Preparation of large area and high performance flexible GaInP/GaAs/InGaAs tandem solar cells

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Abstract: Flexible high-efficiency III-V multijunction solar cells are being developed for use in unmanned Aerial Vehicles (UAVs), wearable devices and space applications. The solar cell epitaxial layers are grown on GaAs substrate by metalorganic chemical vapor deposition (MOCVD) and then are transferred to flexible substrates by cold-bonding and epitaxial lift-off process (ELO). Through the design of ELO apparatus and a large number of experiments on the optimal parameter GaAs solar cell structure can be effectively separated from 4-inch GaAs wafer without defects and degradation in performance. Recently, 30 cm2 large area flexible GaInP/GaAs/InGaAs 3-junction solar cells on 50 μm polyimide film achieved a 1-sun AM0 conversion efficiency of 31.5% with an open-circuit-voltage of 3.01 V a short-circuit current-density of 16.8 mA/cm2 and a fill factor of 0.845. By using the very light PI substrate the unit weight of the solar cell is only 168.5 g/m2 and the specific power is up to 2 530 W/kg.

Key words: solar cells, flexible and high-efficiency, epitaxial lift-off, GaAs

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Introduction

GaInP/GaAs/Ge lattice matched triple-junction solar cells with high efficiency, high reliability and long life have become the main force of the space power supply \(^{1,2,3}\). However, it cannot be ignored that despite the significant increase in the conversion efficiency, the weight of the substrate material seriously restricts the improvement of the specific power of the battery \((\text{W} \cdot \text{kg})^{-1}\). Another problem is that the Ge bottom cell absorbs approximately two times more low energy photons than are needed for current matching with the GaInP and GaAs subcells. It is well known that the Ge bottom junction would be replaced with a 1.0-eV In\(_{0.3}\)Ga\(_{0.7}\)As junction that is lattice matched with the other junctions. Because of the 2% lattice mismatch between In\(_{0.3}\)Ga\(_{0.7}\)As and other subcells\(^4\) graded composition buffer layers (metamorphic) was adopted to avoid the dislocations. However, the remaining threading dislocations would significantly degrade any subsequently grown junctions with higher band gaps. By growing in an inverted configuration this degradation of the top junctions can be avoided\(^5\).

Epitaxial lift-off (ELO)\(^6\) known as peeled film technology was initially developed in the late 1980\(^{7}\). With this technique, III-V multijunction solar cells structures can be separated from their GaAs substrates using selective wet etching of a thin AlAs or Al\(_x\)Ga\(_{1-x}\)As layer by electron beam evaporation\(^8\). As a result, GaAs solar cells can be cemented on arbitrary flat carriers for further processing\(^9\).

As a result, GaAs solar cell structure can be effectively separated from 4-inch GaAs wafers without defects and degradation in performance with a lateral etch rate of about 25 ~ 35 mm/h. However, subsequent processing these thin-film structures into actual ELO solar cells was found to be difficult because the epitaxial layers are very thin and fragile. Therefore, GaAs solar cells were transferred to flexible support substrates such as polyimide or metal foils by a bonding process before the ELO process. The thin-film structures obtained by the ELO process can be cemented on arbitrary flat carriers for further processing\(^10\).

In this work\(^1\) we demonstrate the application of the cold-bonding and epitaxial lift-off technique as an effective substrate thinning method for making ultra-thin high-efficiency flexible triple junction GaInP/GaAs/InGaAs solar cells. Moreover, the growth technology of InGaP/GaAs/InGaAs tandem solar cells has been fabricated.

1 Experimental

In this study\(^1\) the GaInP/GaAs/InGaAs inverted 3-junction cells were grown by Metalorganic Chemical Vapor Deposition and followed by cold-bonding and epitaxial lift-off. Figure 1 shows a typical process flow for flexible GaInP/GaAs/InGaAs tandem solar cells. The process starts with growth of inverted metamorphic (IMM) 3-junction GaInP/GaAs/InGaAs epitaxial layer\(^1\) followed by the cold-bonding between epitaxial layer and PI thin film. After cold-bonding\(^6\) epitaxial lift-off (ELO) process was conducted in order to transfer the solar cell structure to the PI substrate. Finally, flexible solar cells can be prepared by rigid substrate fixation and traditional solar cell technology.

All metamorphic buffer and solar cells layers in this work have been grown by Metalorganic Chemical Vapor Deposition using an AIX2600-G3 reactor with 4-inch substrate. The n-GaAs substrates were used with a 7\(^\circ\) offset from the (001) to (111) B plane and the carrier concentrations were \((1 ~ 4) \times 10^{18} \text{cm}^{-3}\). The hydrid sources arsine and phosphine were used for the group-V-growth. Trimethylgallium at 7.5\(^\circ\)C and ethyldimethylindium at 16\(^\circ\)C were used as the group III precursors. SiH\(_4\) was used as the dopant for n-type doping of III-V compound epitaxial layers\(^6\) while diethylzinc and CCl\(_4\) were used as the p-type dopants. The MOCVD growth conditions for each of these subcells were optimized to yield a high quality material with minimum defects and optimal electrical performance.

Both the epitaxial film of solar cells and the PI film were cleaned and passivated to a high quality of surface finish prior to subjecting the wafers to bonding. First\(^1\) all the epitaxial subcells were cleaned by acetone\(^5\) isopropanol and ethanol sequentially in an ultrasonic bath and then rinsed by deionized water\(^5\) and finally dried with N\(_2\). The polyimide (PI) thin films were first dipped in KOH solution and then cleaned in acetone and ethanol solutions to remove the pollutions of carbon and metal oxides. After that\(^6\) a 50 nm titanium film and then 300 nm gold film were deposited on the surface of PI and epitaxial layer by electron beam evaporation\(^5\) respectively.

The bonding was carried out using Karl Suss SB6e wafer bonding equipment. The bonding parameters such as temperature\(^5\) pressure and bonding time were carefully optimized to achieve high mechanical strength and low electrical resistance across the bonded interface. Scanning acoustic microscopy was used to determine the uniformity and quality of the bonded interface. After wafer bonding\(^6\) the GaAs substrate was removed by using the ELO process. The etch solution concentration\(^5\) temperature and ELO rate were carefully optimized.

2 Results and discussion

Figure 2 shows the schematic of inverted grown GaInP/GaAs/InGaAs 3J solar cells. The solar cell layers are grown in reverse order to be consistent with the ELO process. First\(^1\) a 20 nm AlAs sacrificial layer was grown\(^1\) and then GaInP top subcell\(^1\) GaAs middle subcell and In\(_{0.3}\)Ga\(_{0.7}\)As bottom subcell were grown in turn. GaInAlAs grade buffer structures were grown with a step-graded design between the GaAs and In\(_{0.3}\)Ga\(_{0.7}\)As subcells. The grade consisted of eight 250 nm steps and a 800 nm tuning layer which was varied in composition. On top of the tuning layer\(^1\) a final 200 nm In\(_{0.3}\)Ga\(_{0.7}\)As lattice matched to the 1eV In\(_{0.3}\)Ga\(_{0.7}\)As bottom cell layer was grown.

Figure 3 shows the characterization results for the epitaxial layer of GaInP/GaAs/InGaAs solar cell. High resolution X-ray diffraction in recent decades is the de-
Development of a non-contact and non-destructive crystal quality detection technology especially the reciprocal space map obtained by the three axis diffraction (RSM) can visually display the distribution of reciprocal lattice space scattering intensity. By analyzing the location and shape of reciprocal space map we get directly the information about lattice integrity in defects, stress and strain, mosaic structure and interface matching condition in lattice. The stress state of the buffer layer and the sub-cells can be clearly seen by the HRXRD RSM. It is found that the GaInAlAs grade buffer layer is almost a complete relaxation state and relaxation degree calculated by the software is about 99% as figure 3(a) shown, which indicates that the dislocation caused by the lattice mismatch will be completely released into the buffer layer, making the InGaAs subcell have high material quality. In addition, it can be seen that the bottom subcell was a tensile state which is helpful to attain a high Voc.

Figure 3 (b) shows the PL (photoluminescence spectroscopy) of epitaxial layer of GaInP/GaAs/InGaAs solar cell at room temperature by using a 532 nm laser. Due to the band gap of the InGaAs material at the top is lower than other subcells, the PL signals of GaInP and GaAs cannot be detected since the luminescence below will be absorbed by the InGaAs. The PL peak of InGaAs is located at 1229.5 nm indicating the bandgap of InGaAs is about 1 eV (1240 eV* nm / 1229.5 nm) and the Indium component is 29.9% which is very consistent with the design value 30%. In addition, the FWHM (full width at half maximum) of the PL peak is only 75 nm showed that the In_{0.3}GaAs material has very good material quality.

Figure 4 shows scanning acoustic microscopy test of the GaAs epitaxial wafer/PI film obtained by cold bonding process. The quality of bonding is mainly affected by the sample surface roughness, cleaning methods and bonding parameters. In addition, in order to avoid the influence of impurities, the whole bonding process is conducted in super clean room. After parameter optimization, we obtained a set of process parameters to achieve a large area uniform bonding: Chamber pressure: 2.00E-3 mbar; Bonding temperature: 250°C; Bonding pressure: 20 000N; Bonding time: 1 h. In order to achieve large area and high quality bonding, the cleaning process has been improved. It is can be seen that the hole or cavity in the bonding interface as shown in figure 4(a) were thoroughly removed as shown in figure 4(b).

Figure 5 (a) shows schematic diagram of ELO process. The ELO epitaxial layer structure especially the sacrificial layer was optimized. In order to speed up the separation rate and shorten the process time, we mainly enhance the reaction gas escape rate by apply external forces and control the stress by our design of the ELO apparatus. Figure 5(b) shows the photo of flexible 3-junction InGaP/GaAs/InGaAs solar cell epitaxial layer on PI substrate obtained by bonding and ELO process. It can be seen that the surface of the sample is very bright and uniform without defects such as peeling off, bubbles or cracks, indicating that the transfer process of the epitaxial layer has a high quality.
The solar cell efficiency is increased to 31.5% with the greatest improvement coming from the high-performance InGaP/GaAs/InGaAs solar cell. GaAs solar cells with an open-circuit voltage of 3.01 V have been reached. By using the very light PI film, a low open-circuit voltage of about 0.53 V is achieved for GaAs solar cells. Almost 1.45 V comes from the 1.9 eV GaInP top junction, approximately 1.45 V comes from the 1.9 eV GaInP top junction, and 0.53 V comes from the 1.9 eV GaInP top junction. The GaAs solar cell structure can be efficiently separated from 4-inch GaAs wafers without defects and degradation in performance.

The 4-inch flexible InGaP/GaAs/InGaAs solar cell, as shown in Fig. 6, can be obtained by traditional device processes such as photo-lithography, development, metal deposition, ARC (Antireflection coating), etc. Depending on the application requirements, the flexible solar cells can be cut into different sizes by dicing machine and Fig. 6(b) shows the largest 4 × 8 cm² size (lack of two triangles on the edge the real area is 30 cm²) flexible InGaP/GaAs/InGaAs solar cell.

Figure 7 shows the result of the photovoltaic I-V characteristics of the 30 cm² flexible InGaP/GaAs/InGaAs solar cells with an open-circuit voltage of 3.01 V, a short-circuit current-density of 16.8 mA/cm², a fill factor of 0.845, and AM0 conversion efficiency of 31.5%. The greatest improvement comes from the high Voc of the bottom subcell. Based on our previous work, similar single-junction devices indicated that approximately 1.45 V comes from the 1.9 eV GaInP top junction, 1.03 V from the 1.424 eV GaAs middle junction, and 0.53 V from the 1.9 eV GaInP top junction. Therefore, the bottom In₀.₃GaAs subcell contributes about 0.53 V of open-circuit voltage. Compared with the previous 0.48 V it has been increased about 50 mV due to the tensile state in bottom subcell and the high material quality as Fig. 3 shown. By using the very light PI substrate the unit weight of the solar cell is only 168.5 g/m² as shown in Table 1. Thus, the specific power of flexible InGaP/GaAs/InGaAs solar cells is up to 2 530 W/kg which is very suitable for application in unmanned Aerial Vehicles (UAVs), wearable devices and other space solar cell arrays with high demand for light and flexible thin film solar cells.

### Table 1  Weight composition of each part of the flexible 3J solar cells

<table>
<thead>
<tr>
<th>Component</th>
<th>Unit weight (g/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PI Substrate</td>
<td>71.0</td>
</tr>
<tr>
<td>Epitaxial layer</td>
<td>73.1</td>
</tr>
<tr>
<td>Bonding metal</td>
<td>20.6</td>
</tr>
<tr>
<td>ARC and grid electrode</td>
<td>4.8</td>
</tr>
<tr>
<td>The solar cell</td>
<td>168.5</td>
</tr>
</tbody>
</table>

The external quantum efficiency (EQE) of all three junctions was shown in figure 8. It is obvious that the peak EQE of the GaInP and GaAs subcells is close to 90% while the peak EQE of the In₀.₃GaAs subcell is also more than 80%. In general, the QE curve of InGaAs subcell is downward sloping from 900 nm to 1250 nm but it is relatively flat here which may be related to the photons reflection effect of the back metal (for cold bonding). Other workers have reported enhanced solar cell performance by including a DBR stack in the buffer layers of GaAs solar cells to promote photon recycling.

To further improve the efficiency, the ARC needs to be optimized to reduce the average reflectivity from 300 nm to 1300 nm. In addition, it is necessary to point out that the flexible GaInP/GaAs/InGaAs solar cells can’t be annealed at high temperature above 250°C due to the limitation of flexible thermoplastic substrate. Therefore new flexible substrates or low temperature annealing electrode materials are now in progress in order to further lower the contact resistance of the electrode which enhance the fill factor up to 0.86 and increase the efficiency exceeding 32%.

### 3 Summary

Flexible high-efficiency III-V multijunction solar cells are being developed by using the cold-bonding and epitaxial lift-off process (ELO). Through the design of ELO apparatus and a large number of experiments on the optimal parameter GaAs solar cell structure can be effectively separated from 4-inch GaAs wafer without defects and degradation in performance. Recently, high-quality 1.0 eV In₀.₃GaAs subcell and GaInP/GaAs/In₀.₃GaAs...
GaAs epitaxial layers have been fabricated. Combined with flexible thin film solar cells preparation process 30 cm$^2$ large area flexible GaInP/GaAs/InGaAs 3-junction solar cells on 50 μm polyimide film achieved a 1-sun AM0 conversion efficiency of 31.5% with an open-circuit-voltage of 3.01 V and a short-circuit-current-density of 16.8 mA/cm$^2$ and a fill factor of 0.845. By using the very light PI substrate the unit weight of the solar cell is only 168.5 g/m$^2$ and the specific power is up to 2 530 W/kg.

References


