Research of surface morphology in Ga(In)As epilayers on Ge grown by MOVPE for multi-junction solar cells

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Abstract

The surface morphology of Ga(In)As films grown on offcut Ge substrates was characterized with atomic force microscopy (AFM) and scanning electron microscopy (SEM). We have investigated the effect of growth parameters on the Ga(In)As films by metal-organic vapor-phase epitaxy (MOVPE). The interface properties strongly depended on the growth conditions. We found that two-step growth temperatures can obtain good morphology and suppress the antiphase domains (APDs). Under optimized growth conditions APD-free Ga(In)As film on Ge was obtained. Our results indicate that the 6° offcut Ge substrate with two-step temperatures growth 770 and 650°C, are the optimum set of growth conditions for the buffer layer growth of Ga(In)As/Ge heterostructure solar cells. The root mean square (rms) roughness was approximately 4.58 nm over a 10×10 μm² area. The buffer Ga(In)As films on Ge substrate were developed in preparation for growing multi-junction solar cells and obtained high performance with good morphology.

Keywords: A1. Antiphase domain; A1. Morphology; B3. Solar cell

1. Introduction

GaAs/Ge heterostructures have received a great deal of attention due to their possible application in many electronic and optoelectronic devices, such as bipolar transistors [1], phototransistors [2], and high, efficiency solar cells [3]. The high quality of GaAs films grown on Ge substrates is crucial for the performance of these devices. Recently, the growth of high-efficiency multi-junction solar cells on Ge substrate has been reported [4–6]. Ge is an optimized substrate material in terms of its power-to-weight ratio for high-efficiency tandem solar cells. Although GaAs and Ge are nearly lattice matched (Δa/a = 0.07%) and the thermal mismatch is negligible, there still exist considerable problems related to the epitaxy between polar and non-polar semiconductors resulting in the formation of antiphase domains (APDs) [7–9]. It is well known that when polar material is epitaxially grown on non-polar material, two equivalent orientations, which correspond to an exchange of the location of cation atoms and anion atoms in the two sublattices, are expected in the epilayer [10]. APDs are connected by anti-phase boundaries (APBs), and domains of differing orientation are separated by an APB. Since APBs act as non-radiative recombination surfaces, providing deep levels in the forbidden gap [11], the successful realization of GaAs/Ge devices depends on the ability to grow a high-quality APD-free heterostructure.

In general, the surface roughness in GaAs films on Ge substrates is mainly due to APDs. Though the growth of GaAs on Ge substrates is rather challenging, the formation of APDs can be suppressed by using misoriented substrates and carefully selected initial growth parameters [12]. State of the art the conversion efficiency of InGaP/InGaAs/Ge-based multijunction solar cells has been improved up to
2. Experiments

The Ga(In)As heterostructures were grown by low-pressure metal-organic vapor-phase epitaxy (LP-MOVPE). The p-type Ge substrate with orientation (1 0 0) 6° off towards the [1 1 0] direction were used to grow a Ga(In)As epitaxial layer. The reactants used were trimethylgallium (TMGa) and trimethylindium (TMIn) as group-III materials and pure arsine (AsH₃) as group-V materials. Ultrapure H₂ was used as a carrier gas and for purging the system. The substrates were epi-ready, and no cleaning before the growth was carried out. The growth temperature, as measured from the thermocouple, was varied from 650 to 750°C, and nominal V/III ratios, from 75 to 115. For further sample codes in the article, refer to those in Table 1. Complementary and conventional characterization techniques, atomic force microscopy (AFM) and scanning electron microscopy (SEM), were applied to directly measure the film surface morphology. AFM is a more recently developed technique that is capable of providing atomic scale imagery of surfaces. SEM is used to study the APBs of the GaAs grown on Ge. Surface topographical images, with various scanning sizes of these samples were taken using AFM in order to reveal the surface roughening and other defects, most likely the APBs. Smooth surface morphology is expected to be an indication of good material properties and final device performance. In order to grow a completely APD-free Ga(In)As layer on Ge, the effects of growth temperature and other parameters were investigated.

As summarized in Table 1, the growth consisted of three parts: firstly, the GaAs growth that was carried out at growth temperatures varying from 650 to 750°C, with a V/III ratio of 115 and a growth rate of 50 nm/min to a thickness of about 2 μm, given the sample code of A; Secondly, the GaInAs growth, which was carried out at growth temperatures varying from 650 to 750°C, with a V/III ratio of 100 and a growth rate of 33 nm/min, given the sample code of B; Thirdly, the GaInAs growth, which was carried out at growth temperatures varying from 750 to 770°C, with a V/III ratio from 75 to 85 and a growth rate of 33 nm/min, given the sample code of C. The growth sequences for samples in this study were designed to evaluate the effect of surface roughness in preparation for growing multi-junction solar cells and obtaining high performance with good morphology.

3. Results and discussion

Table 1 summarizes the growth parameters and AFM analytical data for all the investigated films, the values of growth temperature, average roughness, root mean square (rms) roughness, and peak-to valley value. The rms roughness (\( \sigma_{\text{rms}} \)), with

\[
\sigma_{\text{rms}} = \sqrt{\langle [h(r) - \bar{h}]^2 \rangle},
\]

was given in Table 1.

### Table 1

<table>
<thead>
<tr>
<th>Sample</th>
<th>V/III ratio</th>
<th>Growth temperature (°C)</th>
<th>Growth rate (nm/min)</th>
<th>Scan size (μm × μm)</th>
<th>( R_a ) (nm)</th>
<th>( R_{\text{rms}} ) (nm)</th>
<th>P–V (nm)</th>
<th>Epi layer</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>115</td>
<td>650</td>
<td>50</td>
<td>3 × 3</td>
<td>17.82</td>
<td>92.91</td>
<td>166.0</td>
<td>GaAs</td>
</tr>
<tr>
<td>A2</td>
<td>115</td>
<td>700</td>
<td>50</td>
<td>3 × 3</td>
<td>17.27</td>
<td>97.07</td>
<td>172.0</td>
<td>GaAs</td>
</tr>
<tr>
<td>A3</td>
<td>115</td>
<td>750</td>
<td>50</td>
<td>3 × 3</td>
<td>5.87</td>
<td>21.73</td>
<td>42.4</td>
<td>GaAs</td>
</tr>
<tr>
<td>B1</td>
<td>100</td>
<td>650</td>
<td>33</td>
<td>3 × 3</td>
<td>14.37</td>
<td>71.00</td>
<td>149.0</td>
<td>InGaAs</td>
</tr>
<tr>
<td>B2</td>
<td>100</td>
<td>700</td>
<td>33</td>
<td>3 × 3</td>
<td>14.80</td>
<td>101.49</td>
<td>177.0</td>
<td>InGaAs</td>
</tr>
<tr>
<td>B3</td>
<td>100</td>
<td>750</td>
<td>33</td>
<td>3 × 3</td>
<td>4.87</td>
<td>23.81</td>
<td>38.8</td>
<td>InGaAs</td>
</tr>
<tr>
<td>C1</td>
<td>85</td>
<td>750</td>
<td>33</td>
<td>3 × 3</td>
<td>5.87</td>
<td>23.42</td>
<td>40.7</td>
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<td>7.47</td>
<td>23.35</td>
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<td>C2</td>
<td>75</td>
<td>760</td>
<td>33</td>
<td>3 × 3</td>
<td>1.49</td>
<td>7.23</td>
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<tr>
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<td></td>
<td></td>
<td>10 × 10</td>
<td>8.74</td>
<td>11.47</td>
<td>36.7</td>
<td>InGaAs</td>
</tr>
<tr>
<td>C3</td>
<td>75</td>
<td>770</td>
<td>33</td>
<td>3 × 3</td>
<td>1.45</td>
<td>9.03</td>
<td>25.5</td>
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</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>10 × 10</td>
<td>4.17</td>
<td>5.56</td>
<td>23.8</td>
<td>InGaAs</td>
</tr>
<tr>
<td>C4</td>
<td>75</td>
<td>770/650</td>
<td>x</td>
<td>3 × 3</td>
<td>0.72</td>
<td>4.99</td>
<td>16.2</td>
<td>InGaAs</td>
</tr>
</tbody>
</table>

\( R_a \): average roughness, \( R_{\text{rms}} \): root mean square roughness, P–V: peak-to valley value.
where \( \bar{h} \) is the mean height of the surface and \( h(r) \) is the height of the surface at a distance \( r \) on the surface, was calculated from the AFM images.

3.1. Effect of growth temperature of GaAs on six off-oriented Ge substrate

The effect of growth temperature on the layer properties of GaAs grown on Ge was studied by Agarwal et al. [15]. An initial buffer layer of 2 \( \mu \)m of GaAs was grown on the Ge substrate. Three two-dimensional (2D) AFM topographical images of 3 \( \mu \)m \( \times \) 3 \( \mu \)m scanning sizes are shown in Figs. 1(a)–(c) as a function of growth temperature at 650, 700 and 750 °C, respectively, with a V/III ratio of 115 and a growth rate of 50 nm/min. The influence of the growth temperature on the surface morphology can be seen in the AFM image. The samples grown at three different temperatures clearly exhibit the formation of surface roughness. It is obvious that the average, the rms roughness, and peak-to-valley values decrease with increasing growth temperature in 6° off-oriented Ge substrate, except at growth temperature of 700 °C. The 3-D images of samples A1, A2 and A3 are shown in Figs. 2(a)–(c), respectively. It is clear that sample A1 has numerous grave hillocks. The \( \sigma_{\text{rms}} \) are 9, 10, 2 Å for samples A1, A2 and A3, respectively, in 3 \( \mu \)m \( \times \) 3 \( \mu \)m scanning sizes. The film grown at 750 °C shows lower average and rms roughness and has better surface morphology. However, the film grown at 650 °C has a hazy surface. This indicates that a 6° offcut of Ge substrate with a higher growth temperature of 750 °C may be the appropriate choice for the buffer layer growth of the GaAs/Ge solar cell.

3.2. Effect of growth temperature, V/III ratio of Ga(In)As on 6 off-oriented Ge substrate

The investigation of GaIn\(_{0.1}\)As grown on Ge followed that of GaAs discussed above, as in Table 1. Furthermore, the various V/III ratios were studied. The results indicate that the average, the rms roughness, and peak-to-valley values decrease with increasing growth temperature, except at a growth temperature of 700 °C, which is in accordance with GaAs grown on Ge. It seems that the small amount of In incorporated into InGaAs did not affect the surface morphology. A growth sequence which involves raising the growth temperature from 750 to 770 °C was used to evaluate the effect of higher temperature. It can be observed that a much smoother surface was obtained for the sample grown at a higher temperature. In addition, two-step growth was carried out. The first layer (1 \( \mu \)m) was...
grown at the rate of 33 nm/min at the growth temperature of 770 °C. The second step layer (1 μm) was grown at 650 °C. Figs. 3(a) and (b) present the cross-sectional and top-view SEM micrograph of GaInAs grown on Ge at 650 °C. The GaInAs film growth at 650 °C induced a high density of APDs and threading dislocations at the interface, with many huge APDs extending to the epilayer surface. There are crevices and elongated mounds on its surface. Figs. 4(a) and (b) show the cross-sectional and top-view SEM micrographs with Ga(In)As grown on Ge with two-step temperature growth. Cross-sectional TEM image of 650 °C grown GaInAs on Ge (Fig. 3(a)) confirms the presence of numerous APBs punching through the surface, causing the rough surface. In contrast, with two-step temperature growth (Fig. 4(a)), the APDs are completely eliminated. The formation of APDs was found to depend primarily on the growth temperature. Fig. 5 depicts the root mean square roughness as functions of growth temperature for all samples. In the range of 650–750 °C, the rms roughness as functions of growth temperature reveals the same tendency for GaAs and GaInAs. By raising the growth temperature, over 700 °C, a lower rms roughness was obtained for suppressing the APD on Ge epitaxy. But lowering the growth temperature, below 700 °C, a lower rms roughness was obtained for obtaining high quality of GaAs or GaInAs films. The highest purity material is generally agreed to be obtained at the lowest growth temperature, in the range from 600 to 650 °C [20].

The lowest rms roughness is 4.58 nm obtained by two-step growth, in 10 μm × 10 μm scanning sizes. Two-step growth results in a better quality epitaxial layer as compared with single-step growth. This is a result of raising the growth temperature to 770 °C for the first 1 μm to reduce the threading dislocation density. This result clearly demonstrates that the initial growth temperature plays a direct
role in the formation of APDs. Owing to the first buffer layer being grown at a higher temperature, the atoms have sufficient mean free paths to reach their proper and respective sites thereby reducing defects and APDs at the interface. The APDs were suppressed by 750 °C growth and subsequent lower temperature (650 °C) film growth resulting in superior quality. The reasons for growing lower temperature film were mentioned before that the highest purity material is generally agreed to be obtained at the lowest growth temperature, in the range from 600 to 650 °C [20]. The improvements were evident from the surface morphology and rms roughness studied by AFM and SEM. Thus, we conclude that Ga(In)As grown on Ge with two-step temperature growth is much smoother than that with single-step temperature growth. This result is in opposition to that reported in Refs. [7,15,16].

4. Conclusion

Ga(In)As heterostructures were grown by low-pressure metal-organic vapor-phase epitaxial technique and studied with AFM and SEM. Cross-sectional SEM studies showed that APD-free GaInAs growth on Ge was possible if the growth parameters were properly selected. The average, the rms roughness, and peak-to-valley values decrease with increasing growth temperature. Our results indicate that the 6° offcut Ge substrate with two-step temperature growth of 770 and 650 °C and a V/III ratio of 75 are the optimum set of growth conditions for the buffer layer growth of GaInAs/Ge heterostructure solar cells. The lowest rms roughness of 4.58 nm is obtained by two-step growth in 10 μm × 10 μm scanning sizes. Thus, we conclude that two-step temperature growth on the Ge buffer layers is crucial for growing good quality GaInAs films, defined as films that have a smooth surface and the APD-free. The buffer GaInAs films on Ge substrate were developed in preparation for growing multi-junction solar cells and obtaining high performance with good morphology.

References