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(Received 28 October 2012; accepted 19 November 2012; published online 20 December 2012)

Intense terahertz (THz) electromagnetic wave emission was observed in undoped GaAs thin films deposited on (100) n-GaSb substrates via molecular beam epitaxy. GaAs/n-GaSb heterostructures were found to be viable THz sources having signal amplitude 75% that of bulk p-InAs. The GaAs films were grown by interruption method during the growth initiation and using various metamorphic buffer layers. Reciprocal space maps revealed that the GaAs epilayers are tensile relaxed. Defects at the i-GaAs/n-GaSb interface were confirmed by scanning electron microscope images. Band calculations were performed to infer the depletion region and electric field at the i-GaAs/n-GaSb and the air-GaAs interfaces. However, the resulting band calculations were found to be insufficient to explain the THz emission. The enhanced THz emission is currently attributed to a piezoelectric field induced by incoherent strain and defects. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4770267]

I. INTRODUCTION

There has been considerable interest in the molecular beam epitaxy (MBE) growth of GaAs on GaSb substrates to realize vertical cavity light emitting diode[1] and quantum dots emitting at 1.85 μm.[2] In order to contain the strain in the interface and to reduce the defects due to the high lattice mismatch ($\sim$8%), growth techniques such as interfacial misfit arrays (IMF) have been employed.[1,3] In this paper, we report on the successful MBE growth, structural quality, and terahertz (THz) emission of GaAs thin films on GaSb substrates. The GaAs/n-GaSb films are characterized by a THz signal that is as much as 75% of that of p-type InAs.

Among the semiconductor surfaces irradiated by femtosecond laser pulses, bulk p-InAs has been shown to be one of the most intense THz emitters.[4–7] However, recent works indicate that a judicious choice of layers of semiconductors may lead to new sources of THz emission with intensities that are comparable to that of bulk p-InAs. A study revealed that an i-GaAs/n-GaAs epilayer structure has a stronger THz signal than an n-GaAs due to an enhanced depletion width.[8] It was reported that a 70% enhancement from InAs/GaAs quantum dots interspersed in GaAs was attributed to the strain field.[9] Another study explained that the enhanced THz emission of undoped-GaAs/n-GaAs layer on a Si substrate was due to three regions of high electric field.[10]

The quality of the GaAs layers grown on GaSb was established by investigating their enhanced THz emission. In addition, semiconductor surface THz emitters are of interest due to their ease of use and wide applicability in THz spectroscopy. As such, GaAs/n-GaSb may prove promising as an unbiased, unfabricated semiconductor surface emitter.

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II. METHODOLOGY

The layers were grown in a Riber 32P MBE chamber equipped with Ga and tetrameric As cells. The (001) n-GaSb substrates were subjected to chemical preparation steps to achieve a clean epi-ready surface, consistent with previous studies.[11,12] The substrates were immersed in HCl for 1 min to etch off the residual oxides along with pre- and postdegreasing steps. To form a protective oxide layer, the substrates were submerged in 0.2% Br-methanol solution for 30 s. The substrates were outgassed in the loading chamber for 1 h at a nominal temperature of 350 °C. The oxide layer was thermally desorbed in the growth chamber under an As_4 flux of 5×10^{-6} Torr. From the reflection high-energy electron diffraction (RHEED) pattern, the onset of oxide removal occurred at 520 °C. The substrates were then heated to 530 °C for only 5 min to avoid surface decomposition. The pattern was characterized by a spotty diagram with diffused, overlaid chevrons, as shown in Fig. 1. After 2 min at a substrate temperature, $T_S = 530\,^{\circ}$C, the RHEED pattern turned into a (2×4) GaAs-like pattern with short streaks, indicating a smoother surface. The growth of GaAs commenced by opening the Ga shutter. Then for 5 min, a cycle of shutter sequences was implemented to allow the Ga atoms to migrate to favorable sites:[13] (i) Ga and As shutters were opened for 1 s then (ii) only As shutter was opened for 1 s. We observed that the (2×4) GaAs-like diffraction pattern persisted. Previous attempts without the use of the aforementioned method resulted to flaking off of the surface.

Two series of samples were grown. Sample I was comprised of a 0.5-μm-thick GaAs buffer, 40 periods of AlAs (20 Å)/GaAs (20 Å) superlattice buffer, and a 0.4-μm-thick GaAs. The SL was deposited to smoothen the surface.[14] In sample II, we used a buffer material that has a smaller lattice

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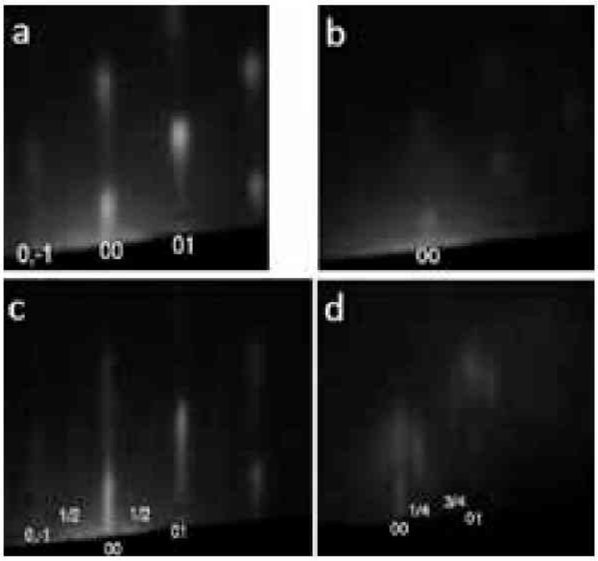

FIG. 1. RHEED (12 keV) images during oxide removal showing (a) patterns superimposed by chevrons for the [110] and (b) the [1$\bar{1}$0] azimuths, and the (c) (2 × 4)-like patterns for the [110] and (d) the [1$\bar{1}$0] azimuths after heating at 530 °C for 2 min.

mismatch with GaSb. A 4000-Å InAs buffer and 60 periods of InAs (20 Å)/GaAs (20 Å) superlattice were grown before the 1.5-μm-thick GaAs layer. Growth interruption was performed during the growth initiation of all the samples. For the case of sample II, where an InAs buffer was used, the shutter sequence was as follows: (i) In and As shutters were opened for 1 s, then (ii) only As shutter was opened for 1 s. The schematic diagrams of the layers are provided in Fig. 2.

The strain and structural quality of the samples were analyzed using x-ray reciprocal space maps (RSM). X-rays were produced by a Cu-target with $K_{\alpha 2} = 1.5443$ Å and $K_{\alpha 1} = 1.5405$ Å. Scanning electron microscopy (SEM) was used to reveal the surface morphology and interface quality of the layers.

A standard THz-time domain spectroscopy (THz-TDS) set-up was utilized to compare the THz emission of the samples with p-InAs and semi-insulating (SI) GaAs wafers in the reflection excitation geometry.[4] The excitation source was a mode-locked Ti:sapphire femtosecond laser emitting at 800 nm wavelength with a pulse width of ~100 fs at a repetition rate of ~80 MHz. The THz-TDS set-up employed an optically grated LT-GaAs photoconductive antenna detector.

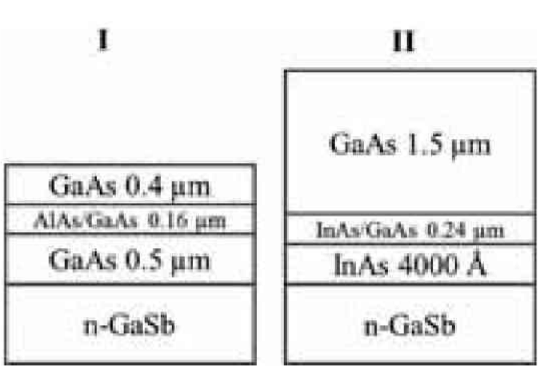

FIG. 2. Cross-sectional structures of the layers.

III. RESULTS AND DISCUSSION

Figure 3 presents the THz time-domain waveforms emitted from the undoped GaAs/n-GaSb samples in comparison with (100) p-InAs and SI GaAs wafers. The curves are offset in the time scale for ease of comparison. The p-InAs wafer still emitted the strongest THz signal (black trace), while a bare GaSb substrate showed the weakest (×6 magnified gray trace). The SI GaAs emission is less intense than that of p-InAs by approximately one order of magnitude. Similarly, earlier works reported a one order of magnitude difference between the THz emission intensity of bulk GaAs and bulk p-InAs.[7,10,15] However, our GaAs/n-GaSb layers demonstrated THz emission intensities comparable to p-InAs. The signal amplitude of sample I is half of that of p-InAs wafer. Sample II generated THz signal which was about 75% that of the p-InAs signal. The relatively stronger THz emission observed for sample II may be attributed to the InAs/GaAs buffer layer. The buffer layer for sample I is GaAs which is highly lattice-mismatched to the (100) GaSb. Notice that the intensity of GaAs/n-GaSb is ~10 times stronger than that of the GaSb substrate. Assuming that both GaSb and GaAs contribute to the THz emission from the GaAs/n-GaSb samples upon excitation, it could be expected that the THz emission intensity is a linear superposition of the THz emissions from bare GaSb and SI GaAs wafers.

At 800 nm pump, the penetration depth of the laser is ~1 μm for GaAs. Previous studies attributed the THz generation in GaAs to surface field acceleration caused by Fermi-level pinning.[4] It is generally believed that an electric field causes enhanced THz emission due to greater acceleration gradient. We calculated the depletion width in the GaAs surface by assuming a surface potential 0.72 eV due to band bending. The carrier concentration in the undoped GaAs layer was estimated to be $N_i = 1 \times 10^{15}\,\mathrm{cm}^{-3}$.[16] The surface electric field and depletion width were then calculated to be 14 kV/cm and 1000 nm, respectively. In contrast, an electric

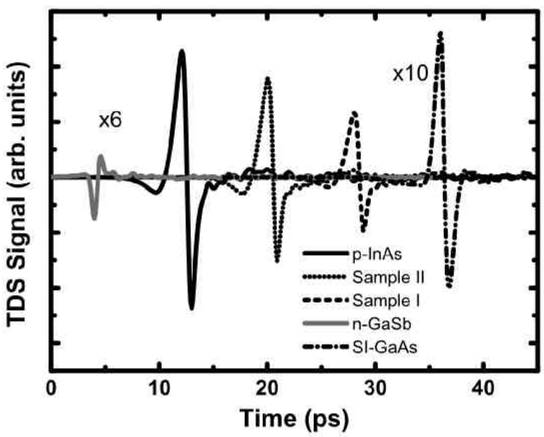

FIG. 3. THz waveforms emitted from undoped GaAs layers grown on n-GaSb are comparable with the THz emission of bulk p-InAs. Offset in the time scale was provided for ease of comparison.

field strength of 35 kV/cm for the i-GaAs/n-GaAs homostructure that yielded 10 times better THz emission than bulk n-GaAs was reported in Ref. 8. It may be noticed that the calculated depletion widths for our layers are insufficient to account for the intense THz emission. This suggests that aside from the surface field, there could be other regions under strong electric fields such as the GaAs/n-GaSb interface. In Ref. 10, the THz emission of undoped-GaAs/n-GaAs layer on Si was attributed to three regions of high electric fields. We then calculated the interface field strength by assuming a 70:30 conduction band-valence band offset. The carrier concentration in the n-GaSb ($N_a = 4 \times 10^{17}\,\mathrm{cm}^{-3}$) was verified using van der Pauw Hall measurements. The calculated electric field and depletion width in the GaAs side of the GaAs/n-GaSb interface are 7 kV/cm and 0.54 μm, respectively.[17,18] The GaSb side is not relevant due to the limit of the laser penetration depth. Even so, the THz emission that could have resulted from contributions from both surface field and interface field could not account for the enhanced THz signal.

Because of the significant lattice mismatch in the heterostructure, the effect of strain and dislocations is anticipated. The $\theta/2\theta$ x-ray scans of the samples reveal broadening of GaAs peaks indicating structural defects, such as dislocations and strain.[19] However, RSM measurements reveal that the layers are relaxed. Figure 4 shows the RSM around the (444) epitaxial GaAs reciprocal lattice points (RELP). The k-space coordinate S_x of the RELP maxima is related to the parallel interplanar spacing and parallel lattice parameter.[20] From the maps, the calculated lattice parameter of the GaAs overlayer is 5.65 Å. Compared to the lattice parameter of a free-standing GaAs (5.6533 Å), the result suggests that the top GaAs layer is relaxed for all samples. It must be noted that at present, we cannot quantify the residual strain and the defect density cannot be inferred from the RSM data.

SEM micrographs in Fig. 5 show etch pits at the substrate-epilayer interface signifying a highly dislocated interface. It is believed there might still be incoherent strain in this region. The plan-view SEM images further attest that the surface is not smooth due to the presence of defects.

To explain the THz emission of the samples, we also consider the design of the metamorphic buffers (MBs). MBs are utilized in order to diminish, by orders of magnitude, the density of threading dislocations.[21] It is believed that not all of the dislocations are prevented by the buffer layers from propagating toward the surface. Furthermore, it is surmised that these dislocations induce a piezoelectric field. When there is biaxial strain in the (1,1,0) plane, the piezoelectric field in the [100] direction is described by the equation,[22] $E_{[100]} = \frac{2e_{14}}{\varepsilon_0(1+\chi)} \frac{C_{11}+C_{12}}{C_{11}+C_{12}+2C_{44}}\,\varepsilon$. We used the elastic stiffness constants given by Ref. 23. For an assumed residual strain of $\varepsilon = 0.001$, the piezoelectric field calculated is 9 kV/cm. It

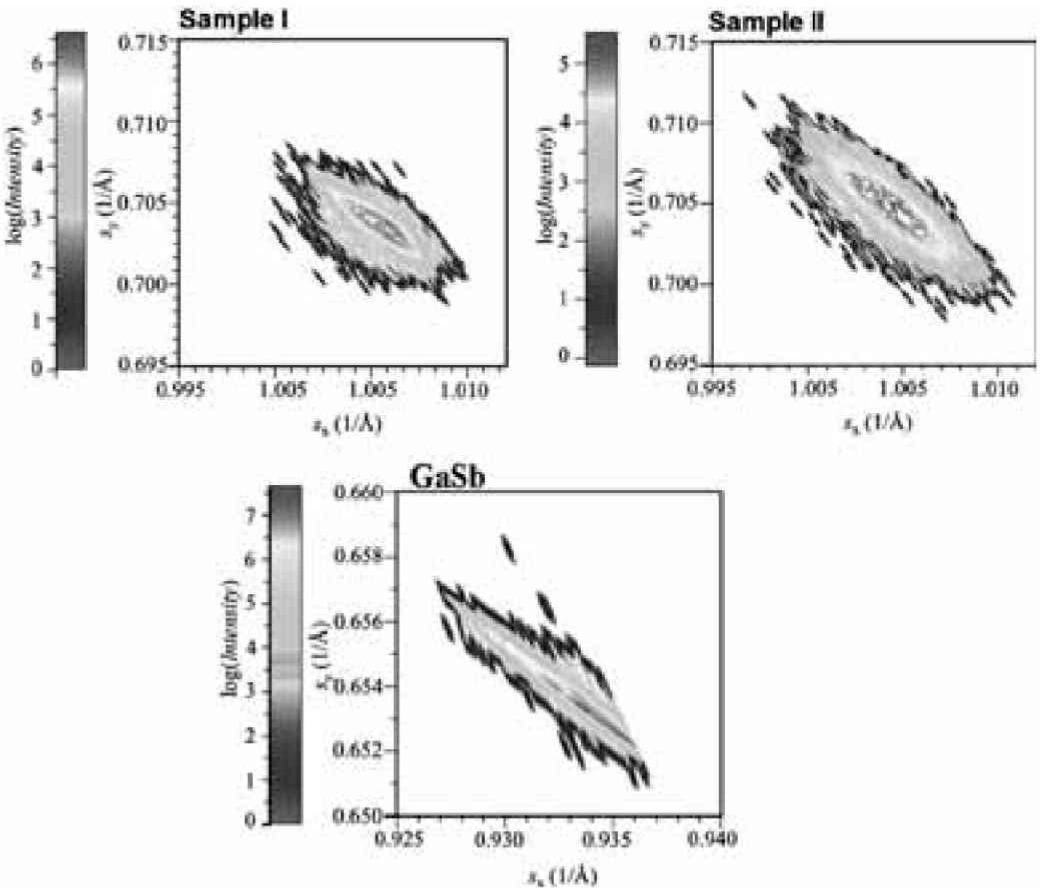

FIG. 4. Reciprocal space maps around the (444) reflection of the GaAs layers. Also shown is the RSM around the (444) reflection of the n-GaSb substrate.

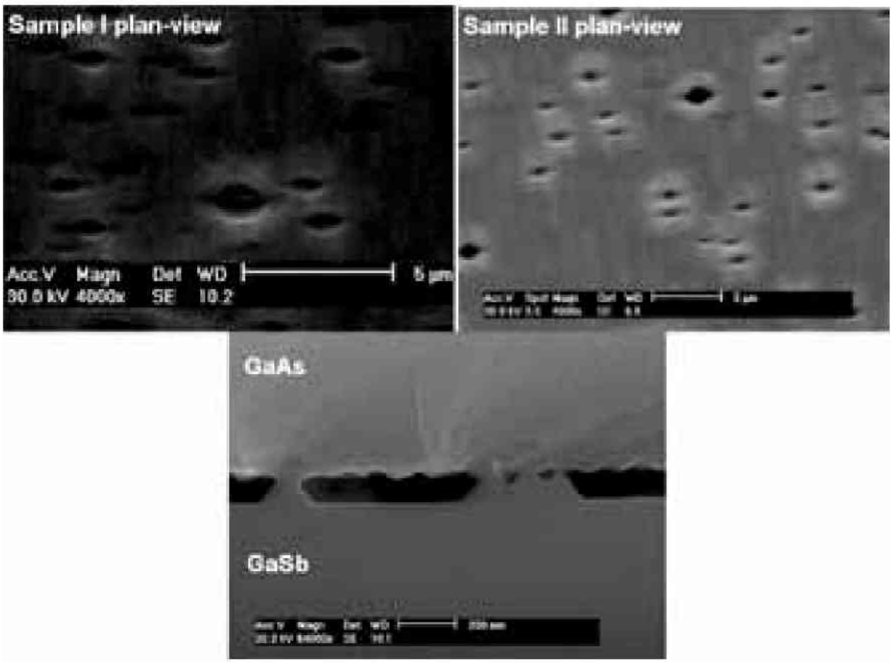

FIG. 5. Plan-view and cross-sectional electron micrographs of the samples. The etch pits at the interface is observed for all samples.

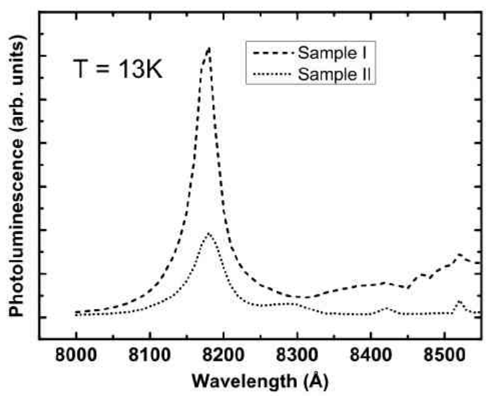

FIG. 6. Photoluminescence spectra (13 K) of the GaAs layers.

could be possible that the piezo-electric field contributes to the depletion width and may have further enhanced the THz emission for both samples.

GaAs/n-GaSb films showed photoluminescence (PL) at 13 K, as displayed in Fig. 6. Although both samples exhibited GaAs-related peaks at 8180 Å, the PL intensity was higher for sample I. Moreover, the PL spectra of sample II showed a carbon-related transition at 8300 Å. At present, the PL results suggest that sample I has better crystalline quality compared with Sample II.

IV. CONCLUSION

In summary, GaAs thin films were successfully formed on (100) n-GaSb by growth interruption during the initiation of epitaxial growth. Metamorphic buffer layers such as GaAs, InAs, AlAs/GaAs SL, and InAs/GaAs SL were used. The GaAs layer grown on InAs and InAs/GaAs SL buffer layers exhibited THz emission that is 75% of that of p-InAs. Our calculations show an enhanced depletion width due to the surface Fermi pinning and the bandgap offset. RSMs reveal that the layers are relaxed. SEM micrographs indicate dislocations and residual strain at the interface which explain the intense THz signal observed for both GaAs/n-GaSb films. PL spectra show sample II with InAs buffer is possibly more defective leading to enhanced THz emission. The results suggest that GaAs/n-GaSb material could be a viable alternative to bulk p-InAs.

ACKNOWLEDGMENTS

C. Sadia is grateful to the Commission on Higher Education (CHED) for the Ph.D. scholarship provided through the CHED-National Institute of Physics as a Center of Excellence Program. This work was also supported by the Department of Science and Technology-Philippine Council for Industry and Energy Research and Development (DOST-PCIERRD), DOST-Grants-In-Aid (GIA) program, and UP System research grant.

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