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Multiple growths of epitaxial lift-off solar cells from a single InP substrate

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We demonstrate multiple growths of flexible, thin-film indium tin oxide-InP Schottky-barrier solar cells on a single InP wafer via epitaxial lift-off (ELO). Layers that protect the InP parent wafer surface during the ELO process are subsequently removed by selective wet-chemical etching, with the active solar cell layers transferred to a thin, flexible plastic host substrate by cold welding at room temperature. The first- and second-growth solar cells exhibit no performance degradation under simulated Atmospheric Mass 1.5 Global (AM 1.5G) illumination, and have a power conversion efficiency of $\eta_p = 14.4 \pm 0.4\%$ and $\eta_p = 14.8 \pm 0.2\%$, respectively. The current-voltage characteristics for the solar cells and atomic force microscope images of the substrate indicate that the parent wafer is undamaged, and is suitable for reuse after ELO and the protection-layer removal processes. X-ray photoelectron spectroscopy, reflection high-energy electron diffraction observation, and three-dimensional surface profiling show a surface that is comparable or improved to the original epiready wafer following ELO. Wafer reuse over multiple cycles suggests that high-efficiency; single-crystal thin-film solar cells may provide a practical path to low-cost solar-to-electrical energy conversion. © 2010 American Institute of Physics. [doi:10.1063/1.3479906]

Epitaxial lift-off (ELO), in which the active region of an electronic device structure is separated from its parent substrate and then transferred to a different host substrate, is attractive for cost reduction in III-V thin film solar cells by reusing the original and costly substrate.¹⁻⁸ However, wafer reuse has been limited by the increased wafer surface roughness caused by wet-chemical etching during ELO. Although chemo-mechanical polishing can restore the surface smoothness,^{2,3} the procedure removes some of the substrate material and can lead to damage. Here, we employ epitaxial protection layers for both the substrate and the lifted-off thin film to provide a high quality regrowth surface without polishing. By combining ELO with cold welding to transfer the epitaxial solar cells to a thin, flexible plastic sheet,⁹ we demonstrate reuse of the original substrate for fabricating efficient indium tin oxide (ITO)-InP Schottky-barrier thin-film solar cells¹⁰ without loss of device performance. Power conversion efficiencies approaching $\eta_p = 15\%$ are achieved.

The epitaxial layers were grown, starting with a 0.2 μ m thick InP buffer layer, on a S-doped (100) InP substrate by gas source molecular beam epitaxy (GSMBE). Then, a 0.1 μ m thick In_{0.53}Ga_{0.47}As etch stop layer, followed by a 1.5 μ m thick InP protection layer are grown. At this point, a 10 nm thick AlAs sacrificial layer is grown which is later etched away to separate the active layers from the parent wafer. Next, a second 1.5 μ m thick InP protection layer is grown, followed by a 0.1 μ m thick In_{0.53}Ga_{0.47}As etching stop layer, and then the active region consisting of a 3.0 μ m thick lightly Be-doped (3×10¹⁶ cm⁻³) InP base layer with

0.1 μ m thick Be-doped (3 × 10¹⁸ cm⁻³) InP Ohmic contact layer is grown. For the second growth, the InP buffer layer thickness was increased from 0.2 to 2 μ m to smooth the surface. In all other respects, the first- and second-growth epitaxial structures were identical.

Immediately following growth, a 100 Å thick Cr adhesion layer followed by a 600 Å Au contact layer were sputtered onto a 50 μ m thick Kapton[®] sheet, and a 600 Å of Au layer was deposited on the highly p-doped InP epitaxial layers by electron-beam evaporation.

After metal deposition, the wafer is mounted Au-side down on the plastic sheet and a cold-weld bond⁹ is formed by applying a pressure of 10 MPa for 60 s. Then, the Kapton[®] sheet is affixed to a rotatable, 7.5 cm diameter Teflon rod with Kapton[®] tape, and immersed into an etching solution of HF:H₂O (1:10). To expedite the ELO process, a 13 g weight is mounted on the plastic substrate, and the Teflon rod is rotated to maintain an external force on the wafer while increasing the gap between the epitaxial layers and substrate as the etching proceeds.

After completing the ELO, the Au residue on the original InP substrate is removed by etching (etchant type TFA, Transene Co.). The InP protection and $In_{0.53}Ga_{0.47}As$ etchstop layers on both the substrate and solar cell are then selectively removed by etching in H₃PO₄:HCl (1:3) and H₂SO₄:H₂O₂:H₂O (1:1:10), respectively. Subsequently, citric acid:H₂O₂ (20:1) is used to fully remove the remaining $In_{0.53}Ga_{0.47}As$. The freshly exposed InP substrate surface is then degreased by sequential dipping in heated trichloroethylene, acetone, and heated isopropanol, followed by the intentional growth of an oxide by exposure to UV/ozone for 10 min. The substrate is then loaded back into the GSMBE

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FIG. 1. (Color) (a) Atomic force microscope images of the surfaces of the original epiready InP substrate, recovered InP substrate surface after the first and second ELO processes following the removal of the protection and etch stop layers, and an ELO surface without the protection layer. (b) Image of lifted-off 2 in. diameter epitaxial layer containing an array of ITO/InP thin film solar cells fabricated by a combination of ELO and cold-weld bonding to a 50 μ m thick Kapton[®] sheet.

chamber and degassed. The second photovoltaic cell is then grown on the original parent substrate using the identical procedure described above, followed by a second round of cold welding and ELO.

To fabricate the ITO/InP Schottky junction, exposure to UV/ozone for 7 min removed the surface contaminants and forms a thin, passivating native-oxide.¹¹ The ITO is sputter-deposited at a rate of 0.3 Å/s through a shadow mask with an array of 1 mm diameter openings. A control, thin-film ITO/InP solar cell cold-welded onto the Kapton[®] sheet with the same epitaxial structure was fabricated via complete substrate removal, as described previously.¹²

Selective removal of the protection and etch-stop layers must result in a pristine wafer surface to allow for repetitive use of the parent substrate. Sulfuric acid based solutions have a high etch rate (0.22 μ m/min), with a selectivity between In_{0.53}Ga_{0.47}As and InP of 80:1.¹³ This etchant results in a 1.7 nm root-mean square (rms) roughness of the InP substrate after In_{0.53}Ga_{0.47}As removal. In contrast, the citric acid:peroxide etch has higher selectivity (470:1) (Ref. 13) but nanometer-scale spikes remain after etching. To minimize the InP surface damage and roughness, and to eliminate residual In_{0.53}Ga_{0.47}As, the sulfuric acid etch was used to remove the majority of In_{0.53}Ga_{0.47}As, followed by sonicat-



FIG. 2. (Color online) The x-ray photoelectron spectra from the fresh and the ELO processed substrate surfaces. Inset: The $2\times$ and $4\times$ surface reconstruction patterns obtained by reflection high-energy electron diffraction for the fresh and the ELO-processed substrate after eliminating oxide layer.

ing in the citric acid etch. The same procedure was employed for $In_{0.53}Ga_{0.47}As$ layer removal from the solar cell epitaxial surface, with 1 min longer exposure to the citric acid solution and without sonication.

Atomic force microscope images [Fig. 1(a)] show that the rms roughness of the InP surface after $In_{0.53}Ga_{0.47}As$ layer removal following both the first and the second growths (0.16±0.01 nm) is less than that of the epiready original InP substrate (0.30±0.02 nm), and the ELO wafer that lacked a protection layer (0.41±0.02 nm).

An image of the completed, 2 in. diameter epilayer with an array of ITO/InP solar cells bonded to the Kapton[®] substrate is shown in Fig. 1(b). As previously, these cells can withstand considerable bending stress without cracking or a degradation in performance.¹²

Figure 2 shows the x-ray photoelectron spectra that compare the surface elemental compositions of the fresh and ELO-processed substrates following protection layer removal. Both substrates show almost identical In and P peak intensities at the same binding energies, indicating that the surface after ELO has not changed from that of the original, epiready InP substrate. Furthermore, the reflection highenergy electron diffraction patterns obtained from the wafer



FIG. 3. (Color online) Current density vs voltage characteristics of the first and the second ELO processed ITO/InP and the control solar cells bonded to a Kapton[®] sheet, and measured under 1 sun, AM 1.5G simulated solar illumination. Inset: Surface morphology of the original substrate (a) after ELO and protection layer removal and (b) after the ELO without the protection layer.

TABLE I. Comparison of device performances under AM 1.5G simulated solar spectrum, and the dark current at $-1 \text{ V} (J_D)$, ideality factor (*n*), and specific series resistance (R_s) in the dark.

	J_D $(\mu A/cm^2)$	п	R_s ($\Omega m cm^2$)	J_{sc} (mA/cm ²)	V_{oc} (V)	FF (%)	$\eta_p \ (\%)$
Previous work ^a	1.6	1.14	5.4	29.6 ± 2.9	0.62	55	10.2 ± 1.0
Control cell	0.6	1.66	0.7	31.2 ± 1.0	0.71	64.3 ± 2.0	14.6 ± 0.3
First ELO cell	128	1.87	0.5	31.8 ± 0.1	0.70	64.4 ± 1.9	14.4 ± 0.4
Second ELO cell	17	1.71	0.7	31.3 ± 0.3	0.71	66.1 ± 0.3	14.8 ± 0.2

^aReference 12.

surface after eliminating the native oxide exhibits the same 2×4 reconstructions as the original wafer (inset, Fig. 2). The streaky pattern also indicates that the roughness has not increased on epitaxial layer removal.

Orange-peel-like roughness was observed on the surface exposed to the ELO etching process in the absence of a protection layer. This is apparent from the three-dimensional (3D) surface profiles in Fig. 3, inset, that compare the millimeter-scale surface morphology between ELO processed substrates with and without the protection layer. The surface images confirm the necessity of employing the protection layer to ensure that the regrowth surface is flat on both the macro and nanoscales.

The fourth quadrant current density-voltage (J-V) characteristics of the first and the second ELO processed photovoltaic cells, and the control device measured under simulated Atomospheric Mass 1.5 Global (AM 1.5G) illumination at 1 sun (100 mW/cm²) intensity are compared in Fig. 3. The characteristics are nearly identical for all devices (see Table I). The reverse-bias dark current (J_D) at -1 V for the control cell is lower than that of either of the ELO cells, as also listed in Table I. However, there is no increase in dark current with number of ELO steps employed, indicating that these differences are due to run-to-run variations in the ITO/ InP Schottky barrier formation process rather than from systematic differences in the epitaxial layer quality. Slight variations in Schottky barrier height and J_D are observed for different ITO sputtering depositions and for different positions on the sample holder.

The ideality factors (*n*) and specific series resistances (R_S) are extracted using the Shockley equation modified to include R_S , $J=J_0 \exp[q(V-IR_S)/nk_BT]+(V-JR_S)/R_P$, where J_0 is the reverse bias saturation current, *q* is the electron charge, *V* is applied voltage, k_B is the Boltzmann constant, *T* is temperature, and R_P is specific parallel resistance.¹⁴ The first- and the second-growth solar cells exhibit no systematic performance loss under 1 sun intensity, with $\eta_p=14.4\%$ and 14.8%, respectively. A compilation of parameters from fits to the characteristics are provided in Table I.

The significant improvement in device performance compared with previous reports of InP solar cells prepared by total substrate removal¹² primarily results from the increased V_{oc} and fill factor (FF) due to the improved ITO/InP interface. To reduce the contact surface damage, a higher ITO sputtering power has been combined with the UV/ozone treatment to form a thin oxide passivation and surface protection layer. The increased sputtering power also leads to an increase in ITO conductivity.

In summary, we have demonstrated the fabrication of ITO/InP thin-film solar cells sequentially grown multiple

times on a single InP parent substrate. After each growth, the epilayers were removed by ELO, and then cold-weld bonded at room temperature to a thin, flexible plastic sheet. The optimized fabrication processes lead to similar performance for both ELO processed and control cells, with $\eta_p = 14.4 \pm 0.4\%$, $14.8 \pm 0.2\%$, and $14.6 \pm 0.3\%$, respectively. Furthermore, including protective epitaxial layers results in an undamaged surface, allowing for its continued reuse. Hence, this work demonstrates a reduced-cost fabrication method for ITO/InP thin-film solar cells based on continuous reuse of a single substrate without the loss of device performance.

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