GaAs 에피 성장 기술의 최근 연구 동향
(New Trends in GaAs Epitaxial Techniques)

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〈Abstract〉

Epilayer growing process has been recognized as a key technology for successful GaAs based devices and integrations. These may include HEMT, multiple quantum well structures, band gap engineering, and quantum confinement heterostructures. The fabrication of epilayers in these devices must meet very stringent requirements in terms of crystallinity, composition, film thickness and interface quality. In particular, the quality of interfaces is getting more important because the film thickness, and flatness, roughness and stability at interface of ultrathin films cause critical effects on the device performance.

This article reviews the current status of modern epitaxial techniques which have been developed in the last few years. First, the new techniques PLE, GI, MEE, TSL based on MBE technique will be reviewed and their technical importance will be stressed. Secondly, MOMBE, GSMBE, CBE which combine the advantages of MBE and MOCVD will also be discussed.

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Thirdly, the new sophisticated epitaxial technique, ALE, of which mechanism is totally different from others, will also be reviewed. Finally, areas which should be exploited more extensively to accomplish these techniques will be addressed.

I. Introduction

The properties and application of ultrathin layers of III-V semiconductiors grown by modern epitaxial techniques have been proliferated at an explosive rate. The advent of new growth techniques such as MBE (molecular beam epitaxy) and MOCVD (metalorganic chemical vapor deposition) opened the way to the growth of semiconductors, atomic layer upon atomic layer. Owing to progress in crystal availability and control, basic understanding of low-dimensional systems, and applicability of heterostructure concepts, the recent years have also been the emergence of a wide family of structures and devices.

MBE and MOCVD have been shown to be useful methods for producing ultrathin III-V compound semiconductors, such as GaAs and AlGaAs, with reasonable quality for device applications. These methods, however, suffer from two problems. One is that heterojunctions grown by these methods have rough interfaces on the atomic scale due to a large number of atomic steps. The other problem is that the growth temperature is too high to realize sharp impurity profiles.

Over the last few years several modified procedures have been used to improve the interfaces produced by MBE. These include phase - locked epitaxy (PLE), the use of growth interrupts (GI) and more recently migration enhanced epitaxy (MEE). The claimed advantages of these methods are based on conclusions drawn mainly from reflection high energy electron diffraction (RHEED) observations combined with photoluminescence (PL) and phtoluminescence excitation spectroscopy (PLE) data.

The purpose of this article is to review the new trends in GaAs processing particularly in GaAs epilayer growing technologies such as MBE related techniques, MOMBE and atomic layer epitaxy (ALE). The comparison of these techniques will also be made wherever it is appropriate.

II. New Approaches in Epitaxial Techniques

1. Improved Techniques based on MBE

MBE, MOCVD and MOMBE(GSMBE, CBE) have been shown to be useful methods for producing ultrathin III-V compound semiconductors, such as GaAs and AlGaAs, with reasonable quality for device applications. The heterojunctions grown by these methods, however, have rough interfaces on the atomic scale due to a large number of atomic steps on the substrate. Therefore, several modified procedures have been developed to improve the quality of interfaces produced by MBE. These include phase-locked epitaxy (PLE)[1], the use of growth interrupts (GI)[2 3] and more recently migration enhanced epitaxy (MEE)[4~6]

1) Phase-locked epitaxy (PLE)

PLE uses the strong intensity oscillations in reflection high energy electron diffraction (RHEED) from GaAs and AlGaAs during MBE growth. This RHEED intensity oscillation has been understood to have its origin in the formation of 2-d nucleation and subsequent propagation of 2-d island as shown in Fig. 1.[7~10] They also have characteristic damping during growth and, often, a characteristic recovery once growth is interrupted.

In the PLE, the phase of the RHEED intensity oscillations is analyzed by computer and the molecular beam shutters are operated at a particular phase. That is, the method is to phase-lock the cyclic molecular beam shutter ON and OFF operation using the RHEED oscillation. The basic concept of PLE is that the oscillation period corresponds precisely to the growth of single monolayer, i.e., a complete layer of Ga + As atoms, or $a_0/2$ in the [001] direction (a_0 ; lattice constant).

This computer controlled PLE has a great advantage for the precise control of very thin film and superlattice structures, because the number of monolayer is adjusted just by the number of RHEED oscillations, not by the deposition time, and so this technique is invulnerable to fluctuations of molecular beam fluxes.[15]

Once the stationary growth conditions were achieved (10 periods were sufficient), the sequences could be stored and repeated automatically by the computer during the rest of the growth process without further feedback from the RHEED. The RHEED does not need to be ON during the rest of growth, and the substrate can be rotated as usual for better uniformity.

This is the technique for controling the grown-layer thickness, not for improving the interface sharpness. Moreover this technique is only applicable for a particular growth condition where RHEED intensity oscillations would occur (i.e., 2-d nucleation growth mode). Also, this technique has a problem of beam shutter breakage.

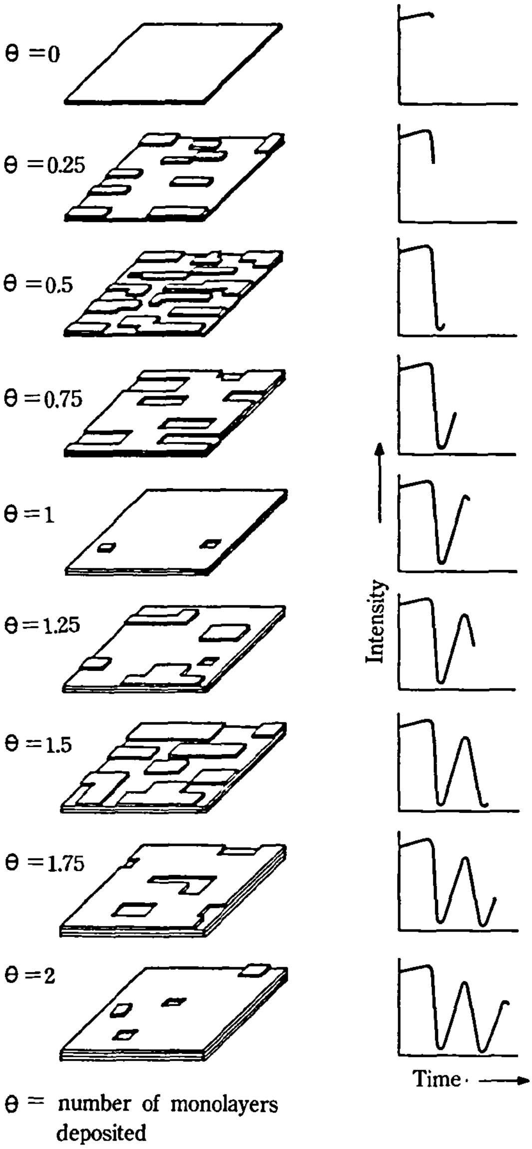

Fig. 1 First-order Growth Model (two monolayers) in Relation to the RHEED Intensity Behaviour.

2) Growth interrupts (GI)

The need to achieve monolayer abruptness at semiconductor heterojunctions grown by MBE has prompted the development of growth processing techniques that utilize interruptions in the incident molecular flux.[2,3] In this technique, the growth is interrupted by interrupting the flux of group III atoms, whilst maintaining the surface crystallinity by continuing to supply group V molecules.

The basic concept of this technique is to allow enough time for adatoms to diffuse (or migrate) until they find proper sites on the surface (i.e., the diffusion distance is increased by increasing the diffusion time).

The idea of smoothing the surface by growth interruption is implicitly obvious from the model proposed by Neave et al. to explain RHEED oscillations and recovering effects.[7]

The recovery of the RHEED intensity following cessation of growth has been identified with an increase in the mean terrace width of the surface and hence a reduction in the surface step density.[11]

This GI technique can provide atomically smooth surface (or interface), which has been revealed by the recovery of RHEED oscillation and the sharp PL peak. However, the optimization of interruption time is required as a function of substrate temperature and As flux.

3) Migration Enhanced Epitaxy (MEE)

Migration enhanced epitaxy has recently been proposed as an improved technique for epilayer growth with good optical quality at low substrate temperatures.[4-6] This technique makes use of the rapid migration of Ga and Al atoms in an As-free atmosphere by alternately depositing Ga and/or Al atoms and As_4 molecules to the growing surface (i.e., diffusion distance is increased by increasing the diffusion constant).

It has been found that MEE is useful for growing high-quality GaAs and AlGaAs layers at very low growth temperatures.[4-6] The possibility of growing III-V compounds by MBE at reduced substrate temperatures has recently attracted considerable interest for various reasons[5]: (1) no interdiffusion occurs in highly-doped heterostructures, (2) high-level doping is possible and, (3) in connection with the growth of III-V on Si, lower thermal-induced strains and better compatibility with previous Si IC process technology are found.

Fig. 2 compares the MEE result with that of the normal MBE growth.[4] In both cases, the beam flux intensities of Ga and As_4 are $J_{Ga} = 6\ X\ 10^{14}/cm^2.s$ and $J_{As4} = 4\ X\ 10^{14}/cm^2.s$, respectively. In the normal MBE growth, the RHEED oscillation almost completely disappears after 20 periods because the surface flatness in the atomic scale deteriorates. In contrast, the oscillation continues during the entire layer growth process when MEE technique is applied. This result indicates that a better surface flatness is maintained during MEE growth even after the growth of thousands of layers. This is most probably caused by the rapid migration of Ga in a very low arsenic pressure.

Furthermore, the investigation on the growth mechanism by RHEED revealed that a flat growing surface is maintained during MEE, even when the number of Ga or Al atoms deposited per cycle is not exactly adjusted to the number of surface site.

It was also found that the composition at the growing surface can deviate considerably from stoichiometry at low growth temperatures because of an excess As deposition.[6] The deviation from stoichiometry on the growing surface deteriorates the photoluminescence efficiency of the grown layers, and makes the layer structure unstable in thermal annealing. This problem was shown to be avoided by optimizing the number of As_4 molecules deposited per cycle. The optimum number of As_4 molecules needed to form one atomic layer was found to be approximately equal to half the number of surface sites.

In this technique, accurate control of beam fluxes (especially group III atoms) is required. Under MBE growth conditions, the sticking coefficient of group III elements is always nearly unity, indicating independence on the surface coverage, so that there is no natural self-regulating mechanism able to avoid accumulation of Ga on the surface once an atomic layer has been completed. However, a sort of artificial regulation or synchronization of the beam alternation to layer-to-layer growth cycle is still possible using the RHEED oscillation (combination with PLE). Also this technique has a problem of beam shutter breakage.

The various modified growth procedures mentioned above, do influence the properties of the interfaces. The claimed advantages of these techniques are based on conclusions drawn mainly from RHEED observations combined with photoluminescence/photoluminescence excitation spectroscopy data. But as Foxon has pointed out,[13] there are doubts concerning the generality of the pictures presented, because important factors were often overlooked in using the techniques which provide information.

Also these techniques use the phenomena of RHEED oscillations. Therefore the epitaxial layer is grown under the conditions where RHEED oscillations would occur (i.e., 2-d nucleation growth mode). It is still questionable which one has better quality between the films grown at 2-d nucleation growth mode and step flow growth mode.(12)

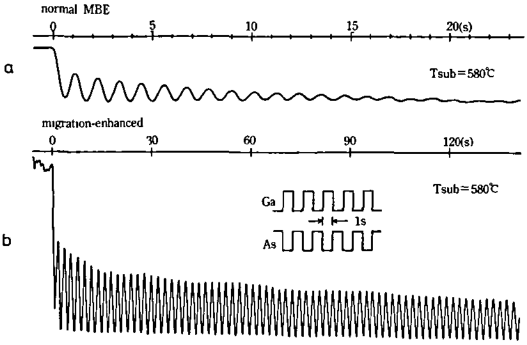

Fig. 2 RHEED Specular Beam Intensity Oscillation during the Growth of GaAs for a Normal MBE Growth (a), and for the MEE Growth (b).

4) Tilted Superlattice (TSL)

In the conventional MBE and MOCVD growth techniques, all the quantum structures have epitaxial layers with composition and effective band gap that could be varied only in a direction perpendicular to the substrate surface. The tilted superlattice (TSL) technique by MBE[12 13 14] and MOCVD[15 16] offers the possibility of producing structures with composition and band gap modulation in directions parallel or at any angle to the substrate surface.

In the case of two A and B compound semiconductors (e.g. GaAs, AlAs), the TSL are produced by depositing alternately A and B submonolayers (or fractional monolayers) on a vicinally misoriented substrate.[13,15] Fig. 3 shows a schematic of a TSL obtained by alternate deposition of $(GaAs)_m$ and $(AlAs)_n$ fractional monolayers in the sequence indicated by the numbers.

In these structures, the tilt angle of the TSL interfaces with respect to the [001] orientation (β) and the TSL period (T) may be adjusted independently by choosing the values of p ($= m + n$) and the misorientation angle (α) of the vicinal substrate plane. A 'vertical' superlattice, with the superlattice interface planes parallel to [001] and period equal to the step width, is obtained for $p=1$. By varing m and n during the TSL growth, various structures may be grown.[15,16]

The basic concept of TSL technique is that all layer nucleation events take place at step ledges and that a step-flow growth mode is established during deposition. Therefore, the TSL structural perfection is intimately tied to the stability of the deposition conditions (beam fluxes and substrate temperature) and to the uniformity of the step spacing, which are not easily controllable parameters.

The experimental results[13~16] confirm that growth conditions on vicinal (001) surface may be chosen to insure a layer-by-layer growth, where growth occurs by the addition of atoms to the step edges, without island nucleation on the step terraces. Successful growth of the TSL also verifies that growth conditions may be selected to insure the presence of a uniformly spaced array of steps on the GaAs surface that are one monolayer (one layer of Ga atoms plus one layer of As atoms) high.

A more fundamental problem concerns the interface sharpness. It has been found that interface are not compositionally sharp by the TEM observations. This could be due to[16] : (a) the presence of thermally induced kinks at step edges, (b) the existence of nucleation centers other than step edges during deposition, or (c) surface interdiffusion during deposition.

A strong optical anisotropy (i.e., anisotropy of the polarized PLE emission) characteristic has been observed in a GaAs-AlGaAs quantum well wire (QWW) array made using the TSL technique.[16] This shows that a modulation of the band gap in two dimenstions can be directly introduced by this technique.

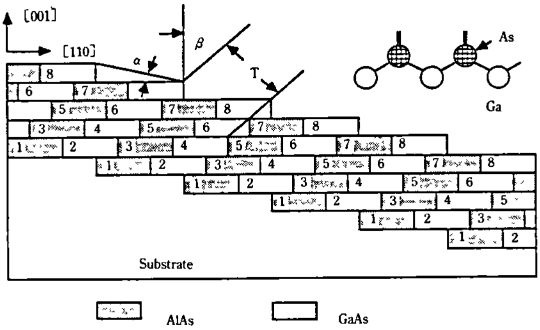

Fig. 3 Idealized Representation of a Cross Section, Perpendicular to the Step Edges, of a TSL with p = 1.25. The Inset Shows the Arrangement of the As Bonds with Respect to the Step Edges.

2. MOMBE/GSMBE/CBE

MOCVD and MBE have become the two dominant crystal-growth techniques used to synthesize sophiscated structures. However, each technique has its own advantages and disadvantages. MOCVD is a gas phase chemical deposition process which takes place at relatively high gas pressure compared to MBE. This technique provides more flexibility than MBE, particularly for the growth of phosphorous compounds and allows uniform wide area growths. However, in CVD process, reactant species have to diffuse through a gaseous boundary layer to reach the substrate. Therefore, the abruptness of heterointerfaces will suffer from the diffusion boundary layer and gas switching transients. On the othere hand, MBE is a physical deposition process that takes place in ultrahigh vacuum. No diffusion boundary layer exists, and thus, allows direct incidence of atoms or molecules in a direct line of sight onto the heated substrate. Several efforts have been made to combine the advantages of both techniques. The different experimental conditions for various epitaxial methods are summarized in Table 1.

CBE is the newest development in epitaxy growth technology,[19] In CBE, unlike MBE, which employs atomic beams(eg. Al, Ga, and In) evaporated at high temperature from elemental sources, all the sources are gaseous at room temperature. They can be organometallic or inorganometallic compounds. Unlike MOCVD, in which the chemicals reach the substrate surface by diffusing through a stagnant gas boundary layer above the substrate, the chemicals in CBE are admitted into the high-vacuum growth chamber in the form of a beam. Therefore, comparing with MBE, the main advantages include : (1) the use of room temperature gaseous group-III organometallic sources, which simplifies multiwafer scale-up ; (2) semi-infinite source supply and precision electronic flow control with instant flux response ; (3) a single group-III beam that guarantees material composition uniformity ; (4) no oval defects even at high growth rates ; and (5) high growth rates if desired.

Compared with MOCVD, these include : (1) no flow pattern problem encountered in multiwafer scale-up ; (2) the beam nature produces very abrupt heterointerfaces and ultrathin layers conveniently ; (3) clean growth environment ; (4) easy implementation of in-situ diagnostic instrumentation, e.g., RHEED and RGA ; (5) compatible with other high-vacuum thin-film processing techniques, e.g., metal evaporation, ion beam milling, and ion implantaion.

Moreover, in addition to being an important technology for epitaxial crystal growth, CBE also offers the promise of elucidating some of the fundamental surface chemical kinetics that are also relevant under MOCVD conditions.

Table 1 Comparison ov Various Epitaxial Methods

Technique	Source		Pressure (Torr)
	III	V	
MOCVD	Gas	Gas	10～760
CBE	Gas	Gas	10^{-4}
MOMBE	Gas	Solid	10^{-6}～10^{-5}
GSMBE	Solid	Gas	10^{-6}～10^{-5}
MBE	Solid	Solid	10^{-5}

3. Atomic Layer Epitaxy(ALE)

Recent development of semiconductor quantum effect devices has been greatly indebted to the progress of the thin film growth technology. However, a novel growth technology is needed for more complicated quantum devices, such as a three-dimensional carrer confinement device. As a promising method to meet with such requirements, atomic layer epitaxy or digital epitaxy was proposed.[20,21]

ALE is attracting considerable attention as the most promising growth technique for preparing ultrathin layers of uniform and precisely controlled thickness to the atomic level. The distinguishing feature of ALE is its self-limiting mechanism, i.e., only a layer of definite thickness is deposited during an exposure cycle over a wide range of growth conditions. Some characteristic features of ALE are illustrated in Fig. 4.

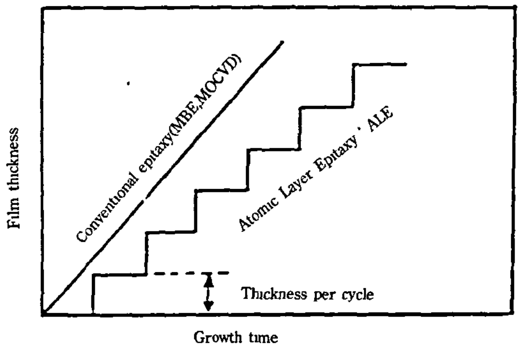

Fig. 4 Illustration of ALE.

A particularly simple way to describe the ALE approach is to say that it makes use of the difference between chemical adsorption and physical adsorption. When the first layer of atoms or molecules of a reactive species reaches a solid surface there is usually a strong interaction (chemisorption) ; subsequent layers tend to interact much less strongly (physisorption). If the initial substrate surface is heated sufficiently one can achieve a condition such that only the chemisorbed layer remains attached.

To make surface chemical reactions occur, the reactants are transported to the substarate alternately as pulses of neutral atoms or molecules, either as chopped beams in high vacuum, or as switched streams of vapor possibly on an inert carrier gas. The incident pulse reacts directly and chemically only with the outermost atomic layer of the substrate. The film therefore grows stepwise, a single monolayer per pulse, provided that at least one complete monolayer coverage is formed before the next pulse is allowed to react with the surface.[22] Unique features of ALE deduced from its mechanism are that

(1) abrupt interfce on the atomic scale
(2) film thickness depends only on the number of pulses or cycles
(3) perfectly uniform epilayer on the large area substrate
(4) selective epitaxy
(5) no hill-lock problem
(6) no interdiffusion at interface because of low growth temperature
(7) no oval defect
(8) no flow pattern problem
(9) impurity planes can be inserted (delta doping)

(10) side-wall epitaxy with monolayer unit accuracy is expected

Even though this technique combined with laser chemistry has great potentials and very attracting features for the fabrication of the sophisticated devices such as 3-d quantum confinement structures, this technology is still at infant stage compared to other techniques.

III. Conclusion

The current status of various epilayer growing techniques including modified MBE, CBE, and ALE were reviewed. These techniques appropriate for the fabrication of superlattices, quantum devices, and new functional devices have been developed very rapidly in the last few years. The driving force behind this is that the precise control of interfaces in terms of film thickness, flatness, roughness, stability, and lattice matching problem have been recognized to play important roles in the fabrication of quantum devices. Therefore, the conventional processes such as MBE and MOCVD have been modified and improved to produce new techniques such as PLE, GI, MEE, TLS, MOMBE, and CBE in the last few years. Much attention has been paid on the new ALE technique of which mechanism is very different from others. Furthermore, to make these epitaxial techniques more practical ones in the near future, more extensive experimental and theoretical studies are requested on the growth mechanisms responsible for these processes and also on the characterization techniques for the properties of ultrathin epilayers and interfaces.

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