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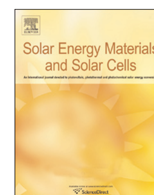

"Zero-charge" SiO_2/Al_2O_3 stacks for the simultaneous passivation of n^+ and p^+ doped silicon surfaces by atomic layer deposition

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ABSTRACT

To achieve high conversion efficiencies, advanced silicon solar cell architectures such as interdigitated back contact solar cells demand that defects at both the n^+ and p^+ doped Si surfaces are passivated simultaneously by a single passivation scheme. In this work, corona charging experiments show that the fixed charge density Q_f is a key parameter governing the passivation of both surface types. Alternatively, Q_f can be controlled from strongly negative to even positive values by carefully tuning the SiO_2 interlayer thickness in SiO_2/Al_2O_3 stacks prepared by atomic layer deposition (ALD). This control in Q_f allows for a superior passivation of n^+ Si surfaces by SiO_2/Al_2O_3 stacks compared to a single layer Al_2O_3. For instance, for SiO_2 interlayer thicknesses of ~3–14 nm, the recombination parameter of an n^+ Si surface having a high surface doping concentration N_s of 2×10^{20} cm^{-3} was reduced from $J_{0n^+}=81$ fA/cm^2 to $J_{0n^+}=50$ fA/cm^2. Simulations predict that the SiO_2/Al_2O_3 stacks outperform Al_2O_3 passivation layers particularly on n^+ Si surfaces having a moderate N_s in the range of 10^{18}–10^{20} cm^{-3}. On p^+ Si surfaces, $J_{0p^+}\leq54$ fA/cm^2 was achieved for all ALD SiO_2 interlayer thicknesses investigated (i.e., 1–14 nm). The SiO_2/Al_2O_3 stacks presented in this work are compatible with SiN_x capping and subsequent high-temperature firing steps, which are typically used in solar cell processing. Furthermore, the results were successfully reproduced in an industrial ALD batch reactor using a low-temperature process. This makes ALD SiO_2/Al_2O_3 stacks a promising candidate for the simultaneous passivation of n^+ and p^+ Si surfaces in solar cells.

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1. Introduction

A high level of surface passivation is a prerequisite to achieve crystalline silicon solar cells with high conversion efficiencies. Less than a decade ago, it was reported that Al_2O_3 films prepared by atomic layer deposition (ALD) provide superior passivation of p and p^+-type Si surfaces, which was technologically challenging at that time [1–3]. The excellent passivation by Al_2O_3 can be related to a very low interface defect density D_{it} ($<10^{11}$ cm^{-2}) on Si, which is essential for *chemical* passivation. Furthermore, a high *negative* (fixed) charge density Q_f in of the order of 10^{12}–10^{13} cm^{-2} (depending on synthesis method) is present at the interface [4], which reduces the minority carrier (i.e., electron) concentration near the Si surface, providing *field-effect* passivation. Even an ultra-thin film of Al_2O_3 of less than 2 nm was found to be sufficient to passivate the Si surface, when combined with a-SiN_x:H (in short SiN_x) as anti-reflection coating (ARC) and/or capping layer [5,6]. The high levels of surface passivation provided by Al_2O_3 allow for solar cells with high conversion efficiency, including p-type concepts (e.g, PERC or Al–LBSF cells [7,8]) as well as n-type concepts such as PERL cells [9]. Due to superior uniformity and passivation performance, ALD of Al_2O_3 is currently piloted in industry [10].

Despite the excellent results on p-type Si, on heavily doped n^+ Si surfaces the passivation of Al_2O_3 is compromised, as the minority carrier (i.e., hole) concentration at the surface is *increased* by the negative Q_f of the Al_2O_3 [11,12]. On lowly doped n-type Si, Al_2O_3 generally passivates the surface well, as it induces strong near-surface inversion. However, inversion layers are associated with a lifetime reduction at low injection levels ($\Delta n<10^{15}$ cm^{-3}), which is within the operating regime of solar cells [13–15]. Secondly, besides affecting the surface passivation, inversion layers potentially act as undesired conduction pathways to metal contacts. Such a pathway (sometimes referred to as parasitic shunting) affects the performance of solar cells via the fill-factor, short-

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circuit current and open-circuit voltage [16]. All of these unfavorable effects occur not only when n or n^+-type Si surfaces are passivated by the negative charge dielectric Al_2O_3, but they also take place when p or p^+ type Si surfaces are passivated by a dielectric containing a *positive* charge density, such as SiN_x.

In conventional diffused-junction Si solar cells, the n^+ and p^+ doped surfaces are located at each side of the cell, and hence can be passivated independently by a dielectric having the right charge polarity. However, in more advanced solar cell architectures such as interdigitated back-contact (IBC) cells, or in even more advanced axial Si nanowire cells [17], the n^+ and p^+ doped Si surfaces are adjacent and preferably passivated simultaneously by one film or film stack. Hence, the aim of this work is to achieve the passivation of both $n+$ and $p+$ Si surfaces via a low-temperature and industrially viable deposition process. To this end, we propose the use of a passivation scheme without significant Q_f, while providing very low D_{it}. Rather than relying on a fixed charge density to reduce one of the two charge carrier types at the surface, such a "zero-charge" passivation approach could rely on high surface doping concentrations of the n^+ and p^+ Si regions, which reduces the minority carrier concentration at the surface and in this way supresses surface recombination.

In literature, there are several ways reported to manipulate the natively present fixed charge density of dielectrics and dielectric stacks, and some of them have been studied to address the simultaneous passivation of n^+ and p^+ Si surfaces [18–22]. For instance, to reduce the negative Q_f of Al_2O_3, the films can be deposited at high deposition temperatures (i.e., 300–400 °C com e.g., 200 °C), although this adversely affects the level of chemical passivation [23]. Secondly, the negative Q_f of thin Al_2O_3 films can be reduced when it is combined with SiN_x capping and a subsequent high-temperature firing step ≥ 800 °C [21,25]. Such Al_2O_3/SiN_x stacks show promising results on both n^+ and p^+ Si surfaces with various sheet resistances, irrespective of whether the Al_2O_3 films are deposited by PE-CVD [18], or ALD [21,22]. Nonetheless, these stacks exhibit a significant negative Q_f of $\sim(1–2)\times 10^{12}$ cm^{-2} [18,21,25], which is likely still affecting the passivation of n^+ Si surfaces. Alternatively, it is known already for a long time that an interfacial SiO_2 layer strongly reduces the fixed charge density of Al_2O_3 layers [26,27]. In fact, various interlayers can be used to reduce the Q_f of Al_2O_3 layers to virtually zero. This includes SiO_2 layers prepared by ALD [13,28] and by plasma-enhanced chemical vapor deposition (PE-CVD) [20], thermally-grown SiO_2 [29], but also other materials such as HfO_2 [30]. Moreover, when SiO_2 is combined with other capping layers, such as in SiO_2/SiN_x or $SiO_2/Al_2O_3/SiN_x$ stacks, very low charge densities can be obtained [7,29,31], although this strongly depends on the SiO_2 thickness and process conditions [7,29]. To achieve passivation of n^+ and p^+ Si surfaces, for example wet-chemical or PE-CVD SiO_2 films in combination with PE-CVD SiN_x capping layers [19,22], and PE-CVD SiO_2/Al_2O_3 stacks with varying SiO_2 film thicknesses have been studied [20].

SiO_2/Al_2O_3 stacks which are fully prepared by ALD are very promising candidates as "zero-charge" passivation schemes for n^+ and p^+ Si surfaces. It has been reported by Dingemans et al. [13] and Terlinden et al. [28] that those stacks exhibit excellent chemical passivation levels (e.g., D_{it} values $<10^{11}$ eV^{-1} cm^{-2} at mid gap [32]), while their Q_f can effectively be tuned from strongly negative up to zero or even positive values by carefully tuning the ALD SiO_2 thickness. It is hypothesized that for these stacks the SiO_2 interlayer acts as a (trap-assisted) electron tunnel barrier, preventing charge-injection from the c-Si base into the electron trap sites in the Al_2O_3 [13,28]. The Al_2O_3 plays a key role in the reduction of Si/SiO_2 interface defect states, which are effectively passivated by the diffusion of hydrogen from the Al_2O_3 film to the Si/SiO_2 interface during annealing [33]. ALD SiO_2/Al_2O_3 stacks have the advantage that they can be prepared at relatively low temperatures (e.g., 200 °C), are conformal even over high-aspect ratio structures and can be prepared in a single deposition run [13,28,34]. The ALD stacks can potentially be used in high-volume manufacturing for the simultaneous passivation of both sides of IBC cells or bifacial cells. Moreover, very promising results containing the passivation of both sides of PERC cells have recently been reported for stacks having 5 nm ALD SiO_2 films [35].

In previous work dealing with the passivation of n^+ and p^+ Si surfaces, the influence of the fixed charge density on the passivation was not fully addressed yet. In this work, first the influence of the charge density on the passivation of these surfaces is investigated via corona charging experiments. The results serve as a benchmark for the next part of the paper, where we will carefully tune the SiO_2 interlayer thickness to control of the fixed charge density of ALD SiO_2/Al_2O_3 stacks and will study its applicability as passivation scheme of both n^+ and p^+ Si surfaces. Next, the role of surface doping concentration on the passivation properties will be discussed. Finally, we will study the compatibility of ALD SiO_2/Al_2O_3 films as "zero-charge" passivation scheme with typical Si solar cell processing, such as the compatibility with SiN_x capping layers and high-temperature processing steps.

2. Experimental

Symmetrical $n^+/p/n^+$ lifetime samples were made by $POCl_3$ diffusions in an industry-scale tube furnace process (Tempress systems TS81003) on double-side chemically polished, p-type Czochralski-grown (Cz) Si (100) wafers. In another tube of the furnace, symmetrical $p^+/n/p^+$ lifetime samples were fabricated using BBr_3 as precursor, on Cz n-type wafers with a random-pyramid textured surface. After diffusion, the surface doping concentration of the $p^+/n/p^+$ sample was increased by a short wet etch [36]. The resulting (active) doping profiles were determined from electrochemical capacitance–voltage (ECV) profiling (see Fig. 1) using a WEP wafer profiler CVP21 table-top unit. Note, that the doping profile of the boron-doped samples can be affected by the texture of the surface, and a heavier doping level can be expected at the pyramid tips [37]. The sheet resistance and the homogeneity of the doped regions were measured using a Signatone four-point probe, in combination with a Keithley 2400 Source Measurement Unit.

After glass removal, the samples received a short dip in diluted HF (1%, 1 min). Subsequently, SiO_2/Al_2O_3 stacks and Al_2O_3 films were deposited on both sides of the samples by plasma-enhanced ALD in an Oxford Instruments OpAL^{TM} reactor at 200 °C in a *single* deposition run. $H_2Si(N(C_2H_5)_2)_2$ and $Al(CH_3)_3$ were used as metal-organic precursors and O_2 plasma as oxidant. More details on the ALD processes can be found elsewhere [34]. A schematic display of the passivated lifetime samples is given in Fig. 2. To activate the passivation, the stacks were either annealed for 10 min at 400 °C in N_2, or capped with 70 nm SiN_x followed by an industrial firing step ($\sim$800 °C for 1 s). The SiN_x was prepared by in-line PE-CVD reactor (MAiA, Roth and Rau), equipped with a microwave plasma source. As deposited, the SiN_x has a refractive index of $n=2.05$ at 633 nm.

A lifetime tester (Sinton WCT 100) was used for quasi-steady state photoconductance decay (QSS-PC) and transient-PC measurements. Using QSS-PC the recombination parameter J_0 was derived at high injection levels of Δn $\sim(0.4–6.2)\times 10^{16}$ cm^{-3} using the method of Kane and Swanson [38], which allows the extraction of J_0 from lifetime measurements via

$$\frac{1}{\tau_{eff}} - \frac{1}{\tau_{Auger}} = \frac{1}{\tau_{SRH}} + J_0 \cdot \frac{(N_d + \Delta n)\Delta n}{q n_i^2 W} \quad (1)$$

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