4．「Si基板上GaA系化合物半導体内欠陥の
　　水素プラズマによる不活性化に関する研究」

Chapter 1 Introduction

1. 1 Background

GaAs and related compound semiconductor heteroepitaxial layers on silicon (Si) substrate have received considerable interest because they can be used to integrate the superior properties of the compound semiconductors with the sophisticated technology of silicon[1-13]. It opens the way to monolithically integrate the vast potential of Si microelectronics with the optoelectronic and long wavelength capabilities of III-V compound semiconductor, which in their majority are of direct band structures (contrary to the indirect Si band structures), as shown in Table 1. 1. Besides allowing monolithic integration, the GaAs epitaxy on Si substrate has many other advantages. The important ones are as follows:

(1) Compound semiconductor substrates are 3 in. or less in diameter compared to 8-. or 12-in. silicon wafers. Larger diameter wafer production requires highly mature technologies. GaAs on Si allows for the exploitation of the mature Si wafer technology to produce large diameter of GaAs epilayers that can just as effectively function as "GaAs substrates".

(2) At room temperature, the thermal conductivity of Si is three times higher than that of GaAs. Therefore, growing GaAs on Si is good for the heat sinking of GaAs devices. For example, GaAs circuits fabricated on Si substrates can exhibit higher resistance to thermal burnout and runaway than those fabricated on GaAs substrates.

(3) Silicon exhibits superior mechanical properties: To achieve the same mechanical strength as GaAs on Si, it requires much thicker GaAs substrate. Thicker substrates increase cost, weight and requiring new processing equipment to accommodate the increase in thickness. Especially for space based applications like solar cells on satellites, etc., the GaAs on Si is a better choice than GaAs on "thicker" GaAs.

(4) Si wafers cost practically a little fraction of GaAs wafers. The process of epilayer growth is expensive, but for structures that inherently require epitaxial growth, i.e., heterostructures,

Table 1. 1 Properties of Si and GaAs at 300 K.

	Si	GaAs
Mobility (cm²/V-s)	1500	8500
Electron arrival velocity (cm/s)	1×10^7	3×10^7
Bandgap energy (eV)	1.12	1.424
Thermal conductivity (W/K-cm)	1.5	0.46
Thermal expansion coefficient (°C⁻¹)	2.6×10^{-6}	6.86×10^{-6}
Density (g/cm³)	2.328	5.32
Lattice constant (nm)	0.5431	0.5653
Intrinsic resistivity (Ω-cm)	2.3×10^5	1×10^8
Dislocation density (cm⁻²)	0	$<10^5$
Band structure	Indriect	Driect
Crystal structure	Diamond	Zincblend
Mechanical strength	Strong	Fragile
Heterojunction	Difficult	Easy
Wafer size (inches)	≤ 12	≤ 4
Price	Cheap	Expensive

MODFETs, quantum wells, etc., the extra effort involved in the growth of GaAs on Si is insignificant[14].

As a result of the impressive progress in epitaxial growth of GaAs on Si, GaAs devices and GaAs buffered devices grown on Si substrates have been successfully demonstrated. All the main electrical and optical GaAs devices have been grown on Si substrate, and their properties have been compared with the same devices grown on GaAs substrates[15-22]. Also, in the field of photovoltaics, GaAs on Si is useful either as single-junction solar cells, or as a technology for making very high-efficiency multi-junction tandem solar cells, where Si is used as the bottom cell and a GaAs-related compound semiconductor material such as AlGaAs as the top cell material[23,24]. However, the performance characteristics of these device structures still can not approach those on GaAs substrates, because Si is not a promising substrate for single-crystal growth of GaAs. The important ones among the many are initial growth of polar (GaAs) semiconductor on a nonpolar (Si) one, 4% lattice mismatch between silicon and gallium arsenide, and 55% mismatch in thermal expansion coefficients. The large lattice mismatch between these two materials results in the formation of a high concentration of defects (dislocations, micro twins, and stacking faults) near the GaAs heterointerface. The thermal mismatch creates further defects, causes wafer bowing and if the epilayer is thick enough, crack of the epilayer may occur. The presences of the defects, propagating through the GaAs epilayer, have a detrimental effect on the optoelectronic properties of any devices fabricated on GaAs/Si. For the GaAs solar cells grown on Si substrates, which are minority carrier devices and the usual solar cell structures are vertical type, the effect of dislocations particularly thick base layer on the substrate side is crucial, because the dislocations act as non radiative recombination centers and reduce the minority carrier lifetime[25]. Many studies have therefore been conducted to investigate the nature of the defects present. Various methods have also been proposed to both decrease the defect density and to suppress those present in a region close to the heterointerface, but the dislocation density is still much higher ($\sim 10^6$ cm^{-2}) than that is acceptable ($\sim 10^4$ cm^{-2}) for practical applications. Therefore, passivation of the electrical activity of these

Table 1. 2 Problems in GaAs Grown on Si Substrate

PROBLEM	CAUSE	SOLUTION
Antiphase domains (APDS)	Difference in crystalline structure (Polar/nonpolar)	Using tilted substrate
Misfit dislocation	Lattice mismatch	Using strained layer superlattice
Propagating dislocation	Three-dimensional initial nucleation, stress	Thermal annealing, using buffer layer, light. electron-beam irradiation
Stress	Mismatch in thermal expansion coefficients	Patterning, low-temperature growth
Surface morphology	Lattice defects, stress, three-dimensional initial nucleation	Surface polishing
Si autodoping	Si reaction with source gas	SiO_2 back and side coverage

defects is becoming a very essential issue.

In the next section, the problems encountered in achieving high quality GaAs on Si and several approaches to overcome these problems, are described. Section 1. 3 is devoted to characterization of semiconductors with hydrogen incorporated. The purpose and organization of this dissertation are then described in section 1. 4.

1. 2 GaAs epitaxial growth on Si

As summarized in Table 1. 2, the most important problems encountered in GaAs growth on Si and the main solutions proposed are as follows:

(1) Antiphase domains (APDs)

Nonpolar semiconductors consist of a single atom, and both sub-lattices of the diamond structure are occupied by the same atom. Polar semiconductor consist of two or more atoms, and each atom resides in a particular sublattice. Epitaxial growth of polar semiconductors on nonpolar substrates often leads to structure defects known as antiphase boundaries (APBs) due to constituent atoms occupying incorrect sublattice. When epilayers of GaAs are grown on Si (100), Ga and As atoms can exhibit ambiguity in choosing lattice sites. The lattice sites on the (100) planes are indistinguishable and so there are no preferential nucleation sites for Ga and As. This results in arsenic-arsenic or gallium-gallium bonds. Such arsenic-arsenic or gallium-gallium bonds boundaries are called anti-phase boundaries (APBs) or anti-phase domains (APDs). In general, these boundaries can be highly charged and degrade overall mobilities, and are therefore deleterious to device performance. It was found that the APDs can be self-annihilated by using a disoriented Si (100) substrate with odd atomic layer height steps[26]. Now it is almost not a problem in growth of GaAs on Si.

(2) Residual free electron concentration due to Si auto-doping

MOCVD grown unintentionally doped GaAs layers on Si usually have n-type conductivity, with

carrier concentration typically in the $10^{16} - 10^{17}$ cm^{-3} range due to Si auto-doping[27]. Some diffusion models are proposed to explain the Si autodoping phenomenon. For example, Freundlish et al. have proposed the "pipe" diffusion along the high density of threading dislocations, as they found Si diffused in GaAs on Si is concentrated in channels with Si cores that correlated with the dislocation density; Nozaki et al. have proposed gas phase transport of Si atoms to the growing layer, etc.[28]. It is very difficult to grow high resistivity undoped GaAs epilayers on Si using the MOCVD technique. This becomes a crucial problem in the fabrication of GaAs metal-semiconductor field-effect transistors (MESFET's) and high electron mobility transistors (HEMT's), because the pinch-off characteristics of GaAs MESFET's and HEMT's grown on Si by MOCVD are often degraded by high carrier concentration in undoped layers[29]. It is found that the SiO$_2$ back coating is useful in growing high resistivity undoped GaAs layers on Si using the MOCVD technique even at a higher growth temperature[30]. But this back coating method increase the complexity of the growth procedure.

(3) Dislocation due to lattice mismatch

The lattice constant of GaAs is larger than that of Si. This mismatch causes strain on the lattice of epilayer. The energy associated with the strain is proportional to the thickness of the epilayer. When the epilayers are thick, the associated strain energy is larger than the misfit dislocation energy. Then dislocations are generated to relieve the strain energy. There are two predominant types of dislocations[31], one whose Burgers vector is contained at the GaAs/Si interface (type I), while the other inclines (type II). The type I misfit dislocations are of the pure edge type; their Burgers vectors and dislocation lines are perpendicular to each other. Type II misfit dislocations are of the mixed type; their Burgers vectors are inclined by 60° to dislocation lines and at 45° to the plane of the substrate. The type II dislocations are undesirable for two reasons: They are less effective in accommodating misfit and they can more easily move on slip planes and generate threading dislocations. The use of a tilted substrate preferentially induces the formation of type I dislocations. Additionally, one can prevent many threading dislocations from reaching the surface by using a

strained-layer supperlattice (SLS)[32-34]. The strain of SLS will bend the threading dislocations and reduce the dislocation density near the free surface. In addition, some other attempts are done to eliminate the threading dislocations density in GaAs active layer, such as the growth of the low temperature buffer layer grown at 400-600°C (two-step growth)[35]; growing thick GaAs epilayer[36] and using in-situ thermal cycle annealing (TCA)[37], etc..

(4) Dislocation and residual stress due to thermal coefficient mismatch

Basically, there are two main considerations concerning the origin of residual dislocation density in GaAs on Si. One is the large lattice mismatch between a GaAs and Si crystal, as described in above. The other is the large difference in the thermal expansion coefficient between them, or in other words, the thermal stress in a GaAs crystal, since the thermal stress in GaAs can reach more than 10^9 dyn/cm^2 at room temperature. It was found that the quality of GaAs is very high at the growth temperature and the etch pit density is only 10^4 cm^{-2}. However, when the crystal is cooled down, the etch pit density increases to about 10^6 or 10^7 cm^{-2}. This proved that the crystal quality of GaAs on Si is determined by the thermal stress produced during cooling and not by lattice mismatch[38]. In addition, the stress applied to the active layer in the laser structure reduces the lifetime[39]. To minimize the stress, selective growth[40], patterning the epilayer after growth[41] and high pressure annealing the epilayer[42], etc. have been proposed, but the residual stress in GaAs on Si is still difficulty to be removed completely.

As discussed in above, numerous dislocation reduction techniques have been attempted, however, none of these techniques has successfully reduced the dislocation density to below 1×10^4 cm^{-2}, where the material becomes really useful for practical applications. This motivate the interest for finding passivation techniques to inactivate the defects, including shallow and deep defects in GaAs on Si.

1. 3 Hydrogen in III-V semiconductors

Since the discovery of hydrogen passivation of acceptors and donors in GaAs, a great deal of research has been performed on hydrogen in compound semiconductors[43]. Usually, hydrogen is not an electrically active substitutional impurity in III-V semiconductors. That is, if hydrogen were incorporated in an intrinsic, undoped semiconductor, the electronic properties of the semiconductor would not be changed by the hydrogen incorporation. However, hydrogen strongly interacts with shallow donor and acceptor impurities and neutralized them[44]. In addition, the passivation of deep levels by atomic hydrogen is also a very effective and relatively thermally stable phenomenon[45-49]. Consequently, the electronic properties of III-V semiconductors are dramatically changed by hydrogen incorporation[50-53]. There are a number of methods available for introducing hydrogen into semiconductors. These may be classed into two groups - methods in which the hydrogen is introduced in an intentional and controlled manner, such as exposure to a plasma or by direct ion implantation, and those methods in which hydrogen is injected into the semiconductor in an uncontrolled, and often unintentional, way during the crystal growth, cleaning and device fabrication processes. As the hydrogen implantation, which is frequently termed proton implantation, creates defects along their implantation path by high-energy protons, the semiconductor thus becomes highly destroyed. On the otherwise, this mechanism is irrelevant if hydrogen is introduced by diffusion, such as plasma exposure[54,55].

1. 3. 1 Hydrogen plasma exposure

Hydrogen can be incorporated from adsorbed hydrogen on the surface into the bulk of the semiconductors by means of diffusion. Atomic (H) and molecular (H_2) hydrogen surface populations can be generated by a low pressure (0.1 to 100 Torr) glow discharge, direct current (dc) or radio-frequency (RF) plasma reactor. Among them, the most common method is exposure
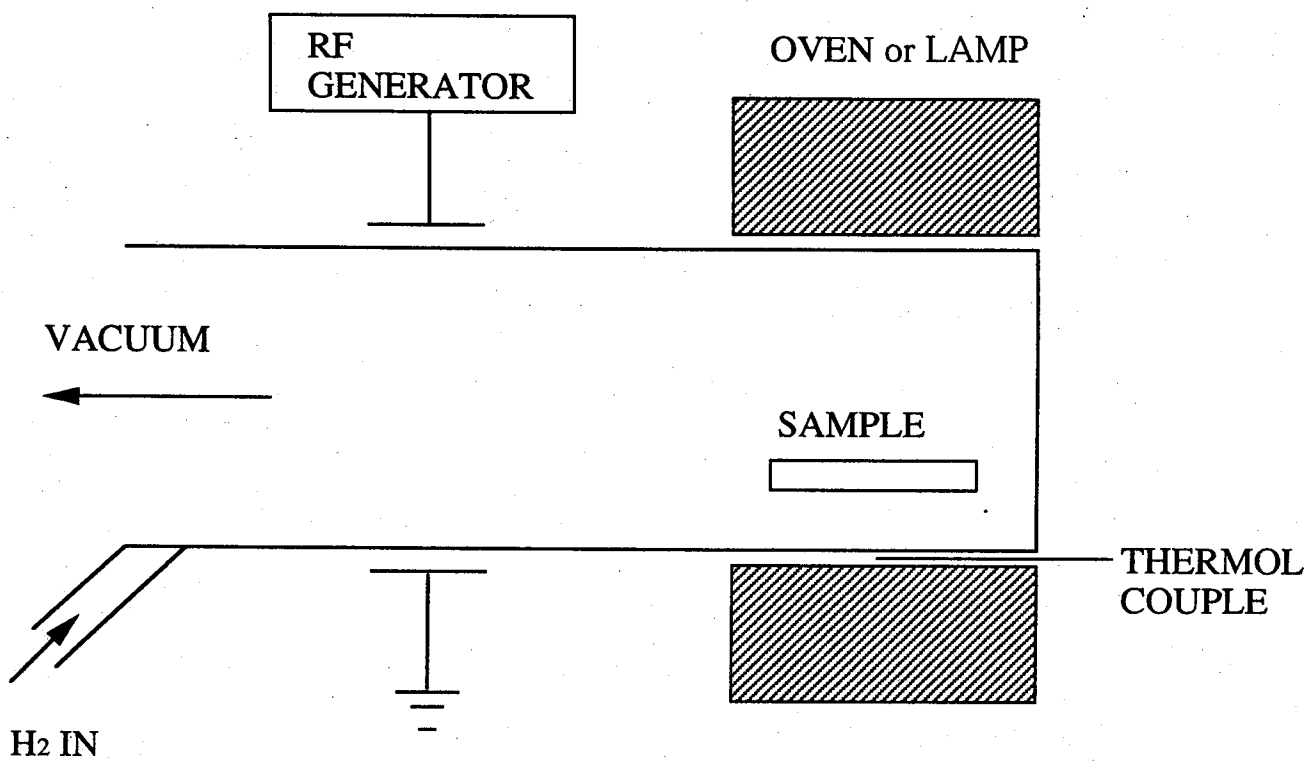
Fig. 1. 1 Schematic representation of radio-frequency (RF) plasma hdrogenation system.

to a radio-frequency plasma reactor in which the sample is mounted 'down stream'. Figure 1. 1 shows one of a schematics of the arrangement used for RF-plasma exposure. The hydrogen plasma itself is excited by radio-frequency (13.56 MHz) power via a high frequency oscillator. The sample is heated typically to 100~400°C for a period of 0.5~3 h downstream in the plasma. This leads to hydrogen incorporation depth of anywhere from a few tenths of a micrometer to as much as 50 μm, depending on the material itself, and the doping density within it. The 'downstream' configuration of the sample implies that the sample is spatially removed from the radio-frequency generator, as seen in Fig. 1. 1. The 'downstream' formation effectively eliminates the direct bombardment damages. The lack of exposure to a high electric field also reduces the non-uniformity of the hydrogen surface population.

1. 3. 2 Passivation of shallow impurity level by atomic hydrogen

Much work on shallow impurity passivation have been performed in GaAs. It is now well established that all of the donor impurities (Si, Ge, Sn, S, Se and Te)[56-59] as well as all of the acceptor species (Be, Mg, Cd, Zn and C)[60-63] can be passivated by association with atomic hydrogen. The commonly accepted atomic model for donor passivation by hydrogen is shown in Fig. 1. 2. The substitutional Si atom is fourfold coordinated. The hydrogen is in an interstitial (AB) site and bound to the Si donor. One of the original Si-As bonds is broken leaving the As atoms opposite the hydrogen atom with a lone pair of electrons. The electron count of the Si-H complex yields that the complex is neutral, i.e. H has passivated the donor atom. Although the atomic model shown in Fig. 1. 2 has been verified for Si-H in GaAs, it is generally assumed that the model also applies to other donor impurities in III-V semiconductors. In addition, the activation energy for the recovery of the electrical activity, i.e. the dissociation of the donor-hydrogen bond, was found to be anti proportional to the donor-hydrogen bond strength. This interdependence between the recovery of the electrical activity of passivated donor and the donor-hydrogen bond strength further supports
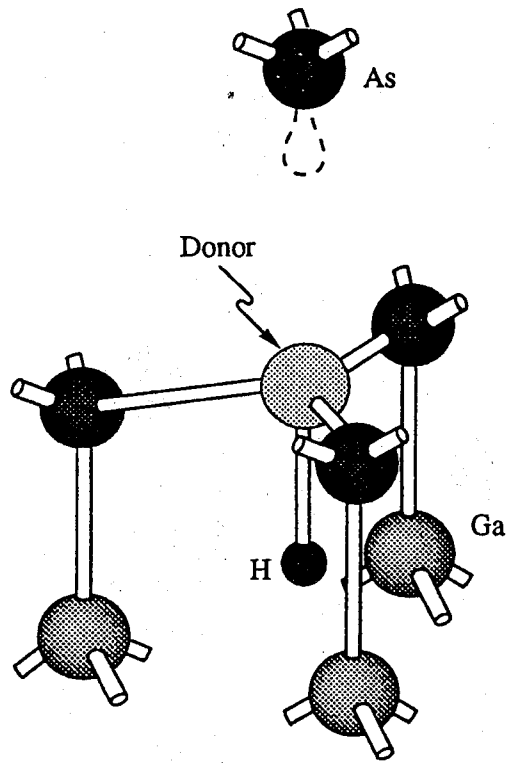
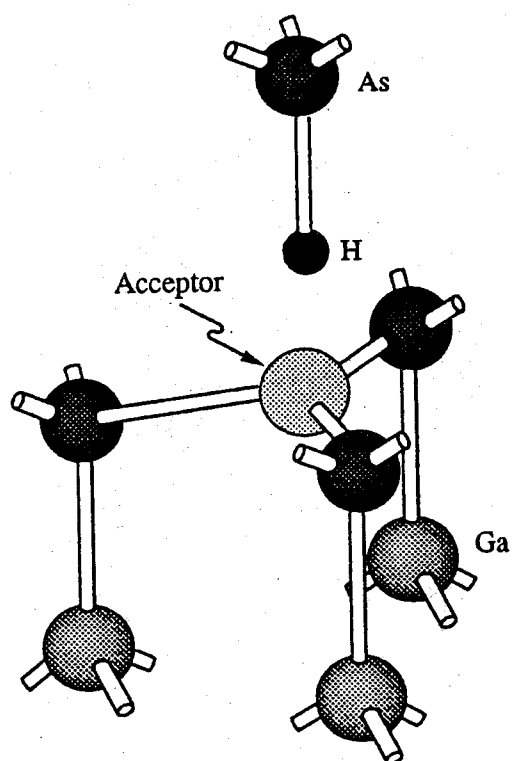Fig. 1. 2 Schematic representation of the group-IV donor-hydrogen complex with hydrogen.

Fig. 1. 3 Schematic representation of the group-II acceptor-hydrogen complex with hydrogen.

the model in which the hydrogen is directly bound to the donor atom. Hydrogen diffuses out of hydrogenated semiconductors, if the samples are heated to sufficiently high temperature, typically larger than 400°C. The original conductivity of hydrogenated samples can be restored by such an annealing cycle.

Hydrogen also passivates shallow acceptors in III-V semiconductors. The atomic model for the substitutional group-II acceptor is shown in Fig. 1. 3. The hydrogen atom is bound to an arsenic atom and occupies a BC-site between the acceptor impurity and the arsenic neighbors. The hydrogen atom thus satisfies the electron pair requirement of the arsenic neighbor. The entire hydrogen-impurity complex is neutral, i.e. hydrogen passivates the acceptor. To date, the atomic model shown in Fig. 1. 3 is generally assumed to apply to all substitutional group-II acceptors.

1. 3. 3 Passivation of deep level by hydrogen

The passivation of point defects-related deep levels by atomic hydrogen is a very effective and relatively thermally stable phenomenon; temperatures in excess of 400°C are normally required to reactivate the deep levels after it has been passivated. The mechanism of hydrogen passivation of point defect-related dangling bond is clear that the dangling bond gives rise to a state in the bandgap. The subsequent attachment of hydrogen to the dangling bond forms bonding and anti-bonding states, the bonding states being in the valence band and no longer electrically active, and the unoccupied antibonding states being in the conduction band and also electrically inactive[64]. Therefore in principle, the presence of an appropriate number of hydrogen can passivate these bonds. However, since the microstructure of all of the deep level in semiconductor is largely unknown, it is not clear if direct attachment of hydrogen to a dangling bond is the passivation mechanism in all cases.

Recently, dislocation passivation via hydrogenation has demonstrated considerable potential as an alternative approach for the reduction or elimination of the deleterious electronic effects of

dislocations in lattice-mismatched heteroepitaxial structures including InP/GaAs[65], GaAs/Si[66], InGaAs/GaAs[67], and GaAs/InP[68], with thermal stabilities well in excess of that observed for point defect passivation. Such an approach would therefore rely on the electrical control of dislocation induced states, rather than the physical or micro structural control of the dislocation themselves. It demonstrates significant changes in the mechanism of charge trapping at dislocations after exposure to hydrogen plasma, which is coupled to the observed reduction in overall trap density. As described in above, the effects of hydrogen on the electrical activity of both shallow dopants and deep level due to point defects have been well documented. In contrast, while strong dislocation passivation by hydrogenation has been observed in heteroepitaxial systems, little else is known about the electrical properties of the hydrogen-dislocation complex, the kinetics of their formation and dissociation, or about any micro structural changes in the dislocation network due to hydrogen plasma exposure.

1. 3. 4 Hydrogen plasma-induced damages on GaAs surface

Although the 'downstream' configuration of the sample during the RF plasma exposure eliminates the direct bombardment damages on GaAs surface to some extent, the hydrogen plasma exposure may still creates deep compensating centers through the impact of energetic H^+ ions on the GaAs surface. The atomic displacement threshold in GaAs is ~ 30 eV, and typical ion energies in a conventional 13.56 MHz plasma are of order 200 eV. In this phenomenon the carrier density will decrease, but the carrier mobility will also decrease because of the additional compensation[69-71]. In addition, a preferential loss of As from the immediate surface region may be induced, because of the vastly different volatilities of arsenic and gallium hydrides[72]. The change in near-surface stoichiomrtry may alter the surface recombination velocity and Fermi-level pinning position. The plasma-induced damages dramatically negate the beneficial effects of H plasma passivation. It needs to minimize the plasma-induced damages in practical hydrogen plasma passivation process[73].

1. 4 Purpose and Organization of dissertation

For further improving the optical and electrical properties of the GaAs on Si grown by MOCVD for practical application, it needs to reduce the dislocation density in GaAs on Si epilayer as low as 10^4 cm^{-2}. Since the reduction of the density of threading dislocations to such a low level in GaAs on Si epilayers can't be simply realized by present growth method and techniques, passivation of the electrical activities of the dislocation related recombination centers is becoming very essential. Passivation of a wide range of shallow and deep impurities in GaAs, AlGaAs and AlGaAs/GaAs quantum well grown on Si by hydrogen (H) plasma exposure is intensively investigated in this study. It is found the addition of H in GaAs on Si helps to reduce the density of electrically active dangling bonds and passivate the electrical activity of most defects and impurity states, and the beneficial effects of the H plasma passivation are stable under usual device processing temperature (below 450°C). However, exposure to H plasma induces surface roughness and depletion of As from the GaAs surface, and the damage effects exceed the H passivation effects, sometimes. It needs to recover these damages without removing the beneficial effects of the hydrogen incorporation. The annealing in AsH$_3$ ambient after plasma exposure is helpful to recover the plasma-induced damages, and and adding PH$_3$ into H$_2$ source during the plasma exposure seems to suppress the generation of damages in the surface region and phophidization of the GaAs surface. The successful improvement of the characteristics of solar cells and Schottky diodes on the GaAs on Si by hydrogen plasma exposure makes sense as a first step in improving the properties of GaAs on Si devices by hydrogen plasma passivation techniques.

This dissertation is divided in to seven chapters, each of which is summarized below.

In chapter 2, the effects of plasma hydrogenation (H$_2$) on the optical and electrical properties of GaAs epitaxial layers (GaAs/Si) and AlGaAs/GaAs multiquantum well (MQW) grown on Si substrate are described. It is shown that a significant improvement in the optical and electrical properties of GaAs epilayers on Si is realized by H plasma passivation. It is also found that the H

plasma exposure significantly increased photoluminescence emission intensity and quantum efficiency at the AlGaAs/GaAs multi quantum well (MQW) grown on Si. These improvements are attributed to the passivation of the defects-related, such as dislocation, deep recombination centers in GaAs on Si.

In chapter 3, a detailed study of H_2 plasma exposure and annealing effects on the photovoltaic properties of GaAs solar cells grown on Si substrates. It is found that the conversion efficiency of the H_2 plasma passivated GaAs on Si solar cell is improved, and this improvement is stable under the 450°C annealing in AsH_3 ambient, which is the usual solar cell process temperature. This can be attributed to the increased minority carrier lifetime by the H passivation of the defect-related non radiative recombination centers in GaAs/Si.

In chapter 4, the hydrogen plasma passivation effects on photoluminescence (PL) characteristics of $Al_{0.13}Ga_{0.87}As$ epilayer grown on a Si substrate are investigated. It has been found that the 4.2 K PL intensity increases for the H plasma passivated sample with reduced carrier concentration, owing to the passivation of nonradiative recombination centers. The passivation of residual impurity carbon in the $Al_{0.13}Ga_{0.87}As$ on Si epilayer was directly confirmed based on the observations of the 4.2 K PL spectra of the samples. The passivation effect is found to persist even following annealing at 450°C for 10 min.

In chapter 5, the PH_3+H_2 plasma passivation effects on MOCVD-grown GaAs on Si were studied in detail. It showed that both the surface phosphidization and defect hydrogenation can be realized simultaneously. The optical and electrical properties of GaAs/Si were effectively improved. In addition, the surface phosphidization suppresses the hydrogen plasma induced damages due to the phosphorus atoms incorporation. As a result, the PH_3+H_2 plasma exposure results in very high open circuit voltage V_{oc} (0.93 V) and fill factor FF (80.9%) for GaAs/Si solar cell, and the conversion efficiency (E_{ff}) is significantly improved.

In chapter 6, PH_3+H_2 passivation effects on GaAs Schottky diodes grown on Si substrates are investigated. It is found that the PH_3 plasma passivation has a merit to realize the phosphidization

and hydrogenation effect on GaAs on Si simultaneously by the P and H atoms incorporation. As a result, the characteristics of Schottky diode made on GaAs on Si are improved. In addition, the PH_3 plasma exposed GaAs Schottky diodes on Si have an excellent thermal stability compared to that of the H_2 plasma exposed diodes.

In chapter 7, the summary of this research is described. The scope for future work is also presented.

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