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Electronic states in $GaAs/Al_{0.3}Ga_{0.7}As$ non-periodic superlattices

Marilyn Usher and Radha Ranganathan

Department of Physics and Astronomy, California State University, Northridge CA 91330, USA

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Abstract. Transmission and photoluminescence measurements in $GaAs/Al_{0.3}As_{0.7}As$ superlattices in which the layer widths vary randomly, illustrate the effects of one-dimensional non-periodic potentials on electronic energy states. The fundamental interband transition energy in the non-periodic structures is lower than that in the periodic case. A relation between this shift and the wavefunction localization length is derived. The PL emission from the non-periodic samples is ten to twenty times stronger than from the periodic samples.

1. Introduction

Non-periodic or disordered potentials affect physical phenomena of deep significance. The properties of periodic Hamiltonians are well understood and the one-electron Schrödinger equation with a periodic potential has served usefully as a mathematical model for perfect crystals. The problem becomes intricate when deviations from periodicity are introduced. In its most general form, the Schrödinger equation for a particle of mass m in a one-dimensional potential, $V_n(z - z_n)$, reads

$$\frac{\hbar^2}{2m}\frac{d^2\psi}{dz^2} + V_n(z - z_n) = E\psi. \tag{1}$$

In the periodic case, $V_n(z - z_n)$ is centred about equally spaced points with identical shape and strength, yielding solutions of the Bloch form for the wavefunction. A non-crystalline or amorphous material, on the other hand, would be modelled by a non-periodic potential. A random potential is obtained if either or both V_n (strength) and z_n (position) in equation (1) are randomly distributed. This is a problem of some prominence, and commands attention in many fields of mathematics, physics and engineering [1]. An area of intense research activity is that of conductivity in a random potential distribution [2, 3]. A profound effect of disorder is the localization of some of the electronic states which gives rise to scaling laws for the conductance quite unlike that of Ohm's law [4]. For one-dimensional disorder however, all states are localized. The localization length has been shown to be of the order of the mean free path (MFP) [2, 5]. The scaling laws would be particularly relevant in this situation because the Ohm's law behaviour is invalid for all sample sizes greater or smaller than the MFP [4]. Samples with controlled and intentionally introduced disorder are desirable for studying the various aspects of localization. The controlled growth of epitaxially layered materials like the semiconductor heterostructure have made possible structures with a potential variation in one dimension (the growth direction), formed by

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the alignment of the individual band gaps. One class of semiconductor heterostructures is the superlattice (SL), constructed by alternately layering two semiconductors with different band gaps. A fundamental unit comprising of a potential-well region (smaller band-gap material) and a barrier region (larger band-gap material) is repeated along the growth direction. Calculation of electronic states for this composite material is vastly simplified by the effective mass approximation (EMA) and the envelope function method, wherein the band gaps of the individual semiconductors align to form a potential consisting of alternate regions of rectangular wells and barriers for electrons in the conduction band (CB), and holes in the valence band (VB) in the direction perpendicular to the layers [6]. The height of the potential barrier depends on the band-gap difference, which in turn depends on the composition of the semiconductors. The electronic energy states group into mini bands separated by gaps. One may then conceive of a situation in which the layer thicknesses and/or compositions are varied randomly from one unit to the next, resulting in a structure that is disordered. Evolution of disorder-induced effects may then be examined by engineering superlattices with controlled amounts of disorder. There has been some work on disordered semiconductor SLs reported in the literature with the main idea of observing Anderson localization and mobility edges [7–11]. Photoluminescence measurements and calculations have been performed which clearly indicate the presence of disorder induced localized states. In this work we have chosen to investigate the electronic states in $GaAs/Al_{0.3}Ga_{0.7}As$ superlattices in which the thicknesses of the GaAs layers are varied randomly, yielding a non-periodic potential distribution in the EMA for electrons and holes, using photoluminescence (PL) and transmission measurements. These measurements yield the energy of transition between the bottom of the valence mini band and the bottom of the conduction mini band. Disorder introduces a red shift in the observed transitions. We examine the quantitative relation between the measured shift and the localization length. The localization length obtained in this manner is compared with that obtained from an independent numerical calculation of the wavefunction. The experimental results are presented first followed by the calculations, comparisons, and a discussion.

2. Experimental procedure

Transmission and PL measurements in the spectral region of 1.2 eV to 2.0 eV were performed on two sets of $GaAs/Al_{0.3}Ga_{0.7}As$ superlattices each consisting of an ordered or periodic SL (OSL) and a corresponding disordered or non-periodic SL (DSL) in which the well widths are randomly distributed about a mean value matching that of the OSL. Each sample consisted of forty GaAs wells separated by $Al_{0.3}Ga_{0.7}As$ barriers of width 9 Å. The period of the OSL, with well width $L_W = 77$ Å was 86 Å (86 OSL) and that of the OSL with $L_W = 40$ Å was 49 Å (49 OSL). In the corresponding DSLs, the well widths had a mean value of 77 Å with a standard deviation, σ, of 14 Å in one and a mean of 40 Å with a σ of 12 Å in the other. The samples were grown by MBE on GaAs substrates separated by a 1500 Å buffer of GaAs followed by a layer of $Al_{0.3}Ga_{0.7}As$ of thickness 2000 Å. For transmission, a window was made by etching away a small region of the substrate and the sample was mounted on a glass plate. Measurements were made at 20 K, produced by a closed-cycle helium refrigerator. The transmission spectrum of white light from a 100 W tungsten lamp was measured for normal incidence. PL was excited by a laser diode emitting at a wavelength of 750 nm and measurements were made at 4.2 K and at various temperatures up to 45 K. The spectra were obtained using a Spex model 1700 II Czerny–Turner spectrometer, a photomultiplier tube and lock-in techniques. The absorption coefficient was extracted from the transmission data

by applying the equation for transmission through an absorbing thin film on a transparent substrate (the glass mount) surrounded by vacuum, given by [12]

$$T = \frac{A}{e^{\alpha t} + Be^{-\alpha t} + C\cos(2\gamma)} \tag{2}$$

where

$$A = (1+g_1)^2(1+g_2)^2(1+g_3)^2 \qquad B = ((g_1(g_2+g_3))/(1+g_2g_3))^2$$

$$C = 2g_1(g_2+g_3)/(1+g_2g_3) \qquad \gamma = 2\pi nt/\lambda$$

where

$$g_1 = \frac{n_0 - n}{n_0 + n} \qquad g_2 = \frac{n - n_2}{n + n_2} \qquad g_3 = \frac{n_2 - n_0}{n_2 + n_0}.$$

The refractive indices of the vacuum, thin film (sample) and glass are denoted by $n_0(=1)$, $n_1(= n - ik)$, $n_2(= 1.5)$, t is the thickness of the film and λ is the wavelength. The absorption coefficient, α, is related to the imaginary part of the refractive index of the sample by $\alpha = 4\pi k/\lambda$. Published values for GaAs were used for the real part n which is dispersive and varies between 3.6 and 3.8 in the region 1.5 eV to 1.8 eV [13]. The transmission spectra were normalized by setting the transmission values below the band gap to be that given by equation 2 assuming zero absorption. Plots of the spectrum of α for the four samples are presented in figure 1. The plots have been displaced vertically for clarity. The position of the band edges rather than the absolute value of α is of relevance. The band edge at 1.51 eV observed in all four samples is from the remnants of the GaAs substrate/buffer that is not etched away. The features above this edge are the transitions of the superlattice. The PL data for the four samples are presented in figure 2. The main PL peak position and the fundamental absorption edge coincide in each case and correspond to the first heavy-hole exciton. The emissions at lower energies broaden and decrease in peak intensity with increase in temperature and are therefore identified to be impurity bound excitonic transitions. The first light-hole exciton is observed at higher temperatures. Transmission and PL data clearly show that the fundamental transitions for the DSLs are red-shifted from those of the corresponding OSLs. The second absorption band observed for the 86 OSL is missing in the 86 DSL. The PL intensities from the DSLs are ten to twenty times stronger than those from the OSLs. Our goal is to understand these observed differences between the OSLs and DSLs and to relate them to quantitative aspects of disorder.

3. Calculations and discussion

We consider first the red shift between the OSL and the DSL. In the EMA, the CB and VB edges of the constituent semiconductors are assumed to form a multiple-square-well potential distribution for electrons and holes. In a periodic potential the wavefunctions are extended. In a non-periodic field, repeated backscattering causes the wavefunction to be localized. The tail of the wavefunction decays exponentially with a decay length defined to be the localization length [4]. The relation between the localization distances and the density of states has been investigated by Thouless [14]. According to Thouless, in the limit of weak

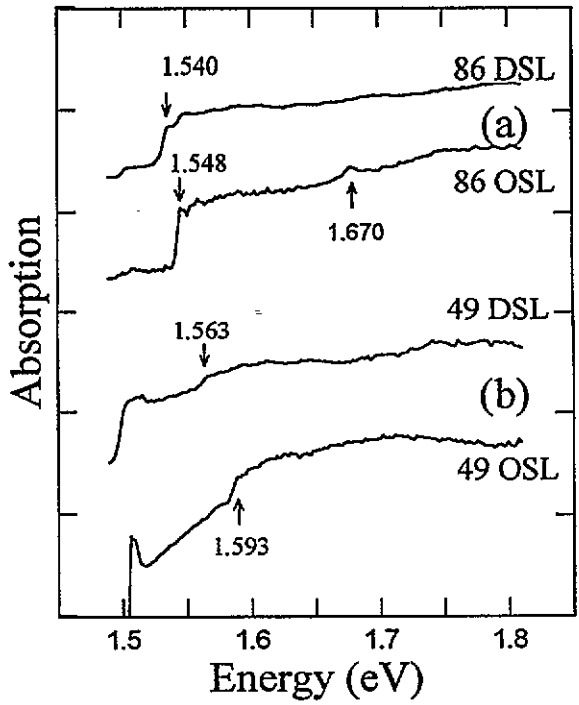

Figure 1. Comparison of the optical absorption (α) of the DSL with that of the corresponding OSL for (a) 86 OSL and DSL, (b) 49 OSL and DSL. The first heavy-hole excitonic transition energy (in eV) indicated for each curve coincides with the main PL peak position for that sample, shown in figure 2.

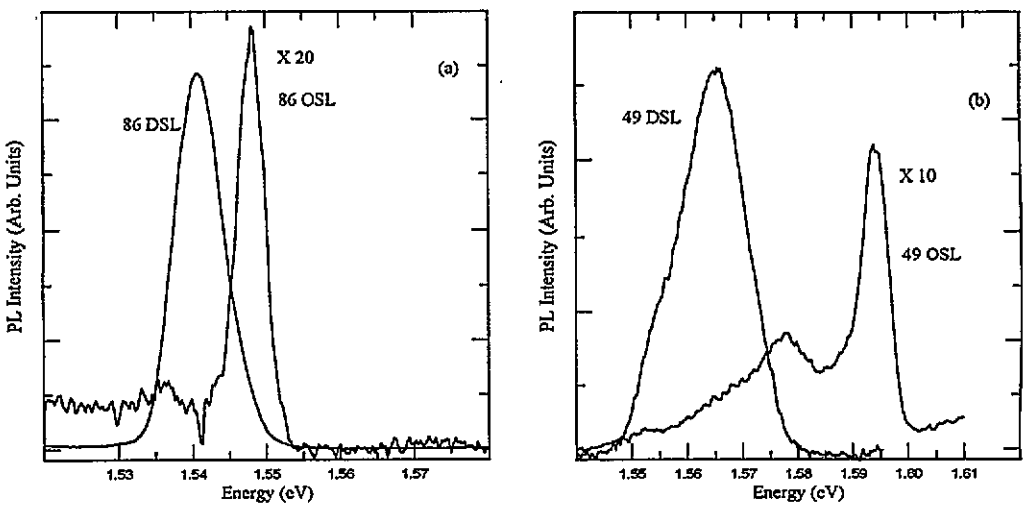

Figure 2. Photoluminescence at 4.2 K in (a) 86 OSL and DSL, (b) 49 OSL and DSL.

disorder, the wavefunction amplitude localization length, ξ, is twice the MFP for backward

scattering, that is

$$\xi = 2v\tau \tag{3}$$

where $v = L/(h\rho(E))$ (L is the length of the sample, $\rho(E)$ is the density of states, h is Planck's constant) is the velocity of the electron, τ is the backward scattering time. The expression in equation 3 was obtained by treating the weak disorder as a perturbation over the periodic field. The matrix element of the perturbation connecting the energy levels of the unperturbed system is of the order of $\hbar/\tau$ which is also the shift in energy produced by the perturbation or the disorder. Therefore the shift in the energy of the electronic ground state, E_{1c}, in a disordered potential may be expressed as

$$\Delta E_{1c} \sim \hbar/\tau. \tag{4}$$

Use of the uncertainty principle yields the same result. Combining the relation (4) with equation (3) gives a relation between the localization length and the shift in electron energy in the limit of weak disorder that reads

$$\xi = 2(L/h\rho(E))\hbar/\Delta E_{1c}.$$

Replacing the density of states, $\rho(E)$, for the periodic system by its average value of $2N/W$, where N is the number of repeating units (or wells) and W is the width of the energy mini band, gives

$$\xi = (L/2\pi N)(W/\Delta E_{1c}) = (d/2\pi)(W/\Delta E_{1c}) \tag{5}$$

where $L/N = d$ is the period of the superlattice.

In the experiments, the measured quantity is the conduction to valence mini band transition energy, modified by the exciton binding energy, E_{nbe} (due to the Coulomb attraction between the electron in the CB and the hole in the VB) and is given by

$$E_n = E_G + E_{nc} + E_{nv} - E_{nbe} = E_{An} - E_{nbe}$$

where $E_G = 1.52$ eV, is the value of the GaAs band gap. The subscript n is the mini band index, and E_{nc} and E_{nv} are the numerical values for the energy position of the bottom of the mini band as measured from the CB and VB edges of GaAs respectively. Transitions occur between conduction and valence mini bands of the same index for the OSLs. Transmission measurements yield E_n and PL measurements yield E_1, while calculations give the values of E_{nc} and E_{nv}. The observed red shift in E_1 between the OSL and the DSL includes the shift in both E_{1c} and E_{1v} and the difference in the exciton binding energy. Thus $\Delta E_1 = \Delta E_{1c} + \Delta E_{1v} + \Delta E_{nbe}$. The exciton binding energy in the superlattices is nearly the same as that in bulk where the value is 4.2 meV. Chomette *et al* have calculated a value of 5 meV for a similar SL of period 60 Å [15]. Numerical calculations of the wavefunction and electronic energies for the 86 DSL show that the wavefunction, although localized has a full width at half maximum (FWHM) of 450 Å and the corresponding exciton binding energy is expected to be bulklike, just as for the OSL, so that $\Delta E_{1be} \approx 0$. Furthermore, the effect of disorder is stronger on the electrons than on the holes so that $\Delta E_{1c} \gg \Delta E_{1v}$ [7]. Thus for weak disorder, the red shift ΔE_1 may be approximated as ΔE_{1c} of equation 5. For the 86 DSL, $\Delta E_1 = 7.5$ meV and the calculated value of W for the 86 OSL is 48.5 meV. Substituting these values into equation (5), gives $\xi = 1.03\ d$. Independent calculations

of the wavefunctions and energies of the valence and conduction mini bands for both the DSLs and the OSLs were also performed using the numerical method described by Kolbas and Holonyak [16]. The potential barrier height was taken to be 250 meV in the CB and 150 meV in the VB [17], the values used for the electron effective mass were 0.067 m_0 in the well and 0.0919 m_0 in the barrier and that for the HH in the VB was 0.62 m_0 [18], where m_0 is the bare electron mass. The ground-state wavefunctions for the 86 OSL and DSL are plotted in figure 3. The wavefunction is oscillatory in both cases. In the OSLs the electron is confined within the SL of length L between boundaries at $z = 0$ and $z = L$ and the wavefunction oscillation amplitudes may be taken to be of the form of $\sin(\pi z/L)$. Disorder causes the tail of the wavefunction to decay exponentially. The localization length, ξ, in the DSL, was then obtained by fitting the wavefunction oscillation amplitude to the form $\sin(\pi z/L)\exp(-z/\xi)$, in the tail region. For the 86 DSL, ξ obtained in this manner was 1.6 d. This is in remarkably close agreement with the value of 1.03 d obtained from equation 5. Calculations were also performed for another DSL of fifty wells with an average well width of 80 Å and a standard deviation of 3 Å. For this sample, ΔE_{1c} was calculated to be 0.5 meV and W was 45 meV, which when substituted in equation 5 yields, $\xi = 14.3\ d$. A fit to the calculated wavefunction gives $\xi = 12.8\ d$. The observed value of E_1 (1.592 eV) for the 40 OSL is higher than the calculated value of E_{A1} (1.585 eV), indicating that the layer widths may be smaller than the assumed values. However, calculations do agree with the experimental value of E_1 (1.563) for the 49 DSL if we assume an exciton binding energy of 8 meV. A value of 8 meV for the exciton binding energy is reasonable for the 49 DSL because the strong disorder in this sample causes the exciton to be more localized leading to a larger binding energy. The red shift of 30 meV observed for the 49 DSL is therefore not the true red shift from the OSL. Using the calculated values of $W = 160$ meV and $\Delta E_{1c} = 9$ meV for the 49 DSL in equation 5, gives $\xi = 2.8\ d$, while a fit to the calculated wavefunction gives 2.75 d.

It is easy to understand the reason for not observing the second absorption edge for the 86 DSL. Calculations show that for the 86 OSL two conduction mini bands exist below the continuum (within the well) and two absorption edges are observed. For the DSLs there are no mini bands or gaps because of the lack of periodicity and consequently only the first transition, E_1, is observed. In the present calculation, the use of the effective mass approximation together with the envelope function method permit a one-dimensional treatment for the superlattice potential. Wang *et al* [19] have calculated the density of states within a tight-binding framework for a three-dimensional periodic random superlattice, made up of two types of layers, in which the layers within a period are randomly distributed while the two-dimensional periodicity in the plane of the layers is preserved. The total number of layers considered is twenty. Their results show that a three-dimensional treatment yields band tail states that extend farther into the band gap. Such a treatment would of course be prohibitive for our samples where the number of layers is 1200. A conclusion from their work that is of relevance here is that although the disorder may be one-dimensional, a three-dimensional treatment is needed for a correct description of the density of band tail states. These states lead to a tail in the absorption edge similar to the Urbach tail in amorphous materials [20] and a broadening of the PL line. These are effects that we observe in our experiments. In disordered systems the position of the band edge in transmission and the PL peak are identified as the mobility edge [20]. A one-dimensional treatment for our samples may not give the correct density of band tail states but gives a good first approximation for the band edge or the mobility edge. In this work we have been concerned with the position of the band edge rather that the width of the distribution of the band tail states. A shrinking of the effective band gap with increasing amounts of disorder is an established result [20]

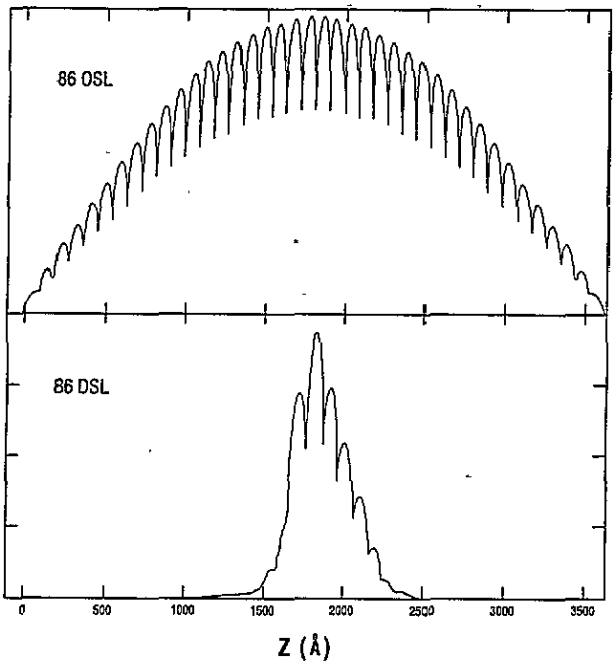

Figure 3. Wavefunction in arbitrary units for the 86 OSL and the 86 DSL in the EMA along the growth direction of the SL (z).

and we observe this for our intentionally disordered superlattices. A line shape analysis is an area for future work.

We now turn to a discussion of the PL intensities in OSLs and DSLs. The free exciton in a 3D bulk crystal has translational symmetry and interacts with photons of the same wavevector forming the excitonic polariton which is a stationary state that in principle does not decay [21]. However interaction with crystal boundaries, imperfections in the crystal and acoustic phonons causes the radiative decay observed as PL. In a pure crystal, the free exciton is expected to have long radiative lifetimes. In a quantum well there is no translational symmetry in the growth direction and the exciton is localized in this direction. The confined exciton can couple to a whole distribution of photons of wavevector $|k| < n\omega_0/c$ (where n is the refractive index, ω_0 is the frequency of the photon at the exciton energy and c is the velocity of light) and radiative decay can occur resulting in a shorter radiative lifetime [22,23]. A similar comparison may be drawn between OSLs and DSLs. In the DSL the breakdown of translational symmetry in the growth direction causes the exciton to be localized. The free exciton in the DSL may therefore be expected to have a short lifetime than in the OSL. PL intensities are inversely proportional to the radiative lifetimes which could account for their enhancement in the DSLs.

We have attempted to illustrate that semiconductor superlattices are model candidates for studying disorder induced phenomena because the strength of disorder and the length scales can be controlled. We have shown that the localization length in disordered superlattices can be obtained very simply from a shift in the ground state energy. Disordered superlattices with varying degrees of disorder can be used for studying the characteristics of polaritonic luminescence.

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