Characterization of GaAs grown by molecular beam epitaxy on vicinal Ge(100) substrates

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In this article we investigate the growth of GaAs on two different vicinal surfaces of Ge (100), cut 6° off the (100) plane toward the (110) plane or toward the (111) plane. Both substrates exhibit evidence of a regular array of double steps. Ge substrates and GaAs films are characterized with low energy electron diffraction, low temperature photoluminescence, scanning tunneling microscopy, electron channeling, and scanning electron microscopy. GaAs grown on wafers cut toward the (111) plane exhibits high quality as compared to reference GaAs samples, whereas GaAs grown on wafers cut toward the (110) plane displays clear evidence of three-dimensional growth and low crystallinity. © 2004 American Vacuum Society. [DOI: 10.1116/1.1774203]

I. INTRODUCTION

III–V optoelectronic materials have a wide variety of applications in today’s electronic devices. Integrating these materials onto elemental semiconductors such as Si and Ge would allow cost reduction, increased performance, and greater versatility of these optoelectronic devices. Much work has already been done on integrating GaAs-based device structures onto Si and Ge,1–3 and integrated material devices have already found useful applications in industry, such as GaAs/Ge based solar cells.4–7

In most of these systems, the interface between GaAs and Ge plays a crucial role in the device performance. Two key issues need to be addressed: antiphase domains (APD) and undesired cross-doping at the interface.8,9 One of the proposed solutions to reduce the former consists of growing GaAs on vicinal substrate surfaces. Substrates with sufficiently large offcuts exhibit double-stepped terrace structures that significantly reduce the number of APD’s. In particular, (100) surfaces of Si, SiGe, and Ge substrates offset 6° toward the (111) plane have been shown to produce regular arrays of double steps.

Little work has been reported so far on the impact of the offcut direction on the growth, and few studies have addressed offcuts on Ge (100) substrates. The goal of the present study is to show that regular arrays of double steps are perhaps the necessary, but not the sufficient, criterion, and that the offcut direction also plays a key role in achieving high quality GaAs epitaxy. We investigate the growth of GaAs on Ge vicinal surfaces cut at 6° off the (100) plane toward the (111) and (110) planes. Significant differences in GaAs quality are obtained from these two orientations.

II. EXPERIMENT

Ge(100) substrates with two different offcut orientations were purchased from Eagle-Picher. The first, referred to as Ge I, was offcut 6° toward the (110) plane, and the second, referred to as Ge II, was offcut 6° toward the (111) plane.

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FIG. 1. Low energy electron diffraction (LEED) pattern from a Ge buffer layer grown on (a) Ge I (112 eV) and (b) Ge II (65.2 eV); (c) LEED pattern from GaAs grown on Ge I (113 eV).
They were initially degreased for 5 min in 80 °C trichloroethylene, dipped in an ultrasonic bath of acetone for 5 min, and rinsed in methanol. The substrates were then exposed for 20 min to ultraviolet-generated ozone to remove surface carbon and grow a protective oxide, transferred to the surface analysis/Ge growth chamber, and annealed in ultra-high vacuum (~2×10⁻¹⁰ Torr) at 550 °C for 1 h to desorb the protective oxide. Desorption was followed by the growth of 0.5 μm of GaAs at temperatures ranging from 450 to 650 °C [measured by thermocouple imbedded in the molecular beam epitaxy sample block] and at a growth rate of 1 μm per hour. Following the GaAs growth, the samples were transferred back to the surface analysis chamber, sputtered to remove contamination, and annealed in UHV at 550 °C for 1 h to reorder the GaAs surface and investigate its structure via LEED. All other measurements (i.e., low temperature photoluminescence, electron channeling, Raman spectroscopy, and scanning electron microscopy (SEM)), were done ex situ.

III. RESULTS AND DISCUSSION

Figure 1 shows the LEED pattern obtained from the Ge buffer layer grown on (a) Ge I and (b) Ge II, and (c) from a GaAs layer grown on Ge II. Both Ge LEED patterns exhibit clear evidence of spot splitting in addition to the 2×1 μm per hour. Following the GaAs growth, the samples were transferred back to the surface analysis chamber, sputtered to remove contamination, and annealed in UHV at 550 °C for 1 h to reorder the GaAs surface and investigate its structure via LEED. All other measurements (i.e., low temperature photoluminescence, electron channeling, Raman spectroscopy, and scanning electron microscopy (SEM)), were done ex situ.

III. RESULTS AND DISCUSSION

Figure 1 shows the LEED pattern obtained from the Ge buffer layer grown on (a) Ge I and (b) Ge II, and (c) from a GaAs layer grown on Ge II. Both Ge LEED patterns exhibit clear evidence of spot splitting in addition to the 2×1 reconstruction due to dimerization of Ge surface atoms.10,11 The spot splitting arises from the convolution in the diffraction from two different periodicities at the surface: that associated

Fig. 2. Scanning tunneling microscopy (STM) micrograph from the Ge buffer layer grown on Ge II. The dimer rows are 8 Å apart.

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Fig. 3. Scanning electron microscopy (SEM) micrograph (E=5 keV) from GaAs grown on (a) Ge I and (b) Ge II. The latter is focused on a particle left on the surface, as the rest of the surface is featureless.
with the atomic structure of the surface, and that associated with the regular array of vicinal steps. By measuring the ratio of distances between split spots to distances between regular lattice spots, the step-induced periodicity and the average size of the step terraces can be measured. One can therefore determine the number of atomic layers present at each vicinal step. For Ge, which has a lattice parameter 5.65 Å, the dimer spacing is 4.00 Å, and the height of a double step is 2.83 Å. For a 6° offcut wafer, the ratio between the double step height and the terrace width is simply \( \tan \theta \) where \( \theta \) is the angle of the offcut. The terrace width is then calculated to be 26.9 Å for a double step. The ratio between terrace width and dimer spacing is predicted to be 6.7. From the diffraction patterns of Fig. 1, the \( \frac{2}{3} \frac{1}{2} \) unit cell dimension, which corresponds to the dimer spacing, can be compared to the spot-splitting, which corresponds to the vicinal terrace spacing. This leads to a ratio of 6.3 for Ge I and 6.8 for Ge II. These numbers are strong indications that both offcut orientations produce double stepped surfaces.

Surprisingly, the LEED patterns also show the presence of \( 2 \times 1 \) and \( 1 \times 2 \) domains. This suggests that there are single steps at the surface, since the orientation of the dimers changes by 90° across single atomic layer steps, but does not change across double layer steps. Figure 2 shows a STM micrograph representative of the whole surface of a Ge buffer layer grown on Ge II. The terraces are predominately double stepped since over a fairly large area of the surface the dimer rows have the same orientation. Since the ratio between spot splitting and unit cell dimension is consistent with the STM image, one concludes that the terraces are separated primarily by double steps. Single steps clearly exist on these offcut surfaces, but their density is significantly smaller than that of double steps.

The LEED pattern from the GaAs layer grown on Ge II [Fig. 1(c)] is consistent with that of a \( 4 \times 2 \)-reconstructed Ga-rich surface, which is expected for UHV annealing at \( \sim 550 \) °C.\textsuperscript{12} Spot splitting on the \( 4 \times 2 \) reconstruction is also consistent with a regular array of double steps (ratio of distances between split spots and regular lattice spots is 6.6). Interestingly, no LEED pattern is observed on GaAs grown on Ge I wafers.

Figure 3 shows SEM micrographs of the surface of GaAs. Films grown on Ge II wafers are very smooth over tens of microns, whereas films grown on Ge I wafers have a rough surface morphology. The lateral size of the features visible on the latter surface varies between 0.5 and 2 µm, suggesting a very poor epitaxy.

Low temperature photoluminescence (LTPL) spectra are shown in Fig. 4. The PL is excited by the 514 nm line of an Ar laser. At this wavelength, absorption in GaAs occurs in a few hundred angstroms. The main peak in each case corresponds to band-to-band recombination across the GaAs gap.
A standard Si-doped GaAs wafer tested for calibration gives a main PL peak at 1.494 eV with a full-width-at-half-maximum (FWHM) measurement of 8 meV. In comparison, the GaAs layer grown on Ge II gives a LTPL peak at 1.508 eV with a FWHM of 6 meV. However, the GaAs layer grown on Ge I gives a LTPL peak at 1.499 eV with a FWHM of 27 meV. The significant peak broadening is due to the poor crystallinity mentioned above.

Figure 5 shows the electron channeling patterns (ECP) obtained from both types of GaAs films. The ECP arises from the Kikuchi lines associated with the backscattering of high energy electrons from the different crystallographic planes of the layer, and probes a few thousand angstroms for a beam voltage of 20 kV. The orientation of the offset is deduced from the position of the center of the channeling pattern with respect to the center of the detector. This center, which corresponds to the normal direction of backscattering from the (100) terraces, is found at the intersection of the channel lines from the (111) and (110) planes. In each case, the sample offset angle is confirmed to be 6°. The substantially sharper definition of the ECP lines obtained for layers grown on Ge II also confirms the much better crystallinity of these layers.

Finally, backscattered Raman spectroscopy was performed using the 514 nm Ar laser line. As in PL, the probing depth in GaAs is on the order of a few hundred angstroms. Two peaks corresponding to the GaAs LO and TO phonons are visible on the spectrum from the layer grown on Ge I, whereas only one peak appears on the layer grown on Ge II. Selection rules on optical transitions forbid the TO phonon transition in pure crystalline GaAs in the (100) direction. This is consistent with what is observed from GaAs grown on Ge II [Fig. 6(b)]. However, the fact that both LO and TO phonon transitions are visible on GaAs grown on Ge I [Fig. 6(a)] is an additional indication that the film is not of high crystalline quality.

The substrate orientation is the only difference between the two types of GaAs films, the growth temperatures (450 to 650 °C) and growth rates being kept identical. Since well ordered step terraces and homoepitaxial buffer layers are obtained on both Ge I and Ge II surfaces, as measured by LEED for both and STM for Ge II, the orientation of the terraces and steps must play a key role in the heteroepitaxial growth process. We note that homoepitaxial GaAs has been successfully grown and modeled on GaAs substrates misoriented toward the (110) plane and toward the (111) plane. This suggests that Ga and As interact differently with a Ge substrate misoriented toward the (111) plane than with one misoriented toward the (110) plane. Although the reason is not understood at this point, we try here to relate the effect to the basic structure of the terrace edges. Figure 7 gives a schematic of the dimers and terraces for (a) Ge II and (b) Ge I. The Ge II structure was verified by STM (Fig. 2) but the Ge I structure is hypothetical. The Ge II terrace edges are parallel to the dimers and perpendicular to the dimer rows. Whereas the Ge I terrace edges are orientated 45° with respect to the dimers and dimer rows. Based on Fig. 2, we suspect that it is energetically favorable to have smooth terrace edges on Ge II, whereas terrace edges on Ge I substrate are inherently rough. The energy necessary to break dimers along the ideal terrace step is indeed greater than the energy needed to form a step. The step edge roughening could give rise to three-dimensional nucleation sites for GaAs, which would then lead to poor epitaxy. Further investigation is needed to explain why the energetics of the system change so drastically when growing on differently oriented Ge substrates.

IV. SUMMARY

We have investigated the growth of both Ge and GaAs on Ge (100) substrates offcut in two different directions: 6° toward the (110) plane (Ge I) and 6° toward the (111) plane (Ge II). We have shown that Ge buffer layers on both Ge (100) substrate orientations can be prepared with regular arrays of double steps. The combination of a range of experimental techniques shows that GaAs grown on Ge II is smooth and of high crystalline quality, whereas GaAs grown on Ge I exhibits poor morphology and crystalline quality. A
possible explanation for this phenomenon lies in the different interaction of Ga and As with the step edge structure for the two different orientations.

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