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Direct and indirect transitions in $(\text{GaAs})_n/(\text{AlAs})_n$ superlattices with $n = 1 - 15$

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ABSTRACT

Photoreflectance and photoluminescence measurements are carried out to clarify optical properties of $(\text{GaAs})_n/(\text{AlAs})_n$ ($n = 1-15$) short-period superlattices, placing main interest in the crossover of direct-indirect transition and zone-folded weak transition. Photoreflectance spectra of $(\text{GaAs})_n/(\text{AlAs})_n$ with $n < 10$ exhibit a weak structure below the main (strong) structure. Photoluminescence peaks appear at the photon energies corresponding to the critical points of these weak and strong structures. Energy band calculations are performed by using the empirical tight-binding method. Calculations of momentum matrix elements between the top valence band and lowest three conduction bands at the Γ point show an existence of weakly allowed direct transition below the strongly allowed direct transition edge in $(\text{GaAs})_n/(\text{AlAs})_n$ with $n \leq 5$. These results strongly suggest that the observed weak structures in the photoreflectance arise from the weakly allowed direct transition, indicating that the conduction band reflects the nature of the zone-folding effect (X_z).

1. INTRODUCTION

Semiconductor heterostructures such as quantum well structures and short period superlattices (SL's) have revealed various interesting electrical and optical properties. These unique properties arise from the unique features of the electronic density of states, which are quite different from those in bulk materials. From the view points of device application the confinement of the electrons in quantum wells and isolation of the electrons from the donors¹ are very important and high electron mobility is achieved in HEMT, which is known to be a very important low noise and high frequency amplifier.² Multi-quantum well laser is also known to give very high efficiency. These devices utilize the confinement of the electrons in GaAs well regions separated by AlAs or AlGaAs barrier regions, and thus these electrons exhibit two-dimensional nature.

Recently many interesting papers have been published on the optical transition in short period SL's of $(\text{GaAs})_m/(\text{AlAs})_n$.³⁻¹¹ The motivation of these researches are due to the question whether the optical transition in these SL's with small m and n is direct or indirect. This question arises from the fact that the lowest conduction band in GaAs locates at the Γ point (direct band gap), while the lowest conduction band in AlAs lies in the X point (indirect band gap) and it is lower than the conduction band of GaAs at the X point. Therefore the conduction bands and the valence bands at the Γ point results in a type I SL, whereas the conduction bands at the X point and the valence bands at the Γ point form a type II SL. In a SL with a large n value, which should be called '*quantum well*' because of the confinement of electrons in GaAs layers, the lowest optical transition is direct, where the transition occurs between the two-dimensional states of electrons and holes in GaAs layers. When we reduce the atomic layer number of AlAs (and thus for a small value of n), it is expected that the SL exhibit indirect transition because of the zone folding of the X point into the Γ point. Up to now two different concepts have been reported on

the mechanisms of the optical transition in short period SL's. The first one is the type II band alignment due to the crossing of the lowest Γ conduction band and the lowest X conduction band. In this concept optical transitions are understood to occur between the electrons in the X valley in AlAs layer and the hole states in the valence bands of GaAs. We have to note that this kind of transition is very weak when the carriers are confined in different layers (electrons in AlAs and holes in GaAs). This is because the strength of the optical transition is proportional to the squared overlap integral of electron and hole wave functions;

$$M_{if} \propto \int \zeta_h(z)\zeta_e(z)dz, \quad (1)$$

where $\zeta_h(z)$ and $\zeta_e(z)$ are the wave functions of hole and electron, respectively. Noting that the electrons and holes are separated in space, the integral of eq.(1) is almost zero. In order to obtain a considerable magnitude of the overlap integral, the wave functions of electrons and holes should be extended from AlAs into GaAs layer and vice versa. This means that the concept is incorrect in the sense that the extended nature of the wave functions means a wave vector dependent electronic energy, resulting in a formation of minibands. In other words, the electronic energy is not flat in the Brillouin zone and we have to take into account the dispersion of $\varepsilon(k)$. This conflicting assignment will be removed if we take into account the second concept, the zone folding effect, which is interpreted in the following way. The Brillouin zone of the short period SL's is folded in the direction of k -vector perpendicular to the layer and thus it is expected that the lowest conduction band of $(\text{GaAs})_1/(\text{AlAs})_1$ SL becomes indirect or pseudo-direct with the lowest state having predominantly the character of the folded X -point state, which is called ' X_z ' state. The ordering of the conduction bands with the character of GaAs Γ conduction band and the zone-folded AlAs X_z character, depends on the layer number n .

Modulation spectroscopy has been successfully used to investigate optical properties of semiconductor heterostructures.^{10,13-16} Most of them use photoreflectance (PR) spectroscopy, but Lee *et al.*¹⁶ has shown that piezoreflectance method gives very fine structures. These methods provide modulation without any electrode contacts and well resolved structures of modulated reflectivity which give an accurate determination of transition energies or the critical point energies, when the analysis based on Aspnes' analytical formula^{17,18} is used. These reports except our papers^{10,19} concern with modulation spectroscopy in quantum well structures and concentrate their interest in the optical transitions of the confined states such as two-dimensional states or excitons of two-dimensional electrons and holes. On the other hand we reported that the modulation spectroscopy provides a very detailed information about the optical transition in SL's such as direct or indirect transition when we combine the measurements with photoluminescence (PL) experiments.

In this paper we will report PR spectroscopy of short period $(\text{GaAs})_n/(\text{AlAs})_n$ SL's with $n = 1-15$ and discuss the controversy stated above. Photoreflectance measurements were carried out in order to determine the critical point energies due to direct transition and PL measurements were also made for a comparison. One of the most interesting findings is the weak structure observed in PR spectra in the low energy region well below the strong direct transition. Since the PR spectra arises from the direct optical transition, the weak structure of the PR is interpreted as the direct transition arising from the zone-folded direct band gap. In order to discuss the selection rule of the optical transition, we carried out energy band calculations of $(\text{GaAs})_n/(\text{AlAs})_n$ SL's based on the tight binding method and also of the matrix elements between the valence bands and conduction bands. These calculations support the assignment that the observed weak structure in the PR measurements arises from the zone-folded direct transition.

2. EXPERIMENTAL PROCEDURES

The samples used in the present work were epitaxially grown at 570°C on (100) semi-insulating GaAs substrates by molecular beam epitaxy. The atomic layer number n ranged from 1 to 15 and was accurately

controlled by monitoring the period of the intensity oscillation of a specularly reflected beam in a reflection high energy electron diffraction (RHEED) pattern. The SLs consist of about 200 multiple layers with good quality. The thickness of the individual atomic layers of the SLs has been confirmed by x-ray diffraction measurements.

The experimental arrangement used in the present work is reported elsewhere^{10,12}, which is basically similar to those used by Glembocki *et al.*^{13,14} The modulation is accomplished by mechanically chopping a laser of photon energy greater than the band gap of the sample. In the present work we used Ar ion laser chopped at a frequency of 210 Hz. The intensity of the Ar ion laser, 488.0 or 476.5nm, was reduced to about 0.2mW by using neutral density filters. The reflectance measurement was performed with a probe beam of white light (Xe arc lamp) dispersed by a 50cm single monochromator (JASCO CT-50S) with a 1200 lines/mm grating blazed at 750nm. The light beam reflected from the sample surface was passed through a filter which blocks the laser light and is detected by a photomultiplier tube. Photoluminescence measurements were made by using the Ar ion laser line 488.0nm of about 1W for excitation and by detecting the luminescence spectra with an 80cm focal length double monochromator (Spex 1401).

It is well known that the modulated reflectance can be expressed as

$$\Delta R/R = \alpha \Delta \epsilon_1 + \beta \Delta \epsilon_2, \quad (2)$$

where $\Delta \epsilon = \Delta \epsilon_1 + i \Delta \epsilon_2$ is the perturbation-induced change in the dielectric constant ϵ , and α and β are the Seraphin coefficients which are a function of real and imaginary parts of the dielectric constant, ϵ_1 and ϵ_2 . A detailed discussion on calculating $\Delta \epsilon$ and its effects on the line shape is given by Aspnes.^{17,18} According to the calculation of Aspnes the modulated reflectance is given by

$$\Delta R/R = \sum_j^p \text{Re}[C_j \exp(i\theta_j)(E - E_{gj} + i\Gamma_j)^{-m_j}], \quad (3)$$

where p is the number of critical points, E the photon energy, C_j , θ_j , E_{gj} , and Γ_j are the amplitude, phase, energy gap, broadening parameter, respectively, of the j -th critical point. The value of m_j is a parameter which depends on the critical point type, $m_j = 3.5, 3.0$ and 2.5 for the one-, two- and three-dimensional critical point, respectively. Very recently similar analysis has been made by Glembocki and Shanabrook²⁰ who showed that PR spectrum of quantum well structures is to be analyzed by the first derivative formula when the electrons and holes are confined in the well region. In the samples used in the present work, however, electrons and holes are extended in the SL and thus their wave functions are three-dimensional-like. Therefore, the experimental data of PR are analyzed by using eq. (3) of Aspnes' formula with $m_j = 2.5$ (three-dimensional critical point).

In SL's, the Brillouin zone is folded and mini-bands are formed, resulting in many critical points in a narrow region of photon energy. This means that the PR spectra consist of a combination of many critical points, and in other words we have to put a value greater than 2 for p in eq. (2). The best fitting procedure was carried out by adopting the method described elsewhere.^{10,12}

3. RESULTS AND DISCUSSION

Figure 1 shows a typical result of photoreflectance (PR) and photoluminescence (PL) spectra in (GaAs)₈/(AlAs)₈ SL at $T = 200\text{K}$, where the solid circles in the lower trace represent experimental data and the solid curve is best fitted to the data using Aspnes' third derivative formula. The experimental data of PR are plotted with a suitable interval so that a comparison between the experimental and best fitted curves becomes clear. In Fig. 1 we find that the PL consists of two main peaks, while the PR spectrum exhibits a very complicated structure due to an existence of many critical points in the

Fig. 1. Photoluminescence (upper trace) and PR (lower trace) spectra observed in $(\text{GaAs})_8/(\text{AlAs})_8$ at $T = 200\text{K}$. The solid curve in the lower trace is calculated from Aspnes' third derivative formula so as to best fit to the experimental reflectance curve and the vertical arrows indicate the transition energies estimated from the best fitting.

Fig. 2. Four components of the reflectance spectra for $(\text{GaAs})_8/(\text{AlAs})_8$ at $T = 200\text{K}$ are separately displayed, where these components are obtained by the best fitting shown in Fig. 1.

Fig. 3. Same as Fig. 1 but for $(\text{GaAs})_{12}/(\text{AlAs})_{12}$ at $T = 200\text{K}$. The PR spectra consists of one weak structure and three strong structures as shown in Fig. 4.

Fig. 4. Photorefectance spectra for $(\text{GaAs})_{12}/(\text{AlAs})_{12}$ at $T = 200\text{K}$ are resolved by the best fitting procedure and the vertical arrows indicate the transition energies.

energy range 1.75 to 2.0 eV. The PL peaks in the photon energy region 1.8 to 2.0 eV seem to correspond to the PR structure in this region. We find very weak emission at lower energy side in PL spectra. In SL's with smaller n , the emission of the low energy side, where we have no structure in PR, becomes stronger and exceeds the emission of the higher energy side. We do not know the origin of the low energy side emission. In order to find the critical point energies we plotted the four components of the PR spectra separately in Fig. 2, where we find that the complicated PR spectra are well resolved and we can estimate the critical point energies with a good accuracy. The vertical arrows in Figs. 1 and 2 indicate the critical point energies obtained from the best fitting. It is very important to point out that the PL peak at 1.815 eV coincides with the critical point energy 1.797 eV of the weak structure in the PR spectra. Taking into account the fact that the PR spectra arise from the critical points of the joint density of states and thus from the direct transition process, the SL of $(\text{GaAs})_8/(\text{AlAs})_8$ has a direct gap at $E_g = 1.797\text{eV}$ at $T = 200\text{K}$, but this transition is very weak compared to the transition at 1.897, 1.915 and 1.951 eV. It is very interesting to remind that the spectra of PR have fine structures and the analysis of the best fitting provides accurate determination of the critical point energies. Similar features are observed in other SL's investigated in the present work.

These features have been observed in our previous work at room temperature, where a weak structure was observed only in $(\text{GaAs})_5/(\text{AlAs})_5$ at lower energy side.¹⁹ Two possibilities are proposed for the lower energy peak; (1) direct transition at the Γ point associated with the zone-folded conduction band and (2) indirect transition between the lowest conduction band located at X or at a point in the Brillouin zone other than the Γ point and the valence band at the Γ point. As stated before, the PR spectra arise not from a indirect transition but from a direct transition because the indirect transition is higher order perturbation compared to the direct transition. As we can see in Fig. 1 the weak structure in the PR spectra gives the critical point energy corresponding to the PL peak, the lower energy structure may be assigned to the weakly allowed direct transition between the zone folded conduction band and the

Fig. 5. Transition energies obtained from the PR and PL experiments at $T = 200\text{K}$ are plotted as a function of the atomic layer number n , along with the calculated curves. The strong (main) structures in the PR spectra are shown by open circles (referred as PR Expt. (Γ)), the weak structure by solid circles (referred as PR Expt. (X_z)) which is ascribed to the zone-folded weak transition, and PL peaks by crosses (referred as PL Expt.). The calculated results from the tight-binding method are shown by the solid curve for the direct allowed transition, by the dashed curve for the weakly allowed transition which arises from the zone-folding effect (referred as TB direct (forbidden)), and the lowest indirect transition (referred as TB indirect).

valence band. In our previous paper,¹⁹ we assigned the lower energy peak of the PL peaks not to the direct transition but to the indirect transition. This is because the weak structure at lower energy region in the PR spectra has been observed only in the SL (GaAs)₅/(AlAs)₅. In the present work, however, we carried out similar experiments very carefully by changing the temperature and improving the sensitivity of the detection, and found the weak structures in almost all the samples we investigated.

Figure 3 represent PL and PR spectra for (GaAs)₁₂/(AlAs)₁₂ at $T = 200\text{K}$, where we find that only one main emission is observed in the PL spectrum and the weak structure of PR in the lower energy region merges into the main structure. The resolved spectra of the PR signals are shown in Fig. 4, where we find three transition energies in a narrow photon energy region from 1.7 to 1.85 eV, whereas the PL signals consist of a main peak and a weak shoulder at higher energy side.

The present results are summarized in Fig. 5, where we plotted transition energies determined from the PR spectra by solid and open circles, and the PL peak energies by the crosses. In Fig. 5 the transition energy for the weak structure in the PR signals is shown by the solid circles. Although the weak structures at lower energy side are not observed in all the samples investigated, it is very interesting to point out that the lowest PL peak agrees well with the transition energy of the weak structure in the PR at the low energy side. Another interesting feature is that the higher energy peak of the PL agrees well with the transition energy of the strongest structure of the PR. Calculated results are also shown in Fig. 5 which will be discussed later.

4. ENERGY BAND CALCULATIONS OF SUPERLATTICES

Up to now there have been published many papers on the energy band calculations of short period SL's by using various methods.²¹⁻²⁹ Most of them show that the lowest conduction band of (GaAs)_n/(AlAs)_n with small n forms indirect band. Our calculations based on the empirical tight-binding method reveals that the SL's are indirect for $n < 10$.¹⁰ In the previous paper¹⁹ we compared our experimental data with the calculations and found that the atomic layer number dependence of the energy gaps determined by PR and PL measurements are well explained by the crossover of the direct and indirect transitions around $n = 10$, although the absolute values of the energy gaps exhibit some discrepancy. The discrepancy was explained in terms of the layer number fluctuation by performing energy band calculations of long period SL's such as (GaAs)_n/(AlAs)_n/(GaAs)_{n+1}/(AlAs)_n. This explanation is supported by the fact that the broadening parameters of the PR spectra are almost independent of temperature, which means that the broadening is determined not by the electron-phonon interactions but by the fluctuation of the layer number.

We have to note here that the absolute values of the calculated critical point energies of the SL's depend strongly on the parameters assumed for the calculations, such as band parameters of GaAs and AlAs, and also the conduction band or valence band discontinuity. The accuracy of the energy band calculation is in a range of 0.1 to 1 eV, and thus a detailed comparison between the experimental and calculated results is impossible. In our previous paper we concluded that the lowest energy peak of the PL spectra arises from the indirect transition for (GaAs)_n/(AlAs)_n with n less than 10, although we observed a weak structure in PR spectra in (GaAs)₅/(AlAs)₅ which corresponds to the lowest energy peak of the PL. If this weak structure is ascribed to the zone-folded direct transition, the conclusion drawn in the previous paper is subject to a change. As stated in Section 1, the main subject of the present work is to clarify whether the low energy peak of PL and the weak structure of the PR arise from the zone-folded direct transition or from the indirect transition. When the weak structure is observed in every SL, it may be ascribed to the zone-folded direct transition because only the direct transition gives rise to the PR signals. In the present work we observed the weak structure in almost all superlattices with $4 \leq n \leq 13$.

In order to discuss the magnitude of the optical transition probability in SL's we carried out energy band calculations using the tight binding method which is described elsewhere in detail.^{10,25} and momentum

matrix elements.^{19,30,31} Our method is based on the empirical tight binding theory which takes into account the second nearest neighbor interactions in addition to the nearest neighbor interactions of sp^3s^* orbitals as done by Yamaguchi.²⁵ In the present paper we use the same computer program prepared by our group and the same parameters.^{10,30} We use a zinc-blende-structure tight-binding Hamiltonian for the basis of quasiatomic functions localized in the unit cell at R_i , $|n, b, R_i\rangle$. The Bloch-type tight-binding states are given by

$$|n, b, \mathbf{k}\rangle = \frac{1}{\sqrt{N}} \sum_i [\exp\{i(\mathbf{k} \cdot \mathbf{R}_i + \mathbf{k} \cdot \mathbf{V}_b)\}] \times |n, b, \mathbf{R}_i\rangle, \quad (4)$$

where quantum numbers n run over the s, p_x, p_y, p_z , and s^* (= excited s) orbitals. The N wave vectors $\mathbf{k}$ lie in the first Brillouin zone. The site index b is either a (for anion) or c (for cation). The anion positions are R_i and the cation positions are $R_i + V_b$ with $V_b = \delta_{c,b}(a_L/4)(1, 1, 1)$ and a_L being the lattice constant, in terms of Kronecker δ . The quasiatomic functions are Löwdin orbitals, which are symmetrically orthogonalized atomic orbitals. The Schrödinger equation for the Bloch function $|\mathbf{k}, \lambda\rangle$ is

$$[H - E(\mathbf{k}, \lambda)]|\mathbf{k}, \lambda\rangle = 0, \quad (5)$$

or, in this basis

$$\sum_{m, b'} [\langle n, b, \mathbf{k} | H | m, b', \mathbf{k} \rangle - E(\mathbf{k}, \lambda) \delta_{n, m} \delta_{b, b'}] \langle m, b', \mathbf{k} | \mathbf{k}, \lambda \rangle = 0. \quad (6)$$

The solutions are

$$|\mathbf{k}, \lambda\rangle = \sum_{n, b} |n, b, \mathbf{k}\rangle \langle n, b, \mathbf{k} | \mathbf{k}, \lambda \rangle, \quad (7)$$

where the band index λ has $10 \times (n + n)$ values for the $(\text{GaAs})_n/(\text{AlAs})_n$. Diagonalization of the Hamiltonian in this $|n, b, \mathbf{k}\rangle$ basis gives eigen values (energy bands) and eigen vectors (wave functions). The momentum matrix element between the valence and conduction bands is defined as

$$M_x = \sum_{b, n, n'} \langle n', b, \mathbf{k} | C_{bn'} P_x C_{bn} | n, b, \mathbf{k} \rangle \quad (8)$$

where P_x is the x -component of momentum operator, and the coefficients C_{bn} and $C_{bn'}$ are those defined by the right hand side of eq.(7) and given by $\langle n, b, \mathbf{k} | \mathbf{k}, \lambda \rangle$ and $\langle n', b, \mathbf{k} | \mathbf{k}, \lambda \rangle$, respectively. Calculation

Fig. 6. Brillouin zone of $(\text{GaAs})_n/(\text{AlAs})_n$ and the notation.

of the momentum matrix element is straightforward. It is evident from the symmetry consideration of the SL's that the momentum matrix elements M_x and M_y are identical, and therefore we calculated M_x only.

In Fig. 6 we present Brillouin zone of the SL with the notation of the critical points, where the growth direction of the SL is (001) (the z -direction). A typical example of the energy band structure of a SL is shown in Fig. 7 for $(\text{GaAs})_1/(\text{AlAs})_1$, where we used the parameters reported elsewhere^{10,19,30} and the valence band discontinuity is 0.54 eV. We can see very clearly the zone-folding in the z -direction. In addition indirect gap appears with the lowest conduction band at the R point but the energy of the minimum is very close to the lowest conduction band edge at the Γ point. The effective mass of the lowest conduction band is heavier than that of the second lowest conduction band. This seems to be due to the fact that the lowest conduction band reflects the property of the conduction band of AlAs. The results for $(\text{GaAs})_5/(\text{AlAs})_5$ are shown in Fig. 8, where we find that the energy bands consist of many zone-folded bands and that the conduction band edges at various points lie in a narrow range of energy. This results and

Fig. 7. Energy band structure of $(\text{GaAs})_1/(\text{AlAs})_1$ calculated by the tight-binding method based on sp^3s^* basis.

Fig. 8. Energy band structure of $(\text{GaAs})_5/(\text{AlAs})_5$ calculated by the tight-binding method.

Fig. 9. Squared momentum matrix elements as a function of atomic layer number for the transition between the top valence band and the lowest, second lowest and third lowest conduction band are shown by the solid (1 - 1), dashed (1 - 2), and dot-dashed curve (1 - 3), respectively.

the accuracy of the energy band calculation indicate that the assignment of the critical points is a very difficult task.

In order to discuss the magnitude of the transition probability we carried out calculations of the matrix element given by eq. (8) and the results are shown in Fig. 9, where squared matrix elements between the top valence band and lowest three conduction bands at the Γ point are plotted as a function of the atomic layer number n . The solid, dashed and dot-dashed curves are, respectively, represent the squared matrix elements for the lowest, second lowest, and third lowest conduction bands. As seen in Fig. 9, the transition to the lowest conduction band is allowed for $n \geq 4$, whereas the transition to the second lowest conduction band is allowed for $n \leq 4$. In addition, the matrix elements oscillate in the region of small number of n . These results clearly indicate an importance of the zone-folding effect, because the zone-folding effect depends on the layer number, even or odd. As the layer number is decreased, the lowest transition 1-1 becomes very weak while the second lower transition 1-2 becomes strong. This feature is very similar to the experimental observation, in which we found that the weak structure in the PR signals appear only in the region of small n .

These calculated results are shown in Fig. 5 along with the experimental result, where the energy of the direct allowed transition is plotted by the solid curve, the energy of the weak transition by the dashed curve, and the lowest indirect gap by the dot-dashed curve. Although the agreement between the present calculations and experiments is not good, the general feature is consistent. The crossover between the strong direct transition and weak direct transition is expected to appear at about $n = 5$, while the present experiment reveals that the crossover occurs at about $n = 10$. The tight-binding calculations are made by using only five atomic states s, p_x, p_y, p_z , and s^* for each atom, and thus the accuracy of the higher energy states, especially X state, is poor. Therefore, the agreement with the experiments is understood to be rather good.

In conclusion we observed weak structures well below the strong structure in the PR spectra of $(\text{GaAs})_n/(\text{AlAs})_n$ with $n < 10$ and emission peak PL appears in the region of the weak structures. This suggests that the weak structures of the PR spectra are interpreted in terms of weakly allowed direct transition and that the transition is induced by the zone-folding effect. This interpretation is supported by the energy band calculations based on the empirical tight-binding method. The higher energy transition observed in the PR spectra agrees well with the PL peak. The present calculation indicates an existence of indirect gap in the region of small atomic layer number ($n < 8$). Although PL spectra show peaks at low energy side below the weak structures in the PR spectra, the low energy peaks are not in good agreement with the present calculation. The origin of the low energy peaks in PL is not clear at the present stage. We investigated the temperature dependence of the peak intensities of the PL at the photon energies corresponding to the weak structure and the strong structure in the PR, and the result gives the ratio of the matrix elements which is in a qualitative agreement with the theoretical calculations given in this paper.

5. REFERENCES

- *Permanent address: Research and Development Center, Toshiba Corporation, Saiwai-ku, Kawasaki City 210, Japan.
- **Permanent address: Fujitsu Laboratories, Ltd., Atsugi City, 243-01, Japan.
- ¹R. Dingle, H. L. Störmer, A. C. Gossard, and W. Wiegmann, "Electron Mobilities in Modulation-Doped Semiconductor Heterojunction Superlattices", *Appl. Phys. Lett.* **33**, pp.665-667, Oct. 1978.
- ²S. Hiyamizu, T. Mimura, T. Fujii, K. Nanbu, and H. Hashimoto, "Extremely High Mobility of Two-Dimensional Electron Gas in Selectively Doped GaAs/ N -AlGaAs Heterojunction Structures Grown by MBE", *Jpn. J. Appl. Phys.* **20**, pp.L245-L248, April 1981.
- ³E. Finkman, M. D. Sturge and M. C. Tamargo, "X-point Excitons in AlAs/GaAs Superlattices", *Appl. Phys. Lett.* **49**, pp.1299-1301, Nov. 1986. See also references in Refs. 1-9 for earlier work.

- ⁴M. Garriga, M. Cardona, N. E. Christensen, P. Lautenschlager, T. Isu, and K. Ploog, "Interband Transition of Thin-Layer GaAs/AlAs Superlattices", *Phys. Rev. B* **36**, pp.3254-3258, Aug. 1987.
- ⁵G. Danan, B. Etienne, F. Molloy, and R. Planel, "Optical Evidence of the Direct-to-Indirect-Gap transition in GaAs-AlAs Short-Period Superlattices", *Phys. Rev. B* **35**, pp.6207-6212, April 1987.
- ⁶K. J. Moore, G. Duggan, P. Dawson and C. T. Foxon, "Short-Period GaAs-AlAs Superlattices: Optical Properties and Electronic Structure", *Phys. Rev. B* **38**, pp.5535-5542, Sept. 1988.
- ⁷D. S. Jiang, K. Kelting, T. Isu, H. J. Queisser, and K. Ploog, "Luminescence Properties of $(\text{GaAs})_l/(\text{AlAs})_m$ Superlattices with (l, m) ranging from 1 to 73", *J. Appl. Phys.* **63**, pp.845-852, Feb. 1988.
- ⁸H. Kato, Y. Okada, M. Nakayama, and Y. Watanabe, "T - X Crossover in GaAs/AlAs Superlattices", *Solid State Commun.* **70**, pp.535-539, May. 1989.
- ⁹M. Nakayama, I. Tanaka, I. Kimura, and H. Nishimura, "Photoluminescence Properties of GaAs/AlAs Short-Period Superlattices", *Jpn. J. Appl. Phys.* **29**, pp.41-47, Jan. 1990.
- ¹⁰T. Nakazawa, H. Fujimoto, K. Imanishi, K. Taniguchi, C. Hamaguchi, S. Hiyamizu, and S. Sasa, "Photoreflectance and Photoluminescence Study of $(\text{GaAs})_m/(\text{AlAs})_5$ ($m = 3 - 11$) Superlattices: Direct and Indirect Transition", *J. Phys. Soc. Jpn.* **58**, pp.2192-2199, June 1989.
- ¹¹R. Cingolani, L. Baldassarre, M. Ferrara, and M. Ligarà, "Type-I-Type-II Transition in ULtra-Short-Period GaAs/AlAs Superlattices", *Phys. Rev. B* **40**, pp.6101-6107, Sept. 1989.
- ¹²T. Nakazawa, T. Matsuoka, T. Ohya, K. Taniguchi, C. Hamaguchi, H. Kato, and Y. Watanabe, "Temperature Dependence of the Energy Gaps of $(\text{GaAs})_n/(\text{AlAs})_n$ Superlattices", *SPIE Int. Conf. on Modulation Spectroscopy*, San Diego, March 19-21, 1990, in this proceedings.
- ¹³O. J. Glembocki, B. J. Shanabrook, N. Bottka, W. T. Beard, and J. Comas, "Photoreflectance Characterization of Interband Transitions in GaAs/AlGaAs Multiple Quantum Wells and Modulation-Doped Heterojunctions", *Appl. Phys. Lett.* **46**, pp.970- 972, May 1985. See also references in Refs. 10, and 12-16.
- ¹⁴O. J. Glembocki, B. V. Shanabrook, N. Bottka, W. T. Beard, and J. Comas, for Semiconductor Technology II, "Photoreflectance Characterization of GaAs/AlGaAs Thin Films, Heterojunctions and Multiple Quantum Well Structures", *Proc. SPIE Conf. on Spectroscopic Characterization Techniques for Semiconductor Technology II*, vol.524, pp.86-94, 1985.
- ¹⁵H. Shen, S. H. Pan, and F. H. Pollak, M. Dutta, and T. R. AuCoin, "Conclusive Evidence for Mini-band Dispersion in the Photoreflectance of a $\text{GaAs}/\text{Ga}_{0.74}\text{Al}_{0.26}\text{As}$ Coupled Multiple-Quantum-Well Structure", *Phys. Rev. B* **36**, pp.9384-9387, Dec. 1987.
- ¹⁶Y. R. Lee, A. K. Ramdas, F. A. Chambers, J. M. Meese, and L. R. Ram Mohan, "Piezomodulated Electronic Spectra of Semiconductor Heterostructures: $\text{GaAs}/\text{Al}_x\text{Ga}_{1-x}\text{As}$ Quantum Well Structures", *Appl. Phys. Lett.* **50**, pp.600-602, March 1987.
- ¹⁷D. E. Aspnes and J. E. Rowe, "Resonant Nonlinear Optical Susceptibility: Electoreflectance in the Low-Field Limit", *Phys. Rev. B* **35**, pp.4022-4030, May 1972.
- ¹⁸D. E. Aspnes, "Third-Derivative Modulation Spectroscopy with Low-Field Electoreflectance", *Surf. Sci.* **37**, pp.418-442, 1973.
- ¹⁹H. Fujimoto, C. Hamaguchi, T. Nakazawa, K. Taniguchi, K. Imanishi, H. Kato, and Y. Watanabe, "Direct and Indirect Transition in $(\text{GaAs})_n/(\text{AlAs})_n$ Superlattices with $n = 1 - 15$ ", *Phys. Rev. B* **41**, April 1990 (in press).
- ²⁰O. J. Glembocki and B. V. Shanabrook, "Electromodulation Spectroscopy of Confined Systems", *Superlatt. Microstruct.* **5**, pp.603-607, April 1989.
- ²¹J. N. Schulman and T. C. McGill, "Electronic Properties of the AlAs-GaAs (001) Interface and Superlattice", *Phys. Rev. B* **19**, pp.6341-6349, June 1979. See also references in Refs. 10 and 21-31 for earlier work on energy band calculations.
- ²²W. A. Harrison, "Total Energies in the Tight-Binding Theory", *Phys. Rev. B* **23**, pp.5230-5245, May 1981.
- ²³G. Bastard, "Theoretical Investigations of Superlattice Band Structure in the Envelope-Function Approximation", *Phys. Rev. B* **25**, pp.7584-7597, June 1982.

- ²⁴T. Nakayama and H. Kamimura, "Band Structure of Semiconductor Superlattices with Ultrathin Layers $(\text{GaAs})_n/(\text{AlAs})_n$ with $n = 1,2,3,4$ ", J. Phys. Soc. Jpn. **54**, pp.4726-4734, Dec. 1985.
- ²⁵E. Yamaguchi, "Theory of the DX Centers in III-V Semiconductors and (001) Superlattices", J. Phys. Soc. Jpn. **56**, pp.2835-2852, August 1987.
- ²⁶Jian-Bai-Xia, "Theoretical Analysis of Electronic Structures of Short-Period Superlattices $(\text{GaAs})_m/(\text{AlAs})_n$ and Corresponding Alloys $\text{Al}_{n/(m+n)}\text{Ga}_{m/(m+n)}\text{As}$ ", Phys. Rev. B **38**, pp.8358-8364, Oct. 1988.
- ²⁷S.-H. Wei and A. Zunger, "Electronic Structure of Ultrathin $(\text{GaAs})_n/(\text{AlAs})_n$ [001] Superlattices and the $\text{Ga}_{0.5}\text{As}_{0.5}$ Alloy", J. Appl. Phys. **63**, pp.5794-5804, June 1988.
- ²⁸R. Eppenga and M. F. H. Schuurmans, "Thin [001] and [110] GaAs/AlAs Superlattices: Distinction Between Direct and Indirect Semiconductors", Phys. Rev. B **38**, pp.3541-3544, Aug. 1988.
- ²⁹S. Gopalan, N. E. Christensen, and M. Cardona, "Band-Edge States in Short-Period $(\text{GaAs})_m/(\text{AlAs})_n$ Superlattices", Phys. Rev. B **39**, pp.5165-5174, March 1989.
- ³⁰H. Fujimoto, C. Hamaguchi, T. Nakazawa, K. Taniguchi, and K. Imanishi, "Crossover of Direct and Indirect Transitions in $(\text{GaAs})_m/(\text{AlAs})_5$ Superlattices ($m = 1-11$)", J. Phys. Soc. Jpn. **58**, pp.3727-3732, Oct. 1989.
- ³¹T. J. Drummond, E. D. Jones, H. P. Hjalmarson, and B. L. Doyle, "GaAs/ $\text{In}_x\text{Al}_{1-x}\text{As}$ ($0 \leq x \leq 0.006$) Indirect Bandgap Superlattices", *Proc. Int. Symp. GaAs and Related Compounds, Las Vegas, Nevada, 1986, Inst. Phys. Conf. Ser. No.83, Chapter 6*, pp.331-336, 1987.