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Passivation of c-Si surfaces by sub-nm amorphous silicon capped with silicon nitride

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A sub-nm hydrogenated amorphous silicon (a-Si:H) film capped with silicon nitride (SiN_x) is shown to provide a high level passivation to crystalline silicon (c-Si) surfaces. When passivated by a 0.8 nm a-Si:H/75 nm SiN_x stack, recombination current density J_0 values of 9, 11, 47, and 87 fA/cm^2 are obtained on 10 $\Omega\cdot\text{cm}$ n -type, 0.8 $\Omega\cdot\text{cm}$ p -type, 160 Ω/sq phosphorus-diffused, and 120 Ω/sq boron-diffused silicon surfaces, respectively. The J_0 on n -type 10 $\Omega\cdot\text{cm}$ wafers is further reduced to $2.5 \pm 0.5 \text{ fA/cm}^2$ when the a-Si:H film thickness exceeds 2.5 nm. The passivation by the sub-nm a-Si:H/ SiN_x stack is thermally stable at 400 °C in N_2 for 60 min on all four c-Si surfaces. Capacitance–voltage measurements reveal a reduction in interface defect density and film charge density with an increase in a-Si:H thickness. The nearly transparent sub-nm a-Si:H/ SiN_x stack is thus demonstrated to be a promising surface passivation and antireflection coating suitable for all types of surfaces encountered in high efficiency c-Si solar cells. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4937732>]

Intrinsic hydrogenated amorphous silicon (a-Si:H) has been demonstrated to provide a very high level passivation to crystalline silicon (c-Si) surfaces.^{1–5} Thanks to the extensive studies into the film bulk and interfacial properties in last decade, the recombination rate has been suppressed significantly, achieving recombination current density J_0 as low as 1 fA/cm^2 .⁵ The high-level of surface passivation by a-Si:H is attributable to a very low defect density at the a-Si:H/c-Si interface, assisted in part by the high concentration ($\sim 10\%$ – 20% atomic) of hydrogen incorporated into the films, which migrates to the interface and passivates remaining c-Si dangling bonds.^{6–8} One of the critical conditions for such a high-level passivation is known to be an atomically abrupt transition between the amorphous and crystalline phases at an a-Si:H/c-Si interface.²

The highly passivating property of a-Si:H has been exploited with remarkable success in silicon heterojunction (SHJ) solar cells, enabling (i) a record-high open circuit voltage of 750 mV in a front-junction SHJ cell,⁹ and (ii) energy conversion efficiencies above 25% in two interdigitated back contact (IBC) SHJ cells (one of which holds the current world record for single junction c-Si solar cells).^{10,11} For both types of SHJ solar cells, the employment of the a-Si:H film on the sunward side requires the film to be as thin as possible to minimize light absorption in it, but this usually causes a trade-off consideration on the surface passivation quality. On the front surface of solar cells, an antireflection coating (ARC) is also required; and in the cases of IBC-SHJ or conventional dopant-diffused devices, it can be non-conductive, for example, silicon nitride (SiN_x). The optimization of the a-Si:H/ SiN_x stack is therefore critical to simultaneously achieve good optical transmission and surface passivation for c-Si solar cells.

Several studies have been published on c-Si surface passivation by an a-Si:H/ SiN_x stack,^{5,12–14} as listed in Table I. It is reported that SiN_x is of great benefit in (i) improving the passivation quality by a-Si:H, and (ii) enhancing the thermal stability of a-Si:H passivation. However, the thickness of a-Si:H film required for an acceptable level of surface passivation, for example, $J_0 < 10 \text{ fA/cm}^2$ on undiffused c-Si surfaces, has so far been relatively high (i.e., 5 nm as shown in Table I), leading to an $\sim 1 \text{ mA/cm}^2$ loss in photo-generation current (as simulated using the optical parameters from Ref. 15 and the simulation software OPAL 2¹⁶). A lower limit on the thickness of the a-Si:H film required for good passivation is set by the penetration depth of energetic ions (in the form of either H-related and/or N-related ions^{17–19}) into the a-Si:H thin film during the deposition of plasma enhanced chemical vapour deposition (PECVD) over-layers, causing degradation in the a-Si:H/c-Si interface quality.

The main motivation of this work is to develop a highly transparent a-Si:H/ SiN_x stack to provide a comparable passivation level to that reported in the literature for thicker a-Si:H films (i.e., $\geq 5 \text{ nm}$). We first evaluate the effect of a-Si:H film thickness on the passivation quality of n -type undiffused c-Si surface, finding that a sub-nm thick a-Si:H capped with SiN_x is sufficient to ensure a high surface passivation quality, with $J_0 < 10 \text{ fA/cm}^2$. The sub-nm a-Si:H/ SiN_x stack is then demonstrated to be capable of providing thermally stable passivation to four types of c-Si surfaces, namely, p - and n -type undiffused, boron-diffused p^+ , and phosphorus-diffused n^+ , establishing the a-Si:H/ SiN_x stack as a highly passivating scheme for most high efficiency silicon solar cells, with either front junction or rear junction and homo-junction or heterojunction architectures. Finally, capacitance–voltage (C – V) measurements are employed to probe the effect of a-Si:H thickness on the electronic properties of a-Si:H/ SiN_x stack, elucidating the physical mechanisms underlying the measured dependence of the surface passivation.

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TABLE I. Summary of state-of-the-art passivation results by schemes featuring an a-Si:H thin film.

Passivating scheme	a-Si:H thickness (nm)	Substrate	J_0 (fA/cm ²)	Reference
a-Si:H	3	FZ <i>p</i> -type 1.5 Ω-cm	25.8 ^a	1
	5	FZ <i>p</i> -type 1.5 Ω-cm	13.4 ^a	
a-Si:H	50	FZ <i>n</i> -type 4.0 Ω-cm	5.8 ^a	20
a-Si:H/a-Si(<i>n</i>)	5	FZ <i>n</i> -type 2.5–3 Ω-cm	2.0 ^a	4
a-Si:H/SiO _x	70	FZ <i>p</i> -type 1.0 Ω-cm	3.7 ^a	21
a-Si:H/SiO _x	3.4	FZ <i>n</i> -type 4.0 Ω-cm	5.8 ^a	22
a-Si:H/SiO _x /SiN _x	10	FZ <i>n</i> -type 1.7 Ω-cm	1.0 ^b	5
	5	CZ <i>n</i> -type 3.6 Ω-cm	1.6 ^b	
a-Si:H/SiN _x	10	FZ <i>n</i> -type 2.5 Ω-cm	3.7 ^a	12
	10	FZ <i>p</i> -type 2.0 Ω-cm	10.8 ^a	
	2	FZ <i>n</i> -type 2.5 Ω-cm	27.5 ^a	
	2	FZ <i>p</i> -type 2.0 Ω-cm	79.6 ^a	
a-Si:H/SiN _x	10	FZ <i>p</i> -type 1.5 Ω-cm	3.5 ^a	13
		FZ <i>n</i> ⁺ 90 Ω/sq	10.0 ^b	
a-Si:H/SiN _x	10	FZ <i>p</i> ⁺ 90 Ω/sq	20.0 ^b	14

^a J_0 is extracted from the reported effective minority carrier lifetime using $J_0 = \frac{qWn_i^2(\Delta n + N_d)}{2} \left(\frac{1}{\tau_{\text{eff}}} - \frac{1}{\tau_{\text{bulk}}} \right)$, where q is the elementary charge, W is the thickness of the silicon substrate, n_i is the intrinsic carrier concentration, N_d is the bulk dopant concentration of the silicon substrate, and τ_{bulk} is the intrinsic bulk lifetime of c-Si parameterised by Richter *et al.*²³

^b J_0 is the reported value in the literature, extracted by the Kane and Swanson method.²⁴

The a-Si:H and SiN_x films were synthesised using a direct PECVD reactor (Oxford PlasmaLab 100) at a frequency of 13.56 MHz and a deposition pressure of 650 mTorr. The deposition temperature for a-Si:H and SiN_x was 200 °C and 400 °C, respectively. The thickness of SiN_x with a refractive index of 2.0 at a wavelength of 632 nm was fixed at 75 nm for all samples, while the thickness of a-Si:H was varied from 0 up to 9.3 nm by varying the deposition time. The deposition rate of a-Si:H was approximately 2 Å/s. The film thickness and refractive index of a-Si:H were determined by fitting polarized reflectance using the Tauc–Lorentz model,²⁵ as measured by *ex-situ* spectroscopic ellipsometry (J.A. Woolam M2000 ellipsometer) on ~1 mm thick glasses. The deposition power and silane gas flow for a-Si:H were optimised to (i) ensure the plasma is ignitable, and (ii) minimize the deposition rate to increase the a-Si:H film thickness control in the sub-nm region. It should be noted that given the inherent difficulties with uniformly depositing and measuring such thin films, we estimated an uncertainty of ±1 Å, which is significant given the range of thickness.

To evaluate recombination at the interface between c-Si and the deposited films, symmetrically coated samples were fabricated on four types of c-Si substrates: (i) undiffused 10 Ω-cm *n*-type wafers (195 μm), (ii) undiffused 0.8 Ω-cm *p*-type wafers (275 μm), (iii) phosphorus diffused (*n*⁺) 100 Ω-cm *p*-type wafers (300 μm), and (iv) boron diffused (*p*⁺) 100 Ω-cm *n*-type wafers (520 μm). All wafers were float-zone (FZ) grown and {100} oriented. The undiffused wafers were etched in tetramethylammonium hydroxide (TMAH, 25 wt. %) at ~85 °C to remove saw damage. Phosphorus and boron diffusions were performed in quartz tube furnaces using liquid POCl₃ and BBr₃ sources, resulting in sheet resistances of 160 and 120 Ω/sq, respectively, as measured with a four-point probe. All samples were then cleaned by the RCA (Radio Corporation of America) procedure and dipped in a 1% diluted hydrofluoric acid solution to remove any remaining oxide prior to PECVD film depositions.

The effective carrier lifetime τ_{eff} as a function of excess carrier density Δn was measured using a Sinton Instruments WCT-120 photoconductance tool.²⁶ The recombination current density J_0 was then extracted from the effective lifetime measurement (taken in transient mode) by employing the technique developed by Kane and Swanson²⁴ with an intrinsic carrier concentration $n_i = 8.6 \times 10^9 \text{ cm}^{-3}$ at 25 °C. The Kane and Swanson technique was originally developed to characterise recombination on heavily diffused silicon wafers (such as *n*⁺ and *p*⁺) and recently has been extended to undiffused wafers, provided that the passivating scheme contains a significant amount of fixed charges (absolute amount $> 1.0 \times 10^{11} \text{ cm}^{-2}$) to strongly accumulate or invert the c-Si surface.²⁷ As shown later, the a-Si/SiN_x stack features a high density of positive charge on the order of 10^{12} cm^{-2} , and hence, the use of J_0 to characterize surface recombination is valid.

Figure 1 presents the effect of a-Si:H thickness on surface passivation of *n*-type undiffused wafers. As can be seen, the surface recombination of the sample coated only with SiN_x is high, with a J_0 of 427 fA/cm², despite a likely existence of oxygen atoms at c-Si surface due to unavoidable exposure to air or moisture before film deposition. By contrast, when a thin a-Si:H film is inserted between SiN_x and c-Si, the surface passivation quality is markedly enhanced and it exhibits a strong dependence on a-Si:H thickness, as also evidenced from the injection-dependent lifetime curves plotted in the inset. As the a-Si:H interlayer thickness increases, the J_0 first decreases and then tends to saturate at a value of 2–3 fA/cm² when the a-Si:H thickness exceeds 2.5 nm. The trend is similar to the one presented by Koyoma *et al.*,¹² but the passivation by a-Si:H/SiN_x in this work saturates at a thinner a-Si:H (i.e., 2.5 nm comparing to ~10 nm presented in Ref. 12). Remarkably, the surface recombination current is already considerably low, $J_0 < 10 \text{ fA/cm}^2$, with a sub-nm (0.8 nm) a-Si:H film. An optical simulation by OPAL 2 shows that the 0.8 nm a-Si:H/75 nm SiN_x stack is nearly

FIG. 1. The recombination current density J_0 as a function of the a-Si:H thickness on undiffused FZ 10 Ω -cm n -type Si wafers when passivated by an a-Si:H/SiN_x stack. The inset shows the injection-dependent minority carrier lifetime $\tau_{\text{eff}}(\Delta n)$ curves for the a-Si:H/SiN_x stack with three different a-Si:H film thickness: 0, 0.8, and 2.5 nm.

transparent, with a low light absorption loss of 0.17 mA/cm² when used as a front antireflection coating. Compared to the ~ 5 nm a-Si:H films commonly reported in the literature to obtain a similar level of passivation, the sub-nm a-Si:H provides an improvement in the photo-generation current density of ~ 0.83 mA/cm²—a significant gain given the advanced state of c-Si solar cell development.

Besides the good passivation of n -type undiffused c-Si surfaces, the a-Si:H/SiN_x stack is also found to be capable of passivating the other three commonly used c-Si surfaces, namely, undiffused p -Si, diffused n^+ -Si, and p^+ -Si. Figure 2 shows the Auger-corrected inverse τ_{eff} as a function of excess carrier density of the four samples when passivated

FIG. 2. Auger-corrected inverse $\tau_{\text{eff}}(\Delta n)$ of (i) undiffused 10 Ω -cm n -type wafers, (ii) undiffused 0.8 Ω -cm p -type wafers, (iii) phosphorus diffused n^+ , and (iv) boron diffused p^+ c-Si wafers when passivated by an 0.8 nm a-Si:H/SiN_x stack. The inset shows the thermal response of the four samples annealed in N₂ at 400 $^{\circ}$ C over a period of 1 h.

by the 0.8 nm a-Si:H capped with SiN_x. It can be seen that the data follows a linear trend in the (high injection) range of excess carrier densities used to extract J_0 , and therefore, a reliable value of J_0 can be determined. The a-Si:H/SiN_x stack provides a very similar level of passivation to both n - and p -type undiffused wafers, with a J_0 of 11 and 9 fA/cm², respectively, corresponding to implied open-circuit voltages of 722 and 712 mV. On the phosphorus-diffused n^+ surface, the passivation quality by sub-nm a-Si:H/SiN_x is comparable to that given by a passivating PECVD SiN_x, with a J_0 of 47 fA/cm². This value of J_0 is approximately five times higher than the one presented by Gatz *et al.*¹³ (see details in Table I), mainly attributable to an approximately five times higher surface dopant concentration of the n^+ diffusion in this work (5×10^{19} cm⁻³, measured by electrochemical C-V dopant profiling) versus 1×10^{19} cm⁻³ in Ref. 13. It should be noted that these values of J_0 include recombination in the bulk of the highly doped region. On the other hand, the passivation of the p^+ boron-diffused surface by the a-Si:H/SiN_x stack is inferior, with a J_0 of 87 fA/cm². Nevertheless, compared to the J_0 achievable on such p^+ surface by using SiN_x only (>500 fA/cm²), the sub-nm a-Si:H results in a reduction of J_0 by a factor of ~ 6 . The superior passivation of n^+ surfaces over that of p^+ surfaces could be due to the presence of a significant positive fixed charge density in the a-Si:H/SiN_x stack, and this indeed will be confirmed later.

While the sub-nm a-Si:H/SiN_x stack has been demonstrated to provide good passivation of c-Si surfaces, the thermal stability for process integration in silicon solar cell fabrication remains an important consideration. The above-mentioned four types of passivated samples were subjected to a temperature of 400 $^{\circ}$ C in a N₂ ambient for various lengths of time. The inset in Figure 2 shows the J_0 as a function of annealing time, indicating that the surface passivation by the stack is essentially stable on the four types of c-Si surfaces. Notably, a short annealing of 20 min even improves the passivation of undiffused n - and p -type silicon samples. The a-Si:H/SiN_x stack therefore has the potential to be compatible with the sintering of vacuum evaporated metals or low temperature screen printed pastes for the fabrication of silicon solar cells. In addition, a preliminary experiment of light-soaking the samples at ~ 40 $^{\circ}$ C under the one-sun spectrum for up to 160 h shows that (i) the passivation quality by a 2.5 nm a-Si/SiN_x stack is stable for the duration monitored, and (ii) the passivation quality by a 0.8 nm a-Si/SiN_x stack is slightly improved after the first 60 min exposure and then tends to plateau.

C-V measurements were performed to elucidate the physical mechanisms underlying the evolution in surface passivation with an increase in a-Si:H thickness. Double-sided polished n -type FZ {100} c-Si with a resistivity of 1.0 Ω -cm and a thickness of 290 μ m were coated on one side with the passivating film stack. Three thickness of a-Si:H were used for this experiment: 0, 0.8, and 2.5 nm. The metal-insulator-semiconductor capacitors were fabricated by evaporating aluminium through a shadow mask onto the films, creating circular dots of diameter ~ 700 μ m and thickness ~ 100 nm. The rear contact was formed with a GaIn eutectic paste. High frequency (HF) and quasistatic (QS) C-V measurements were taken using an HP 4284 A

Precision LCR Meter and HP 4140B Picoammeter/DC Voltage Source. In order to minimize charge injection upon voltage biasing, the voltage was swept from 0 or +1 V until the applied voltage caused deep depletion or inversion. The interface defect density at midgap D_{it} was determined by applying the Castagne method²⁸ to HF and QS C–V measurements. The effective insulator charge density Q_{eff} was calculated by assuming that all charge was located at the a-Si:H/c-Si interface.

Figure 3 gives the interface defect density and the effective charge density as a function of a-Si:H thickness, while the inset shows the actual HF and QS C–V measurements. We note the anomalous shape of QS curve for the 2.5 nm a-Si:H/SiN_x stack, causing some uncertainty in extracting D_{it} . This behaviour is attributable to the leakage current in the stack system, as also observed from C–V measurement of a thick a-Si:H film (40 nm) at room temperature.²⁹ Nevertheless, the conclusions derived from this study rely on trends, rather than absolute values of D_{it} . As we can see, without a-Si:H interlayer, the D_{it} is significantly higher, of the order of $10^{13} \text{ eV}^{-1} \text{ cm}^{-2}$. The insertion of a 0.8 nm a-Si:H interlayer results in one order of magnitude reduction in D_{it} . The interface defects are further reduced by increasing the a-Si:H film to 2.5 nm, yielding another order of magnitude decrease in D_{it} to a value of $1.8 \times 10^{11} \text{ eV}^{-1} \text{ cm}^{-2}$. These results suggest that the insertion of a thin a-Si:H film can (i) prevent c-Si surface from ion damage during SiN_x over-layer deposition and (ii) terminate the dangling bonds at c-Si surface.

The trend observed for D_{it} correlates directly with that for J_0 , but the passivation quality is also affected by the charge density in the films. As shown in Figure 3, the charge density exhibits a similar trend to that of the interface defect density, decreasing as the a-Si:H becomes thicker. Without an a-Si:H film, the SiN_x itself has a strong positive charge of $2.2 \times 10^{12} \text{ cm}^{-2}$, in line with values found in the literature.³⁰ After insertion of the 0.8 nm a-Si:H interlayer, the stack charge

density was reduced to $1.2 \times 10^{12} \text{ cm}^{-2}$. A further increase of a-Si:H to 2.5 nm results in a slight reduction in Q_{eff} to $1 \times 10^{12} \text{ cm}^{-2}$. The decreasing trend for Q_{eff} of the a-Si:H/SiN_x stack might be related to the a-Si:H interlayer acting as a barrier for the thermal-assisted charge injection from c-Si to SiN_x layer, similar to the behaviour exhibited by SiO_x/SiN_x or AlO_x/SiN_x stacks.^{31–34} The substantial improvement in surface passivation by a-Si:H/SiN_x is therefore primarily attributable to the reduction in the defect density, outweighing the loss of the charge-assisted passivation due to the decreased positive charge upon inserting a-Si:H thin film.

In conclusion, a thermally stable, highly passivating scheme for c-Si surfaces has been achieved by a sub-nm a-Si:H film capped with PECVD SiN_x. A recombination current density J_0 of 9 fA/cm² was obtained on undiffused *n*-type silicon using 0.8 nm a-Si:H, and J_0 was further reduced to 2–3 fA/cm² when the a-Si:H film thickness exceeded 2.5 nm. The surface passivation by the sub-nm a-Si:H/SiN_x stack was found to also be good on the other three types of c-Si surfaces (namely, undiffused *p*-, diffused *n*⁺-, and *p*⁺-Si) commonly implemented in c-Si solar cells. The passivation was shown to be thermally stable at 400 °C in a N₂-ambient for 60 min, demonstrating the compatibility with the sintering of contacts for fabricating high efficiency c-Si solar cells. Finally, C–V measurements revealed that an increase in the a-Si:H thickness induced a reduction in the density of interface defects and film charges, corroborating that the enhancement in passivation by inserting a-Si:H into the SiN_x/c-Si interface is primarily attributable to the reduction in the defect density. The results in this work are especially significant to c-Si photovoltaics, given that the nearly transparent a-Si:H/SiN_x stack (with sub-nm thick a-Si:H film) provides a high level and thermally stable passivation to four types of c-Si surfaces.

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FIG. 3. Extracted interface defect density D_{it} at midgap, and effective insulator charge density Q_{eff} from extracted the C–V measurements (see the inset) on a-Si:H/SiN_x coated samples, where the thickness of a-Si:H varies at 0, 0.8, and 2.5 nm.

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