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Hybrid GaAs nanowire-polymer device on glass: Al-doped ZnO (AZO) as transparent conductive oxide for nanowire based photovoltaic applications

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1. Introduction

III-V semiconductor nanowires (NWs) are being actively pursued to build next generation solar cells [1–5]. A common consensus among the NW research community is that the NW based solar cell could be the logical progression to improve dramatically the efficiency and costs associated with mainstream Si based solar cells. Currently, the best NWs (GaAs) based solar cell shows power conversion efficiency of 15.3% [1], which is likely to be improved in the near future.

Today, commercially available Si (polysilicon) and CdTe thin film based solar cells in the market show average power conversion efficiency of about 17% [6]. Year-on-year, the prices of Si solar panels have fallen significantly but further cost reduction is needed for solar cells to be deployed at a large-scale in majority of the households. Therefore, an important factor in solar cell research is to increase the efficiency and reduce the device cost at the same time. III-V NWs fits this criteria well, as unlike Si, they have direct band gap (which means less material consumption compared to Si; ~1 µm thick GaAs absorbs an equivalent amount of light as ~100 µm thick Si), high carrier mobility [7] and III-V NWs can easily be integrated on lattice mismatched Si substrate [8].

Further, unlike the conventional solar cells, no antireflection coating is needed for NW solar cells because NWs are excellent light trappers wherein NWs act as light concentrators similar to a mirror (a single NW can concentrate 15 times the sun light [3]). For this reason, a single p-n junction based on NW can even exceed the theoretical Shockley-Queisser efficiency [3] limit of ~33%. III-V NWs grown on Si substrate is an ideal paradigm of achieving record high efficiencies solar cells at lower costs.

Another possibility the NWs offer is the growth on much cheaper substrates than Si, such as glass [9,10]. However, NW based solar cells on glass are not directly viable due to the non-conducting surface of glass. To circumvent this limitation, one option is to explore the NW growth on glass coated with transparent conductive oxide (TCO). In pursuit of future NW based solar cells based on axial or radial p-n junction, an important and crucial component is the search for a low-cost TCO material for top or the bottom contact. So far, indium tin oxide (ITO) is the preferred choice of TCO [11,12] in industrial applications. However, ITO is becoming more expensive due to depleting indium reserves worldwide and is also toxic to the environment. The desirable characteristics of a material to qualify as TCO is to have resistivity of the order of 10⁻³ Ohm cm and optical transmittance exceeding 80% [11,12]. Al-doped ZnO (AZO) [13,14] is fast emerging as an alternative low-cost TCO. AZO is inexpensive, non-toxic and can be readily deposited in large scale at low-cost using atomic layer deposition (ALD) [13,14]. In this report, we add functionality to glass substrate by coating it with optimized AZO as transparent TCO and subsequent GaAs NW growth on TCO. By fabricating a simple GaAs NW-polymer hybrid device on glass, we show that ALD grown AZO is an excellent TCO as well as a promising growth substrate for GaAs NWs.

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2. Experimental

Al-doped ZnO (AZO) growth using atomic layer deposition: AZO films were deposited using a Beneq (TFS 500) ALD system on borosilicate glass. Prior to ALD, the samples were rinsed in isopropanol and in DI water. Precursors for zinc, aluminum and oxygen were diethylzinc (DEZn), trimethyldiaminotin (TMAI) and \( \text{H}_2\text{O} \), respectively, and nitrogen was used as a carrier gas. Reactor temperature during the deposition was kept at 260 °C and the pressure was \( \sim 2 \text{ mbar} \). Typical growth rate was 6 nm/min. A deposition loop consisted of 30 cycles of ZnO followed by one cycle of \( \text{Al}_2\text{O}_3 \), yielding an aluminium content of \( \sim 3 \text{ at.-%} \) in the film.

**GaAs nanowires growth:** GaAs NWs were fabricated on AZO coated glass substrates in a horizontal flow atmospheric pressure metal organic vapor phase epitaxy (MOVPE) system with trimethylgallium (TMGa) and tertiarybutylarsene (TBAs) as precursors. Diethylzinc (DEZn) was used as a dopant. Hydrogen was used as a carrier gas and the total reactor gas flow rate was \( \sim 5 \text{ l/min} \). 40 nm diameter colloidal gold (Au) nanoparticles were used as catalysts for the vapor–liquid-solid (VLS) growth. For proper adhesion of Au nanoparticles, the poly-L-Lysine (PLL) was applied to the substrate for 1 min. Prior to the actual growth, the samples were annealed in-situ at 500 °C for 3 min to desorb surface contaminants. MOVPE growth was started by switching on the appropriate sources simultaneously. The growth duration was 1 min yielding \( \sim 4 \mu\text{m} \) long NWs. The nominal V/III ratio during the growth was \( \sim 25 \), and the growth temperature was 470 °C. The MOVPE temperatures reported in this work are thermocouple readings of the lamp-heated graphite susceptor, which are somewhat higher than the real glass substrate’s surface temperatures as glass is a poor conductor of heat.

**Structural characterisation and transmission measurements:** Structural properties of the GaAs NWs and AZO films were studied using scanning electron microscopy (SEM) (Zeiss Supra 40). Light transmission measurements were performed using a Perkin Elmer Lambda 850 Uv–Vis spectrometer.

3. Results and discussion

Due to their lateral nanoscale dimension and specific growth process, NWs offer possibilities for growth on a wide variety of low-cost substrates \([9,10,15,16]\). We have demonstrated earlier that high quality GaAs NWs can be grown directly on a soda-lime (window) glass substrate \([9]\) (Fig. 1a-b). The remarkable properties of the GaAs NWs on glass include a single-phase zinc-blende (ZB) structure and strong photoluminescence emission light even at the room temperature which manifest the high quality of the NWs. Recently \([10]\), we have also reported the growth of ultra-long InP NWs with a record growth rate \( \sim 25 \mu\text{m/min} \) on a glass substrate with crystal quality comparable to NWs grown on Si substrate. Since glass consists of a large quantity of silica \((\sim 70–80\%)\), the Au-Si liquid eutectic alloy for glass is expected to be similar to the Au-Si alloy on a Si substrate indicating similar VLS growth temperature window \([10]\). Therefore, in principle, majority of III-V NWs can be grown readily on transparent glass. Generally, NW growth on glass offers many advantages such as low-cost, light transparency and ease of NWs transfer to a target substrate without breakage by using a short hydrofluoric acid (HF)etch \([10]\). However, as stated previously, a disadvantage associated with glass is its non-conducting surface, thereby making it challenging for NW based solar cells. To circumvent this limitation, one possibility is the deposition of thin buffer layers of TCO such as ITO or AZO on glass, and subsequent NW growth on TCO. In that respect, for GaAs NWs, among the two, only AZO offers the possibility of Au-assisted growth on its surface using the commonly used PLL-mediated Au nanoparticle deposition \([15]\) (Fig. 1c-d). Although growth on ITO is also possible, it requires more specialized Au nanoparticle deposition methods \([17,18]\).

To realize the discussed possibility, we reported previously \([15]\) the successful Au-assisted growth of GaAs NWs on ALD grown AZO (TCO) thin films as shown in Fig. 1c-d. In that work \([15]\), the AZO films were grown at 210 °C with observation of worm-like structure and with a large grain size (Fig. 1c), which is a typical feature for AZO films. Typically, a large grain size of the film results in less scattering of carriers thereby increasing the conductivity (decrease in the sheet resistance). However, GaAs NWs grown on this AZO with rough morphology (uneven large grain size) typically suffers from initial crawling and kinking on the surface as can be observed in Fig. 1d. We reported resistivity of \( 2.5 \times 10^{-3} \text{Ωcm} \) for these films \([15]\).

In this report, we have further optimized the AZO growth parameters in order to improve the surface morphology of the films. A detailed growth process for AZO films grown using the ALD process is provided in the experimental section. As shown in Fig. 2(a), as-grown AZO film grown at 260 °C shows relatively smooth surface morphology (smaller grain size) compared to our previous report. The as-grown AZO film (ZnO doped with \( \sim 3 \text{ at.-% aluminum, as-grown AZO is n-type} \)) is polycrystalline and shows a resistivity of \( 5 \times 10^{-3} \text{Ωcm} \) as measured by Van der Pauw method using indium contacts. Similar values were reported elsewhere \([19,20]\). The resistivity value reported here is two times more than mentioned in our previous work, while reduction in the grain size and increased surface roughness of the AZO film can be observed in Fig. 2a. In our previous work, the GaAs NW growth initiated as in-plane growth on the AZO surface, and subsequently switched to out-of-plane growth. Here, it is assumed that the different surface morphology of the AZO film favors out-of-plane growth at an earlier stage of the growth. In addition, an increase in resistivity could be attributed to a smaller grain size (more carrier scattering) \([19,20]\).

In literature, for ALD grown AZO films, the best reported resistivity is \( 7 \times 10^{-4} \text{Ωcm} \) \([13]\). Overall, the best AZO resistivity is reported to be \( 2 \times 10^{-4} \) using the magnetron sputtering \([13]\). On the other hand, the best commercially available ITO coating in the market show resistivity in the range \((1–3) \times 10^{-4} \text{Ωcm} \) \([11,12]\). Hence, compared to ITO, we see that the achieved resistivity for AZO is reasonably good.

Further, as shown in the inset of Fig. 2b, a continuous 300 nm thick AZO film is conformally deposited on all sides of the glass substrate thus making its surface conducting. Consequently, using a custom-made shadow mask, Au (80 nm)/Ti (6 nm) contacts were evaporated on the top and bottom of the AZO/glass/AZO structure (Fig. 2b). Current-voltage (IV) measurements (as shown in inset of Fig. 2b) shows that the Au/Ti contacts to AZO (n-type) yield perfect ohmic behavior with the current flow in milliamperes (mA) range. Furthermore, as shown in Fig. 2(c), the 600 nm thick AZO film wrapped around glass shows light transmission of \( > 85–90\% \) in visible to near infrared region (500–1200 nm). These light transparency values are similar to the ITO \([11,12]\). Next, GaAs NWs (40 nm diameter, \( \sim 4 \text{μm} \) length) were directly grown on AZO coated glass using metalorganic vapor phase epitaxy (MOVPE). As seen in Fig. 2(d), GaAs NWs with high density are visible on a 300 nm thick AZO film deposited on glass. The NWs are consistent in length, diameter and density throughout the substrate. The good visual appearance of GaAs NWs seen here is due to the improved AZO surface. Further, as discussed previously, GaAs NWs grown on AZO typically suffers from initial crawling and kinking on the surface (Fig. 1d). In contrast, such tendency is significantly suppressed in this work by growing the GaAs NWs on an optimized AZO surface, and by performing an additional 5 min pre-growth annealing step at 450 °C.

The optical quality of the GaAs NWs grown on AZO was studied by photoluminescence (PL). Fig. 3a shows the PL spectra were obtained at both room temperature and at 45 K in order to estimate the internal quantum efficiency (IQE) of the NWs (SEM image of the measured sample is shown in Fig. 3b). The NWs were slightly Zn-doped in order to enhance the PL intensity \([21]\). The IQE was estimated by comparing the integrated PL intensity at room temperature to that in 45 K as follows:

\[
\text{IQE} \sim \frac{\text{PL}_{300K}}{\text{PL}_{45K}} \times 100\%
\]

This suggests that the IQE is \( \sim 55\% \), while it should be noted that the IQE is in reality somewhat lower, since the
low-temperature measurement was moderately higher than ideal 0 K.

To evaluate the suitability of AZO as TCO for GaAs NWs, a simple NW-polymer hybrid device was fabricated to study the electrical contact between the NW-TCO and the Au/Ti interface, the schematic for which is presented in Fig. 4(a). In the first step, the n-type GaAs NWs were covered with benzocyclobutene (BCB) polymer from Dow Chemicals. BCB polymer is chosen since it has excellent reflow, planarization and insulating properties. After the dropcast, the BCB polymer was cured for 1 h at 250 °C in hydrogen ambient. This resulted in pinhole free 2.5 µm thick BCB film partially covering the GaAs NWs as shown in Fig. 4(b). Next, a short oxygen plasma was performed to remove the BCB residues from the exposed top part of the GaAs NWs. After this procedure, only the tips of NWs were visible, while many of the NWs remained fully buried under the BCB. Fig. 4c shows the top view SEM image of the GaAs NWs covered with BCB. A clear contrast between the protruding NWs and the ones buried within the polymer can be seen clearly (Fig. 4c). Next, the Au (80 nm)/Ti (6 nm) contacts were evaporated on top of the BCB applied NWs for the top contact. Further, for the bottom contact, the Au (100 nm)/Ti (30 nm) layers were already evaporated before the GaAs NW growth on AZO as shown in Fig. 4a. In this simple device, the current pathway is from the top Au/Ti to the NW-AZO interface via NWs and further to the Au/Ti contacts.
at the bottom of the device. This simple device design eliminates any pathway of error in the I-V measurements. Since we already know that the electrical contacts between AZO and Au/Ti are ohmic, for an overall ohmic behavior of the device, also the contacts between the Au/Ti-NW and the NW-AZO interfaces need to be ohmic. If either of these two interfaces is of Schottky-type, that in turn will render the overall carriers pathway to non-ohmic. Fig. 4 (d) shows the I-V measurement between the top and the bottom contact of the hybrid device. As can be seen, a perfect ohmic behavior is observed between the top and bottom contacts. However, the current flow between the contacts is in microampere range. The reason for such a low current (the current flow values are in milliampere range without NWs) could either be the high contact resistance of Au/Ti at the top of the NW or surface depletion effects present in the GaAs NWs [22]. Since GaAs NWs are known to be very sensitive to the surface states, therefore, in the absence of passivation, that could result in high resistivity of NWs [22]. We believe that the device performance could be improved further by using the surface passivated GaAs NWs [23–25]. Thus, the results of this NW-hybrid confirm that AZO can serve as good TCO as well as excellent low-cost growth substrate for GaAs NWs. It is to be noted here that a similar NW-polymer device was demonstrated earlier [26]. However, in that device InP NWs were grown on an ITO coated glass. We believe that using this simple geometry and replacing BCB with a transparent planarization material such as spin-on-glass, axial or radial p-n junction solar cells can be fabricated on AZO coated glass. An advantage of such a solar cell would be that the substrate, contacts and the planarization material are transparent from bottom to the top. Thus, the sun light can be directed into the device either from the top or the bottom. In the latter case, glass will also provide an excellent hermetic seal from the environment.

4. Conclusion

We have successfully demonstrated a simple GaAs-NW polymer hybrid device on AZO coated glass substrate. The device confirms that the AZO on glass can serve both as TCO as well as a low-cost growth platform for GaAs NWs. Based on this concept, a unique device idea is Fig. 3. Internal quantum efficiency (IQE) measurement of GaAs nanowires. a) PL spectra measured at T = 45 K and T = 300 K. IQE was calculated from ratio of PL signal. (b) SEM image of the measured NWs.

Fig. 4. (a) Schematic for GaAs NW- BCB polymer hybrid device, (b) SEM image of GaAs NWs partially immersed in insulating BCB polymer, (c) SEM image of the top view of the device showing some NWs tips exposed and (d) IV measurement showing perfect ohmic behavior between the top and bottom contact of the device (0.16 cm² active area).
proposed to fabricate semi-transparent GaAs nanowire based solar cells on TCO coated glass in the future.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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