GaAs Substrate Reuse Using Molecular Beam Epitaxy of NaCl Layers

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High-efficiency photovoltaics require single-crystal III-V substrates which are very expensive and account for nearly 1/3 of the end cost of the device. [1] Several methods for substrate reuse techniques have been demonstrated using mechanical means or the dissolution of a sacrificial release layer. However, many of these techniques are slow due to re-polishing steps, involve hazardous wet etches, limit heterostructure flexibility, and leave behind insoluble etch byproducts. This work presents an alternative method to conventional III-V substrate recycling methods via growth of NaCl on nearly-lattice matched GaAs (001) substrates. This NaCl liftoff method does not leave behind insoluble etch byproducts or limit future heterostructure designs due to the solubility in water.

Both NaCl and GaAs thin films were deposited on GaAs (001) substrates in an Epi930 MBE reactor with base pressure of 4×10^{-9} torr. Reflection high energy electron diffraction (RHEED) with beam energy of 15 keV was employed both as an *in-situ* diagnostic tool as well as a source of high energy electrons to help promote nucleation of GaAs on NaCl. [2] To ensure a clean and oxide free surface for subsequent GaAs and NaCl deposition, the GaAs substrates were heated to 620° C for 25 minutes under exposure to excess As flux from a valved cracker source. Following this, a 300 nm GaAs buffer layer was deposited at 580° C with As/Ga = 1.68, and subsequently cooled to 150° C before an NaCl layer was deposited using congruently sublimated NaCl from a conventional effusion cell operating at ~480°C. After NaCl deposition at a rate of 3 nm/min, the RHEED beam was moved across the surface until the diffraction pattern dimmed. After this, the sample was heated at a rate of 20° C/min to the temperature desired for an initial low temperature GaAs nucleation step ($350-400^{\circ}$ C) before high temperature (580° C) growth, using a Ga flux that would give a growth rate of ~33 nm/min (with co-deposition) measured using RHEED oscillations at 580° C.

High-resolution x-ray diffraction (XRD) was performed on a Rigaku Smartlab. Scanning electron microscope (SEM) and electron backscatter diffraction (EBSD) was used to look at surface morphology and crystallographic properties ex-situ. EBSD data were acquired in an Oxford system using a Symmetry detector with a CMOS sensor. The acquisition voltage was 20 kV, and the analysis was performed with the sample tilted by 70°. Transmission electron microscopy (TEM) images and electron diffraction patterns were acquired a with JEOL 2100F. The GaAs substrate was tilted so that incident electrons were along <110> and an accelerating voltage of 200 kV was used. Bright field TEM imaging was performed to show the layers and atomic structure of defects. Electron diffraction patterns of areas of 100 nm in diameter were acquired to identify the local phases and crystalline orientation. For removal of the GaAs film from the substrate, the film was first attached to Kapton tape and then placed in water to dissolve through the NaCl layer.

Figure 1 (a) shows a scanning transmission electron microscopy (STEM) image with fast Fourier transforms (FFTs) of both the NaCl and GaAs substrate. The FFTs reveal that the NaCl layer is oriented the same as the substrate and has a nearly identical lattice constant. Figures 1 (b) & (c) shows a STEM



image and the corresponding energy dispersive X-ray spectroscopy (EDS) line profile. Figure 2 compares TEM and diffraction patterns between a RHEED exposed area and a non-RHEED exposed area to understand the effect of RHEED on the nucleation of GaAs on NaCl.

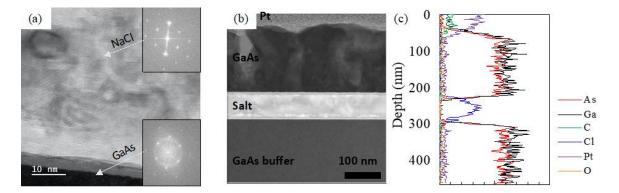


Figure 1. (a) STEM image of the NaCl layer with insets showing FFTs of the (top) NaCl layer and (bottom) GaAs interfacial area (b) STEM image of a NaCl layer capped with GaAs, and (c) EDS linescan of the NaCl layer contained between GaAs layers.

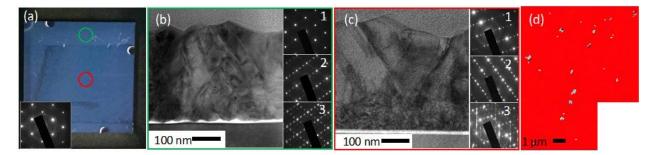


Figure 2. (a) Image showing the central area exposed to the RHEED beam prior to GaAs deposition (inset) STEM diffraction pattern of the substrate. STEM images of areas (b) never exposed to RHEED (from green circle) and (c) exposed to RHEED prior to GaAs deposition (from red circle) and the corresponding diffraction patterns from (1) the top of the film, (2) the center of the film, and (3) near the NaCl interface. (d) EBSD orientation map from the RHEED exposed area.

References:

- [1] K.A. Horowitz, T.W. Remo, B. Smith, and A.J. Ptak, Technical Report, https://www.nrel.gov/docs/fy19osti/72103.pdf (2018).
- [2] B.J. May, J.J. Kim, P. Walker, H.R. Moutinho, W.E. McMahon, A.P. Ptak, and D.L. Young, ACS Omega, *submitted* 2022
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