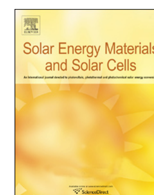
Solar Energy Materials & Solar Cells 120 (2014) 376–382

Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat



Metal-oxide-based hole-selective tunneling contacts for crystalline silicon solar cells

S. Smit [a,*], D. Garcia-Alonso [a], S. Bordihn [a,b], M.S. Hanssen [a], W.M.M. Kessels [a]

[a] Department of Applied Physics, Eindhoven University of Technology, P.O. Box 513, 5600MB Eindhoven, The Netherlands
[b] Hanwha Q.Cells GmbH, Sonnenallee 17-21, 06766 Bitterfeld-Wolfen, Germany

ARTICLE INFO

Available online 10 July 2013

Keywords:
Atomic layer deposition
Passivation
Tunneling
c-Si solarcells
Heterojunctions
Metal oxides

ABSTRACT

The goal of this work is to investigate selective hole contacts for crystalline silicon solar cells that are highly transparent, passivate the silicon surface and have low contact resistance. Stacks of Al_2O_3 and ZnO films are suggested for this purpose. The charge transport mechanism through these stacks is tunneling recombination and it is shown that such stacks can achieve a contact resistance of ~1.5 Ω cm^2 for an Al_2O_3 thickness of 1 nm. Furthermore, it is demonstrated that the surface passivation of such stacks can be greatly improved by the insertion of a 3 nm film of hydrogenated amorphous silicon (*a*-Si:H) between the Al_2O_3 and the crystalline silicon, achieving an effective surface recombination velocity of ~20 cm/s. The stacks with an *a*-Si:H layer achieve a contact resistance of ~5 Ω cm^2. Furthermore, from applying the theory of tunnel diodes to the charge transport through the contact, three important elements have been identified for the reduction of the contact resistance: the negative fixed charge density in the Al_2O_3; the doping concentration in the ZnO; and the dielectric properties of the Al_2O_3.
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1. Introduction

In all solar cells that rely on the principle of generation of free electrons and holes, two key components can be identified: the photon absorber and the selective contacts [1]. In the photon absorber, the energy of the light is converted into chemical energy in the form of electron–hole pairs. This conversion is subject to the thermodynamic limit on efficiency, similarly to any other conversion of heat to a form of higher-grade energy. Next, the selective contacts in the cell force the electrons to exit the absorber along a different path than the holes, converting the chemical energy into work by creating an electric current. In principle, the efficiency of this second conversion is not subjected to thermodynamic limitations because no heat is involved. It is the conversion from chemical energy to work that is the focus of the research described in this paper.

The selective contacts in a traditional crystalline silicon (*c*-Si) solar cell are the diffused emitter and the back surface field (BSF). High *n*- and *p*-doping densities of these contacts result in an increase of the conductivity for one carrier type while the conductivity for the other carrier type is simultaneously reduced.

In silicon heterojunction (Si-HJT) solar cells, it is not just the doping in the deposited hydrogenated amorphous silicon (*a*-Si:H) layers that makes the emitter and rear of the cell selective to one type of carrier, but the band alignment at the *a*-Si:H/*c*-Si interface plays a significant role as well [2]. In fact, higher selectivity can be achieved than with diffused junctions by tuning the valence and conduction band offsets at the *a*-Si:H/*c*-Si interface, which leads to the well-known high V_{oc} values of Si-HJT cells [2]. One disadvantage of the *a*-Si:H layer is that they absorb a portion of the light without converting it into usable current, a phenomenon known as parasitic absorption [2].

The goal of our research is to investigate the possibility to create selective contacts by depositing stacks of selected materials on *c*-Si. The general boundary conditions under consideration in this work (other than high selectivity to either electrons or holes) are:

(a) The interfaces between the layers and the *c*-Si should be well-passivated to avoid surface recombination. An effective surface recombination velocity of 100 cm/s is considered the maximum feasible limit.
(b) The contacts should be highly transparent. The light absorption of the contact should be less than the parasitic absorption in Si-HJT solar cells to be considered feasible.
(c) The contact resistance should be low enough to compete with current technologies. Ultimately, the goal is to obtain a contact resistance on the order of 50 mΩ cm^2.

Other considerations are that the deposition methods used should be industrially scalable and the materials used should be abundant and preferably environmentally friendly.

Abbreviations: *a*-Si:H, Hydrogenated Amorphous Silicon; ALD, Atomic Layer Deposition; BSF, Back Surface Field; *c*-Si, Crystalline Silicon; HIT, Heterojunction with Intrinsic Thin layer; ICP-CVD, Inductively-Coupled Plasma Chemical Vapor Deposition; PVD, Physical Vapor Deposition; SE, Spectroscopic Ellipsometry; Si-HJT, Silicon Heterojunction; SRV, Surface Recombination Velocity; TCO, Transparent Conductive Oxide; XPS, X-ray Photoelectron Spectroscopy; ZnO, Al: Aluminum-doped ZnO.

* Corresponding author. Tel.: +31 40 2474919.
E-mail address: s.smit@tue.nl (S. Smit).

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http://dx.doi.org/10.1016/j.solmat.2013.06.016

With these requirements in mind, stacks of Al_2O_3 and ZnO films are suggested to ultimately achieve these goals. The choice for Al_2O_3 is motivated by the advances in recent years in silicon surface passivation by Al_2O_3 films, prepared by atomic layer deposition (ALD) or otherwise [3,4], making Al_2O_3 a very promising material to fulfill requirement a). The choice for ZnO is based on the combination of its high bandgap (and thus high transparency), low resistivity and natural abundance.

In this work, the focus is on three points:

(i). To assess the potential of stacks of Al_2O_3 and ZnO films as passivating, transparent and selective contacts.
(ii). To gain a theoretical understanding of charge transport through these stacks.
(iii). To find ways to improve their performance.

Point (i) is addressed in Sections 3.1 and 4.1. Section 3.1 will demonstrate how the negative fixed charge density at the Al_2O_3/*c*-Si interface [4] can be used to create a homojunction that separates electrons from holes. Section 4.1 will show the experimental results on the passivation of *c*-Si by the Al_2O_3/ZnO stacks.

Point (ii) is addressed in Sections 3.1, 3.2 and 4.2. In Section 3.1 the charge transport mechanism through the Al_2O_3/ZnO stacks will be identified as tunneling recombination and will be discussed in more depth in Section 3.2. Section 4.2 will show that the measured currents corroborate the theory presented in Section 3.2.

Point (iii), which is addressed in Sections 3.3 and 4.3. Section 3.3 will identify from a theoretical perspective three key elements in the Al_2O_3/ZnO stacks that influence the current through them and that can be optimized to reduce the tunneling resistance. It will also discuss the potential of amorphous silicon interlayers to reduce tunneling resistance and increase passivation. Section 4.3 demonstrates that the passivation of the stacks can be significantly improved by the insertion of an *a*-Si:H interlayer between the *c*-Si and Al_2O_3. The *a*-Si:H films that have been used are significantly thinner than those used in typical Si-HJT cells.

2. Material and methods

Both the surface passivation and the tunneling current investigations were performed on two types of stacks: Al_2O_3/ZnO:Al (type A) and *a*-Si:H/Al_2O_3/ZnO:Al (type B). These stacks are shown in Fig. 1a.

For the passivation study, low resistivity (~3–4 Ω cm) *n*-type and *p*-type *c*-Si wafers (281 ± 1.3 μm thick) were used. The wafers were treated with a diluted HF (1% in DI-H_2O) solution right before carrying out the first film deposition. The surface passivation was always investigated on samples with the thin films deposited on both sides symmetrically.

The 1-nm-thick Al_2O_3 and 100-nm-thick ZnO:Al films were deposited using an ALD open-load reactor (OpAL, Oxford Instruments). A plasma ALD process was used for the deposition of the Al_2O_3 films ($Al(CH_3)_3$, O_2 plasma) at 200 °C and a thermal ALD process was used to deposit the ZnO films ($Zn(C_2H_5)_2$, H_2O vapor) at 180 °C. The ZnO films were doped with different Al concentrations by varying the ALD cycle ratio *x* (1 Al_2O_3 doping ALD cycle after every *x* ZnO ALD cycles). For stacks of type A, the Al doping concentration (represented as Aluminum fraction (*AF*)) was varied from *AF*=0 to 0.31; *AF* being defined as *AF*=[Al]/ ([Al]+[Zn]), with [Al] and [Zn] are the atomic percentages of Al and Zn as measured by X-ray photoelectron spectroscopy (XPS). For stacks of type B, *AF*=0.10.

The *a*-Si:H layers of different thicknesses (1.5–6 nm) were deposited at 50 °C by inductively-coupled plasma chemical vapor deposition (ICP-CVD) (PlasmalabSystem100 ICP 180, Oxford Instruments) using SiH_4 gas.

The stacks of type A were annealed at 400 °C for 5 min in a controlled N_2 atmosphere before the ZnO:Al film deposition. For stacks of type B, the annealing was carried out at a lower temperature of 300 °C for 1 min in a controlled N_2 atmosphere to prevent loss of hydrogen in the *a*-Si:H. The annealing was performed after the complete stack was deposited. The passivation performance of the stacks was evaluated from the effective lifetime, τ_{eff}, of the minority carriers on the silicon wafers. τ_{eff} was determined with photoconductance decay in the transient mode and quasi steady-state-mode (for $\tau_{eff} < 100$ μs) using a Sinton lifetime tester (WCT 100). The upper level for the effective surface recombination velocity (SRV), $S_{eff,max}$, was extracted at an injection level of $1 \cdot 10^{15}$ cm^{-3} by the expression $S_{eff,max} = thickness$ Si-*wafer*/$2\tau_{eff}$. Spectroscopic Ellipsometry (SE) was employed to measure the thickness of each layer of the stacks. SE was also used to measure the optical bandgap ($E_{g,opt}$) and the carrier concentration (n_e) of the ZnO:Al films using a Drude model [5].

Current density–voltage (*J*–*V*) analysis was carried out to test the tunneling behavior of stacks type A and type B. The front side of the tunneling test structure consisted of the stack under investigation and the rear-side of a 10-nm-thick Al_2O_3 single film. On both sides 2-μm-thick Al metal contacts with an area of 1.96 cm^2 were deposited by physical vapor deposition (PVD). Subsequently, high quality p^+-doped contacts with a pitch of 200 μm were produced by laser firing [6] on the rear side. Fig. 1b shows a schematic cross section of the tunneling samples and the stack configurations used.

3. Theory

3.1. The ZnO/Al_2O_3/c-Si system as a selective hole contact

To demonstrate how Al_2O_3 stacks can function as a selective hole contact, Fig. 2 shows the calculated band diagram of the ZnO/Al_2O_3/*c*-Si system, which is based on AFORS-HET simulations [17]. In the simulation, a negative fixed charge density (Q_f) of 5×10^{12} q cm^{-2} [4] has been assumed at the interface between Al_2O_3 and Si. The values for band gaps and band offsets (eV) have been taken from the literature [4,7,8]. It is known that there is also an SiO_x layer between the Al_2O_3 and *c*-Si at the interface [4]. This oxide is not included in the model explicitly, but its presence was taken into account by using an effective dielectric constant $k=6$. This is valid because only the capacitance of the oxide interlayer has an influence on the equilibrium band diagram calculation. The SiO_x/Al_2O_3 thickness in the model is 5 nm. It should also be noted that the distance of the Fermi level in the ZnO to the conduction band is larger in reality than this simulated band diagram shows. This is due to the fact that AFORS-HET is limited to Maxwell–Boltzmann statistics, while Fermi–Dirac statistics are needed to correctly determine the Fermi level in a degenerate semiconductor.

The simulation demonstrates how the Al_2O_3/ZnO stack serves as a hole-selective contact on the *c*-Si. The high density of negative fixed charges at the interface leads to accumulation of holes at the Al_2O_3/*c*-Si interface. Similarly to a diffused junction, in the inversion layer the conductivity for holes is much higher than for electrons, so the inversion layer serves as a homojunction. However, the doping is achieved without adding impurities to the silicon, thus keeping the bulk recombination low.

If the film is thin enough, a tunneling current through the Al_2O_3 is possible by recombination of electrons from the ZnO and holes in the Si without a significant voltage loss due to the alignment of the ZnO conduction band with the *c*-Si valence band. This is

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