Structural and Optical Properties of GaSb Films Grown on AlSb/Si (100) by Insertion of a Thin GaSb Interlayer Grown at a Low Temperature

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We have investigated the structural and the optical properties of GaSb films with a thin AlSb buffer layer and a GaSb interlayer grown on Si (100) substrates by using molecular beam epitaxy. Reflection high-energy electron diffraction and atomic force microscopy measurements of the thin AlSb buffer layers showed that the surface had uniformly-sized quantum dots with a low defect density. The surface roughness of a GaSb film with a thin GaSb interlayer grown at a low temperature was decreased by a factor of about 5 compared with the roughness of the GaSb film without the thin GaSb interlayer. In addition, double-crystal X-ray diffraction and photoluminescence results showed that the structural and the optical properties of the GaSb layer with the GaSb interlayer were improved significantly. We suggest that the significant reduction of the dislocation density in the GaSb film was due to the dislocations being prevented from propagating into the GaSb overlayer by the thin GaSb interlayer.

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I. INTRODUCTION

Lattice-mismatched epitaxy of Sb-based materials on Si substrates has attracted considerable attention due to numerous advances in optoelectronic devices, such as laser diodes [1–3], detectors [4], and transistors [5]. Therefore, much effort has been devoted to the growth of Sb-based layers of high crystalline quality on Si substrates.

In spite of various performance advantages of III-Sb-based compound semiconductors on Si substrates, growth of high-quality III-Sb layers on Si substrates is problematic because the large difference in lattice constants between the epitaxial layer and the substrate leads to the generation of stress and dislocations in the epitaxial film. In other words, the thickness of heteroepitaxial film for device applications is limited because the strain energy in the film is released by the formation of misfit and threading dislocations beyond a critical thickness [6]. One approach taken to overcome these problems is to employ buffer layers such as Sb-terminated layers, buffer layers grown at low temperature (LT), and compositionally graded buffer layers between III-Sb layers and substrates. In particular, a growth method for GaSb layers on Si (100) substrates involving the introduction of an AlSb initiation layer has been demonstrated by several research groups [7–11]. However, there are few reports related to the growth of GaSb films using a thin GaSb interlayer (buffer) grown on a thin AlSb nucleation layer at LT. Also, the effects of the thin buffer layer on the crystalline quality of the heteroepitaxial film have not been deeply considered from a microstructural point of view. The insertion of a thin GaSb layer grown at LT may be effective in enhancing the films quality by suppression of the formation of antiphase domains (APDs) in the GaSb film through the formation of a 3-dimensional (3D) layer.

In the work reported here, we studied the structural and the optical properties of GaSb films on Si (100) substrates with insertion of a thin GaSb interlayer grown at LT. A thin GaSb interlayer (8 nm) was deposited on a 3D AlSb nucleation layer on Si (100) prior to the growth of the GaSb layer. The LT GaSb buffer layer

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greatly enhanced the structural and the optical properties of the GaSb films. We recognized that the GaSb films grown at relatively lower temperatures are n-type while the films grown at higher temperatures are p-type. Thus, we also investigated the dependence of the structural properties of the unintentionally-doped GaSb films on growth temperature. The properties of the GaSb films were investigated using reflection high-energy electron diffraction (RHEED), atomic force microscopy (AFM), X-ray diffraction (XRD), and photoluminescence (PL) measurements.

II. EXPERIMENTS

The GaSb films were grown on p-type Si (100) substrates by using a conventional solid source molecular beam epitaxy (MBE, Riber 32). The substrate temperature was monitored with an infrared pyrometer. Prior to the films growth, the surface of the Si substrate was hydrogen passivated in an HF solution. The hydrogen was then removed by annealing the substrate at temperatures up to 540 °C in the loading chamber. The remnants of the oxide layer on the Si surface were thermally removed at 850 °C in the growth chamber. The removal of the oxide was verified by examination of the RHEED pattern. The substrate temperature was then lowered and stabilized at 540 °C followed by soaking for 5 minutes in an Sb4 overpressure. The Si surface maintained a (2 × 2) reconstructed structure during the soaking.

Three kinds of undoped 1-µm GaSb epilayers were prepared for this study. Their specifications and structures are illustrated schematically in Fig. 1. AlSb nucleation layers of 3.5 nm in thickness were grown on the Si substrates at 540 °C with a Sb/Al flux ratio of 10 under the same conditions. Following this, undoped GaSb layers of 1 µm, thickness at a Sb/Ga flux ratio of 8 were grown at LT (480 °C) and at high temperature (HT, 580 °C) for samples A and B respectively. Sample C, as specified in Fig. 1(c), was grown with a different type of buffer layer. After a thin AlSb nucleation layer had been deposited, a GaSb buffer layer of 8 nm was grown at LT with an Sb/Ga flux ratio of 8, then, a GaSb layer of 1 µm in thickness was grown at 580 °C. The growth rates of AlSb and GaSb were set to 0.3 and 0.7 monolayer (ML)/sec, respectively. During the growth process, the sample surfaces were monitored in situ by using RHEED. AFM measurements were performed to measure the roughness of each surface. The structural and the optical properties of the grown GaSb films were measured by using XRD and PL.

III. RESULTS AND DISCUSSION

First, we used RHEED and AFM measurements to investigate the growth mode of the AlSb layers employed as buffers for the growth of GaSb films. Figure 2 shows the variation of the lattice mismatch between AlSb and Si during the deposition of the AlSb buffer. The values were deduced from the RHEED patterns of the film grown for 24 s at 540 °C. Also, the insets of Fig. 2 display the RHEED pattern and AFM images obtained after deposition of 1.2 nm (3.8 ML) and 3.5 nm. The lattice parameter, which is the reciprocal of the
in-plane lattice parameter, between the (10) and the (10) diffraction streaks can be determined directly from the RHEED patterns. The variation of the surface lattice parameter with growth time should give some insight into mechanism of strain relaxation in the AlSb films. We note that the process of the AlSb-island formation can be divided into three growth regions: the formation of an AlSb wetting layer (WL), the formation of AlSb islands on the WL, and the growth of the islands by a ripening process in strain relaxation [13]. The AlSb WL is formed from the initial nucleation of AlSb, which was observed for AlSb deposition times up to \( \sim 12.5 \) s, corresponding to a deposition of a 0.3-ML film (marked with an arrow in Fig. 2). Because the initial AlSb layer experiences a compressive strain in a metastable growth process, the lattice constant of the AlSb WL changes slightly due to elastic distortion of the AlSb lattice during the layer-by-layer growth. A transition of the AlSb growth mode from a 2-dimensional layer structure to an island structure occurs when the thickness of the AlSb deposition layer is increased to 0.3 ML. The initial formation of the AlSb islands on the AlSb WL result in an abrupt increase in the lattice constant. Further deposition of AlSb up to a deposition time of \( \sim 16 \) s, at which the film’s thickness reaches 1.4 MLs, led to an increase in the size of the initial islands and to an increase in the lattice constant in order to permit relaxation of the compressive strain in the AlSb layer. However, during depositions lasting longer than \( \sim 16 \) s, the lattice mismatch becomes essentially constant. In fact, the obtained value of the lattice mismatch of \( \sim 13\% \) is lower than what is well-known to occur in bulk AlSb. Although the result is slightly different from the theoretical result reported in the literature [14], the continuous deposition of AlSb will lead to lattice constants in the growth structures containing AlSb islands that are similar to the lattice constants of bulk AlSb. Also, we noted that the difference in lattice constants arising in the island between the island AlSb structures and bulk AlSb correlates closely with growth parameters such as the growth temperature, growth rate, and V/III flux ratio.

We also used AFM measurements to investigate the surface structures of GaSb layers grown with different growth temperatures and thicknesses on AlSb buffer layers. The insets of the graph of Fig. 3(a) show the AFM images of the 5-nm-thick GaSb films grown at 480, 500, and 580 °C. The surfaces of all films display a non-uniform distribution of irregular GaSb islands. The average height of the islands increased linearly while the density decreased with increasing growth temperature, as shown in the plot of Fig 3(a). The average height of the islands grown at 580 °C is about 67 nm, which is about 2 and 3 times higher than the heights attained at lower growth temperatures of 500 °C and 480 °C respectively. The size increase with increasing growth temperature is caused by the formation of larger islands through coarsening processes, in which smaller islands grow larger. The driving force of the processes might result from a minimization of the surface and interface energies of the islands and from an enhanced migration of GaSb at higher temperatures. In addition, we examined the surface structures of the GaSb films grown as a function of thickness. Figures 3(b) - (d) show AFM images of the films with 10-, 18-, and 23-nm thickness grown on AlSb (3.5 nm)/Si(100) at 500 °C. For a 10-nm-thick film, the surface displays a distribution of round 3D islands of various sizes (Fig. 3(b)). The GaSb islands might have been formed due to the island-structured surface of the AlSb buffer layer and/or to the strain energy induced by the lattice mismatch between the GaSb layer and the AlSb layer. For an 18-nm-thick film, we could observe that the islands grew preferentially by coalescence processes in which nearby islands collapse into each other to form larger islands (Fig. 3(c)). The continuous island coalescence leads to the initiation of inhomogeneous epitaxial layer growth. For a further increase in thickness up to of 23 nm, we could observe an initial transition of the islands into planar growth of the GaSb layer (Fig. 3(d)). However, twin boundaries are observed to remain in the layer before the formation of complete layer structures.

We carried out further investigations of the surface structures of thicker GaSb films (1-µm thickness) grown under different growth conditions. Samples A and B were grown at 480 °C and 580 °C respectively (Figs. 4(a) and (b)). Alternatively, sample C was grown by using a two-step growth process, such as LT and HT growth, on which a 1-µm GaSb film was grown at 580 °C on an 8-nm GaSb interlayer grown at a lower temperature of 480 °C (Fig. 4(c)). Sample A has a rough, stepped surface morphology whilst sample B shows a smooth, mirror-like
surface over the partial wafer. Nevertheless, there are some twin boundaries on the surface. The roughnesses of both samples A and B were greatly reduced compared with that of thinner films, as shown in Fig. 3. The root-mean-square (RMS) values of the surface roughnesses of samples A and B are about 8 and 7 nm, respectively. In contrast, sample C with a thin GaSb interlayer grown at LT shows a smoother surface and fewer twin boundaries than samples A and B, which do not have the thin GaSb interlayer. The RMS value of sample C is about 1.5 nm, which is much smaller than those of samples A and B, and quantitatively, the RMS value of sample C is 20% of the RMS value of sample A. We can conclude that the surface structures of GaSb films can be improved considerably by using growth at a relatively higher temperature and by inserting of a thin interlayer grown at a lower temperature with the same V/III flux ratio as was employed in experiments reported here.

In order to compare the structural and the optical properties of samples A, B, and C, we performed XRD and PL measurements. Figure 5 shows the high-resolution XRD rocking curves of the samples. The main peak (14384 arcsec to Si (100)) corresponds to the (004) plane of the GaSb layer. The width of the peak from sample A is relatively broad with a full width at half maximum (FWHM) of 781 arcsec (Fig. 5(a)). This broad peak reflects the fact that the GaSb layer grown at LT has more defects induced by the large lattice mismatch between the GaSb epilayer and the Si substrate. The FWHM of the peak was reduced to 563 arcsec for the sample B grown at HT (Fig. 5(b)). Thus, HT growth of the GaSb film is more effective in enhancing of the crystalline quality than LT growth. In addition, the FWHM was further reduced to 460 arcsec for sample C, which was grown at HT with a LT growth interlayer (Fig. 5(c)). Namely, sample C has the highest crystalline quality among the GaSb films grown in this experiment. These results are consistent with the AFM images in Fig. 4. Conclusively, the presence of a thin LT GaSb interlayer is obviously useful for improving the crystalline quality of GaSb films on Si substrates.

Figure 6 shows the PL spectra obtained at 15 K with a fixed excitation power of 20 mW for the three samples. From Hall measurements, we recognize that samples B and C, grown at HT, are p-type while sample A grown at LT is n-type. Although we did not observe the emission peaks related to the free and bound excitons in the PL measurements at this temperature, the peak positions (emission energies) varied with the growth conditions. The PL spectrum of sample A does not show luminescence peaks, as shown in Fig. 6(a). This means that the existence of many defects in the film leads to non-luminescent radiation. However, for sample B, two main emission peaks are observed, 0.778 eV and 0.739 eV. The
peak at 0.778 eV is ascribed to the well-known transition of donor-acceptor pairs (DAP) while the peak at 0.739 eV may be due to deep acceptor-related transitions. According to the literature, the peak of the DAP is associated with the A-PL band [15] related to the native double-acceptor complex of $V_{Ga}GaSb$ [16]. For sample C, new peaks were observed at 0.768 eV and 0.751 eV which are related to the DAP of a shallow and a deep, different from the emission peaks observed at 0.778 eV and 0.739 eV for sample B, as shown in Fig. 6(c). Also, the peak intensity increased abruptly in comparison with that obtained from samples A and B. Even though the detailed origins of the two peaks are not yet been identified, the remarkable increase in the intensity may be explained by a decrease in the density of non-radiative recombination centers. The defects in the films, such as APDs, which play important roles as donors and/or acceptors, generate non-radiative recombination centers. The reduction in the defect density, along with the enhanced crystalline quality, may lead to a decrease in non-radiative recombination centers. Evidence for this was provided by the AFM and XRD measurements shown in Figs. 4 and 5. Other evidence was found in the variation of the intensities and the positions of both peaks in the PL measurements as a function of temperature. The intensities of both peaks decreased rapidly whilst the positions of the peaks shifted to lower energy with increasing PL measurement temperatures (not shown). This behavior is similar to that of the Y-band in ZnSe [17] and that of the DAP band in the CdTe/GaAs heterostructure grown by MBE [18]. Thus, we can conclude that the structural and the optical properties of GaSb films are improved considerably by insertion of a thin GaSb buffer grown at LT. Also, the two-step growth processes can be employed to enhance the crystalline quality of III-V materials films grown on Si substrates.

IV. CONCLUSIONS

We have demonstrated that an enhancement of the structural properties of a GaSb layer can be realized with the introduction of a relatively thin GaSb interlayer and AlSb buffer layer. The RMS value of the surface roughness is decreased by a factor of about 5 compared with the roughness of the thin GaSb interlayer grown at LT. Also, from the XRD and PL measurements, we confirmed that the defect densities and the non-radiative recombination centers were decreased. From the results, we know that many of the dislocations in the GaSb buffer layer are prevented from propagating into the overlying GaSb by the GaSb interlayer grown at LT, leading to a significant reduction in the dislocation density in the layer. These observations can help improve our understanding of the structural and the optical properties for growing Sb-based materials on Si substrate for promising applications in devices.

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