm 1,\pm 2$ excitons are clearly distinguishable, along with peaks due to the higher in-plane states at the same axial separation. The main peaks all have a finite width, which at $\sim 1\,\text{meV}$ is much larger than the numerical broadening. The line shapes of these peaks are characteristic of Fano resonances [3], rising up above the
continuum on one side, but falling well below it, almost to zero, on the other.

The broadened Fano line shapes are a result of the coupling, by the Coulomb interaction, between the main exciton peaks and resonant continuum states from lower energy transitions. This is a purely excitonic effect, which occurs because the Coulomb term allows tunneling between states of different in-plane momentum. It should be distinguished from the lifetime broadening of Stark localized states due to Zener tunneling between minibands [14], which is spectrally insignificant in the field range of interest here, contributing only \( \sim 10^{-6} \) meV to the line widths at \( F = 28 \) kV/cm.

At fields below 14kV/cm, the peaks move closer together and the Fano line widths become larger, since the coupling is strongest when the wave-vectors of the resonant continuum states are small. It becomes less appropriate to think in terms of two dimensional excitons associated with localized Stark ladder states; it is better to consider the applied electric field as perturbing the exciton spectrum, giving it a periodic structure with period \( \sim eFd \), but with an envelope very similar to the zero field shape. At \( F = 7 \) kV/cm, the structure still takes the form of recognizable Fano resonance line shapes, but with all the strength in the region of the band edge and saddle point exciton features. When \( F \) is reduced to 3.5 kV/cm, the width of the resonances becomes large compared to their mutual separation, and they interfere. The resulting spectrum, though basically periodic, contains very complicated structure with unusual line shapes. Calculations at lower field show that the structure becomes finer in scale, but remains present even for \( F < 1 \) kV/cm.

To conclude, a new method for solving coupled exciton problems has been described, and applied to obtain very high resolution spectra for saddle point and Stark ladder features. This has introduced new physics, in the form of additional structure in the spectra and novel peak line shapes. Though the weaker structure is probably experimentally inaccessible, it seems likely that small improvements in the quality of presently available superlattices would allow the observation of excited saddle point exciton states and the Fano line shapes of Stark ladder peaks.
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FIGURES

FIG. 1. Exciton spectra for GaAs-AlGaAs superlattices with narrow minibands, of widths less than or comparable to the exciton binding energy. For each structure, three spectra shown with different magnifications and numerical broadening of 2 meV (lower), 1 meV (middle) and 0.1 meV (upper). M0 and M1 are the critical points at the top and bottom of the miniband.

FIG. 2. Exciton spectra for GaAs-AlGaAs superlattices with wide minibands. For each structure, two spectra are shown with different magnifications and broadening of 2 meV (lower) and 0.2 meV (upper). The emergence of the broad saddle point exciton feature (SPE) below the top of the miniband (M1) is clear.

FIG. 3. Comparison of theoretical (solid line) results with experimental (dashed line) Stark ladder spectra for the AlGaInAs-GaInAs structure of Bleuse et al. The theoretical results have been broadened by 10 meV. The same normalizing factor is used at all fields.

FIG. 4. High resolution results corresponding to the low field spectra of Fig. 3. The line width is 0.2 meV, except for the dashed part of the zero field spectrum which has been broadened to 2 meV.
