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Influence of base pressure and atmospheric contaminants on *a*-Si:H solar cell properties

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The influence of atmospheric contaminants oxygen and nitrogen on the performance of thin-film hydrogenated amorphous silicon (*a*-Si:H) solar cells grown by plasma-enhanced chemical vapor deposition at 13.56 MHz was systematically investigated. The question is addressed as to what degree of high base pressures (up to 10^{-4} Torr) are compatible with the preparation of good quality amorphous silicon based solar cells. The data show that for the intrinsic *a*-Si:H absorber layer exists critical oxygen and nitrogen contamination levels (about 2×10^{19} atoms/cm^3 and 4×10^{18} atoms/cm^3, respectively). These levels define the minimum impurity concentration that causes a deterioration in solar cell performance. This critical concentration is found to depend little on the applied deposition regime. By enhancing, for example, the flow of process gases, a higher base pressure (and leak rate) can be tolerated before reaching the critical contamination level. The electrical properties of the corresponding films show that increasing oxygen and nitrogen contamination results in an increase in dark conductivity and photoconductivity, while activation energy and photosensitivity are decreased. These effects are attributed to nitrogen and oxygen induced donor states, which cause a shift of the Fermi level toward the conduction band and presumably deteriorate the built-in electric field in the solar cells. Higher doping efficiencies are observed for nitrogen compared to oxygen. Alloying effects (formation of SiO$_x$) are observed for oxygen contaminations above 10^{20} atoms/cm^3, leading to an increase in the band gap. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3009384]

I. INTRODUCTION

In large-area industrial deposition systems, the incorporation of residual gases is usually higher than in small-area research deposition systems due to the higher base pressure.[1] Here, we focus on the question as to what degree of high-quality *a*-Si:H solar cells can be produced under such a high base pressure. In general, in *a*-Si:H *p*-*i*-*n* solar cells the device quality is known to depend strongly on the properties of the intrinsic absorber layer (*i*-layer). Its performance deteriorates severely by impurity incorporation, for example, oxygen or nitrogen at high concentration.[1] Previous studies investigated predominantly the effects of contaminations on the optoelectronic properties of single films.[2,3] In their studies, Morimoto *et al.*[2] and Shimizu *et al.*[4] developed a model describing the incorporation of oxygen and nitrogen into the *i*-layer based on conductivity and electron spin resonance (ESR) measurements. These impurities are assumed to lead to donorlike states, shifting the Fermi level toward the conduction band. By means of ESR studies, Stutzmann *et al.*[5] observed that the defect density in *a*-Si:H films is increased when exceeding an impurity concentration of approximately 10^{20} atoms/cm^3 for oxygen and nitrogen. Below that level no influence on the ESR signal was seen. Post-transit photocurrent spectroscopy of *a*-Si:H films showed that oxygen donors have to be located at energy levels lower than 0.35 eV below the conduction band edge.[6] The relationship between light-induced degradation and contamination of *a*-Si:H solar cells was also investigated. The authors attributed a disturbance of the built-in electric field in solar cells to the incorporated donorlike impurities.[3] Kroll *et al.*[7] showed that for a constant impurity concentration in the gas phase, the incorporation of oxygen or nitrogen depends on the applied power and the resulting deposition rate. We distinguish between two main sources of contamination, namely, air leakages or outgassing (virtual leakage) of the process chamber and contaminated process gases. We simulate these sources of contamination by intentionally contaminating the solar cell *i*-layer with oxygen or nitrogen.

This paper addresses the preparation conditions at relatively high base pressures for the *i*-layer deposition (up to 10^{-4} Torr). Two deposition regimes were examined, low total process gas flow and deposition pressure, and high total process gas flow and deposition pressure. The contaminating gases, oxygen and nitrogen, were provided to the deposition chamber through a needle valve at a chamber wall, simulating chamber leaks and the outgassing process. The *I*-*V* characteristics for various nitrogen or oxygen concentrations in the *i*-layer were compared for both applied deposition conditions. Special attention was paid to the fill factor (FF) under long wavelength illumination because of its sensitivity to

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the impurity concentration. Finally, the solar cell *I-V* parameters were compared to single *i*-layer electrical and optical properties.

II. EXPERIMENTAL DETAILS

All samples were prepared in a multichamber plasma-enhanced chemical vapor deposition (PECVD) system with capacitively coupled parallel plate electrodes. The excitation frequency was 13.56 MHz. As substrates for the *a*-Si:H *p-i-n* solar cells, commercial Asahi type U (SnO_2:F) transparent conductive oxide coated 10×10 cm^2 glasses were used. Single *a*-Si:H intrinsic films were deposited on Corning Eagle 2000 glass and *c*-Si substrates under the same conditions as those for the solar cell absorber layers. Seed layers (similar to the solar cell *p*-layer but undoped) were applied to grow these films as similarly as possible to the *i*-layer embedded in the *p-i-n* solar cell. In this study, two deposition regimes were applied for fabrication of *a*-Si:H *i*-layers. One was a low flow [SiH_4|H_2 of 12|100 SCCM (SCCM denotes cubic centimeter per minute at STP)] and low pressure (3 Torr) regime [low flow, low pressure (LFLP)] at a heater temperature (T_H) of ~260 °C (standard regime). In earlier studies this regime was used to produce state-of-the-art *a*-Si:H solar cells and was therefore taken as a reference.[1] The second regime was a high flow (SiH_4|H_2 of 7.8|360 SCCM) and high pressure (10 Torr) regime [high flow, high pressure (HFHP)] at T_H of ~200 °C. The applied discharge powers were 0.014 and 0.2 W/cm^2, yielding deposition rates of ~0.14 and ~0.45 nm/s, respectively. This choice of deposition parameters was made with the intention to lower the oxygen concentration in the plasma and thus in the growing film, due to the higher total gas flow. Additionally, the power was increased to enhance the deposition rate, as a previous study reported this was beneficial for the reduction in oxygen contamination in *a*-Si:H.[7] The other parameters were adjusted to produce state-of-the-art solar cells. The *i*-layers had a thickness of approximately 350 nm. Thermally evaporated silver (Ag) contact pads were used as a back reflector and defined the solar cell area of 1 cm^2.

The contamination gases, oxygen and nitrogen (purity of 5.0), were induced through a controllable leak at the reactor wall to simulate chamber leakages and poor vacuum conditions. By varying the contamination flow, base pressure values between 10^{-8}–10^{-3} Torr were achieved. Note that the process gases SiH_4 and H_2 used for the *i*-layer deposition were lead through a gas purifier (Entegris/Aeronex CE-35KF-SK-4R) prior to feeding them into the reactor in order to avoid unintended contamination.

The *p-i-n* solar cells were characterized by *I-V* measurements under AM1.5 illumination (Wacom, class A solar simulator with 100 mW/cm^2) at 25 °C For the solar cell properties in the short and long wavelength range, additional filters were applied to study the collection of photocarriers generated close to the *p*/*i*-interface (blue light) and in the whole *i*-layer (red light). The oxygen and nitrogen contamination was measured by secondary ion mass spectrometry (SIMS) using a quadrupole instrument (Atomika 4000). The detection limits for oxygen and nitrogen were approximately 5×10^{17} cm^{-3} and 7×10^{16} cm^{-3}, respectively, for a sputtering rate >0.75 nm/s. Details of the setup used for SIMS measurements are published elsewhere.[8]

The electrical properties of single intrinsic films were determined by dark- and photoconductivity measurements in a coplanar two-point configuration with evaporated silver contacts, which were found to be Ohmic. The measurements were carried out in vacuum (~10^{-6} Torr) after 30 min of annealing at 160 °C. A tungsten lamp calibrated with respect to the AM1.5 solar simulator was used for the photoconductivity measurements. The intrinsic films were characterized by constant photocurrent method (CPM) and Fourier transform infrared (FTIR) spectroscopy (on *c*-Si) using a Nicolet 5700 spectrometer. In the latter experiment, the absorption of the Si–H wagging mode at 640 cm^{-1} and of the Si–H stretching modes at 2000 and 2100 cm^{-1} were analyzed to quantify the amount of silicon-bonded hydrogen in the *i*-layer (deposited on float zone *c*-Si substrates).[9] The microstructure parameter R^* serving as measure for the compactness of a material is defined by $R^*=I_{2000}/(I_{2000}+I_{2100})$ (Ref. 10) and I_{2100} are the integrated absorptions of the 2000 and 2100 cm^{-1} Si–H stretching modes, respectively. For highly oxygen-contaminated films ($>2.5\times10^{20}$ cm^{-3}), the silicon-bonded oxygen concentration can be evaluated from IR absorption data in a similar way to the concentration of Si-bonded hydrogen.[11]

III. RESULTS

A. Influence of the impurity concentration on the solar cell performance

Solar cells were prepared with different intentionally contaminated *i*-layers. In Fig. 1, the *I-V* characteristics (efficiency η, FF, and short circuit current density J_{SC} under AM1.5 unfiltered, red, and blue illumination and the open circuit voltage V_{OC}) are shown as a function of oxygen [Fig. 1(a)] and nitrogen [Fig. 1(b)] concentration. For oxygen, results for two deposition regimes (LFLP and HFHP) are shown, while for nitrogen, only data for the HFHP regime are presented. For both deposition regimes, the efficiency decreases by increasing the impurity concentration when a critical contamination level is exceeded. Below this level, the *I-V*-characteristics were not significantly affected. These critical levels can be determined from the solar cell efficiencies but even more sensitively by the FF versus *i*-layer impurity concentration. In general, the FF is known to be most sensitive to the incorporation of defects, donators, and acceptors, etc. in the *i*-layer.[1] The FF under red illumination shows the quality of carrier extraction from the bulk. It is influenced by the *i*-layer properties. Consequently, from the FF under red illumination, the oxygen and nitrogen critical contamination levels are seen more sensitively than from the cell efficiencies (see Fig. 1). A unique correlation between efficiency and FF with the oxygen concentration is observed in Fig. 1(a). This correlation is independent of applied deposition regimes. The solar cell deterioration starts with a slight red FF reduction above the critical level of ~2×10^{19} oxygen atoms/cm^3 and becomes stronger with

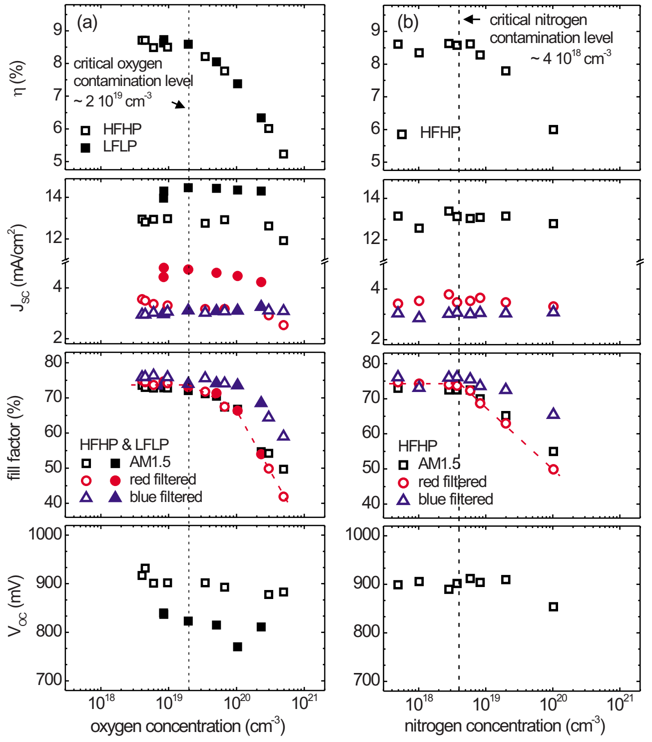

FIG. 1. (Color online) Solar cell efficiency η, FF, J_{SC}, and V_{OC} of *p-i-n* *a*-Si:H solar cells vs impurity concentration in the *i*-layer varied by oxygen (a) and nitrogen (b) flows into the deposition chamber. For oxygen, two deposition regimes are applied, LFLP and HFHP.

increasing oxygen content. Above an oxygen concentration of about 10^{20} atoms/cm^3, the solar cell quality decreases drastically. For nitrogen, the red FF reduction starts for a concentration of approximately 4×10^{18} atoms/cm^3 [see Fig. 1(b)]. The critical level for nitrogen is approximately one order of magnitude lower than the oxygen critical level. In related studies of the atmospheric contamination of hydrogenated microcrystalline silicon (μc-Si:H) deposited under conditions similar to the HFHP regime, similar critical levels were found, namely $(1-2)\times10^{19}$ atoms/cm^3 for oxygen and $(6-8)\times10^{18}$ atoms/cm^3 for nitrogen.[12]

Under blue illumination, the FFs representing the *p/i*-interface quality tend to remain unaffected or even increase for increasing impurity concentration. Only at very high concentrations does the FF under blue illumination decrease. Compared to nitrogen, the losses in the blue FF are more dominant for oxygen. The short circuit current density and the open circuit voltage as a function of the oxygen contamination reveal differences for the two deposition regimes but not for the contamination species. In the LFLP regime, a high J_{SC} ($\sim$14.3 mA/cm^2) and a low V_{OC} ($\sim$825 mV) are obtained whereas the HFHP is associated with a lower J_{SC} ($\sim$13 mA/cm^2) and a higher V_{OC} ($\sim$900 mV).

Apart from the highest oxygen concentrations, the V_{OC} and J_{SC} values measured under AM1.5 and blue illumination exhibit no significant changes due to contamination. However, under red illumination for both deposition regimes, J_{SC} decreases slightly for increasing oxygen concentration [see Fig. 1(a)]. Nitrogen incorporation does not influence J_{SC} and V_{OC}, except for the highest concentration in the examined range as it is shown in Fig. 1(b). In contrast, for μc-Si:H the short circuit current density is strongly affected by the impurities.[12]

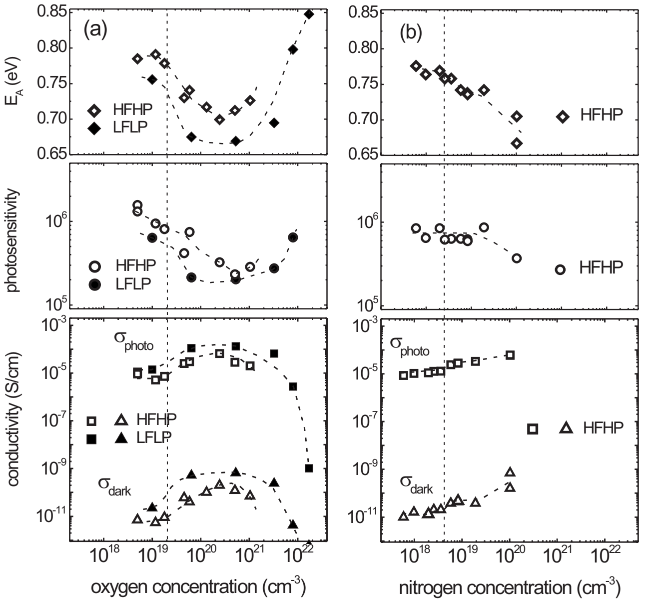

FIG. 2. Activation energy E_A, photosensitivity ($\sigma_{\mathrm{photo}}/\sigma_{\mathrm{dark}}$), dark-($\sigma_{\mathrm{dark}}$) and photoconductivity (σ_{photo}) of intrinsic films deposited at HFHP (open symbols) and LFLP (filled symbols) as a function of oxygen (a) and nitrogen (b) concentrations. The dashed lines are guides for the eyes.

B. Influence of the impurity concentration on the electrical and optical properties of the intrinsic absorber layer

The dependence of activation energy, dark conductivity and photoconductivity, as well as the photosensitivity on oxygen concentration for the two deposition regimes (HFHP and LFLP) is shown in Fig. 2(a). The results for both regimes are quite similar. Dark- and photoconductivities increase up to an oxygen concentration of about 10^{20} atoms/cm^3 and drop at higher oxygen concentrations. The inverse characteristic is seen for the photosensitivity and the activation energy E_A, determined by $\sigma_{\mathrm{dark}}=\sigma_0\exp[-E_A/(k_BT)]$ from the dark-conductivity data. Here, σ_0 is conductivity prefactor and k_B is the Boltzmann constant.[13] Quantitatively, it is seen that for the photo and dark-conductivity, the values obtained from the LFLP deposited films are by approximately half an order of magnitude higher compared to the HFHP films. Activation energy and photosensitivity are somewhat lower. Differences were also observed by FTIR measurements for the two deposition regimes. It was found that for the LFLP regime the (bound) hydrogen concentration was 12.5% and for the HFHP regime it was 16%, both independent of the impurity concentration. Differences were also obtained for the microstructure parameter. For the LFLP regime the material tends to be more dense ($R^*\approx0.03$) than for the HFHP regime, where $R^*\approx0.13$ is observed. This somewhat more porous film structure along with the increased H concentration may be caused by the three to four times higher deposition rate, or by the lower temperature used in the HFHP regime.

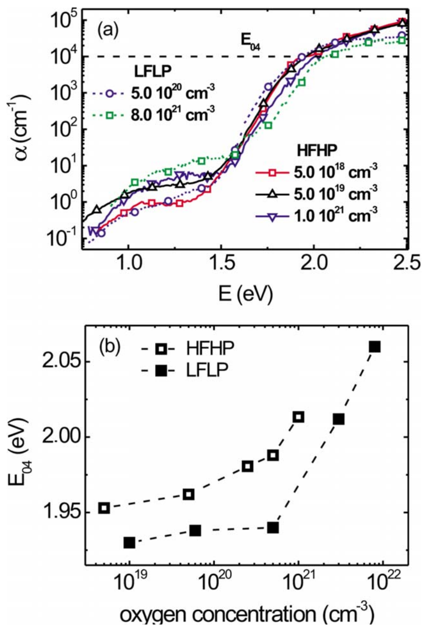

FIG. 3. (Color online) Absorption spectra of oxygen-contaminated films deposited in the HFHP and LFLP regimes (a). The E_{04}-gap of *a*-Si:H films deposited at HFHP (open symbols) and LFLP (filled symbols) as a function of the oxygen concentration (b). The dashed lines are guides for the eyes.

The electrical properties for intentionally contaminated nitrogen films are shown in Fig. 2(b). With rising nitrogen concentration, both dark- and photoconductivities start to increase. The corresponding inverse effect is seen in activation energy and photosensitivity. In this series of samples, the nitrogen concentration did not exceed 10^{20} atoms/cm^3, therefore the high impurity concentration effects observed for oxygen incorporation could not be studied.

Figure 3(a) shows the absorption spectra measured by constant photocurrent measurement (CPM) for selected *i*-layers deposited in the LFLP and HFHP regimes for different oxygen concentrations. In both deposition regimes, the subgap absorption increases with rising oxygen concentration. However, the HFHP samples show this increase at lower oxygen concentrations than the LFLP samples. For example, at oxygen concentrations of about 5×10^{20} atoms/cm^3 (LFLP) and 5×10^{18} atoms/cm^3 (HFHP) both films show similar subgap absorption. An increase in subgap absorption is also observed for increasing nitrogen contamination (data not shown).

Figure 3(b) shows the E_{04}-gap values (determined by the energy at which the absorption coefficient α is 10^4 cm^{-1}) as a function of the oxygen concentration. At low oxygen concentrations, the E_{04} -gap of *i*-layers deposited in the HFHP regime (1.96 eV) is approximately 0.03 eV larger than for LFLP films. This is likely due to the higher hydrogen content. With increasing oxygen concentration the E_{04} increases for both deposition regimes particularly for oxygen concentrations $>4\times10^{20}$ oxygen atoms/cm^3 (i.e., >1 at. %). This is in good agreement with the decreasing conductivity in the alloying range. No significant variation in the E_{04}-gap was observed for the investigated nitrogen concentration range.

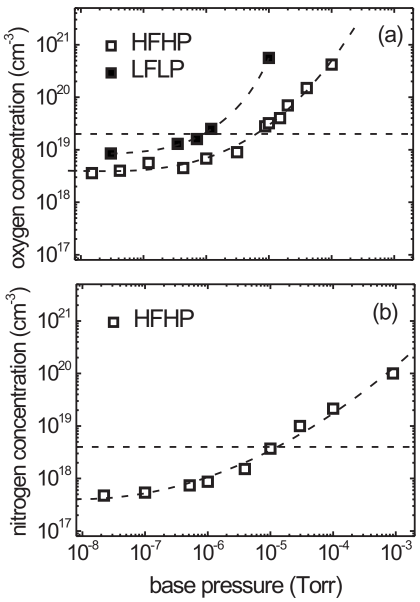

FIG. 4. Oxygen (a) and nitrogen (b) concentrations of *a*-Si:H multilayers deposited at different base pressures varied by a controllable chamber leak. The films were fabricated using two deposition regimes: HFHP (open squares) and LFLP (filled squares). The dashed lines are guides for the eyes.

C. Correlation of base pressure and impurity concentration

Since we see a correlation between the impurity concentration and solar cell characteristics irrespective of the applied deposition regime, deposition conditions leading to low impurity contents at a given base pressure and the dependence of impurity concentration on base pressure are of interest. Figure 4 shows the contamination concentration of the *a*-Si:H absorber layers as a function of the base pressure for the HFHP and LFLP regime for oxygen and for the HFHP regime for nitrogen. In order to keep the deposition conditions as similar as possible, this study was performed on sandwich layers, i.e., a multilayer system composed of *i*-layers of three to five different contamination flows. The base pressure in the *i*-chamber for the *i*-layer deposition was varied from 10^{-8} to 10^{-3} Torr, and the resulting contamination concentration of the *i*-layer corresponds directly to the increasing base pressure. The general trend is independent of the applied deposition regime and contamination gas. For the case of oxygen [Fig. 4(a)], the *i*-layers deposited in the LFLP regime exhibit higher oxygen concentrations compared to the HFHP regime for the same applied oxygen flow. At pressures between 10^{-8} and 10^{-6} Torr, the difference in oxygen concentration between the two deposition regimes is approximately half an order of magnitude. At 10^{-5} Torr, this difference in oxygen concentration is already of more than one order of magnitude. Thus, the amount of incorporated oxygen is strongly influenced by the change in deposition parameters, such as total gas flow.

For nitrogen in the HFHP regime [Fig. 4(b)], the situation is similar to oxygen, save that the total concentration of incorporated species is within 10^{17}–10^{20} atoms/cm^3. This is considerably lower than the oxygen content of 10^{18}–10^{21} atoms/cm^3 over the whole base pressure range investigated. Thus, the incorporation probability for nitrogen is approximately one order of magnitude lower than that of oxygen.

IV. DISCUSSION

The existence of critical impurity concentrations for oxygen and nitrogen in *a*-Si:H solar cells and the similarity to the findings for μc-Si:H solar cells[12] are interesting results deserving further investigation.

In the presented *a*-Si:H solar cells, the *I*-*V* parameters deteriorate mainly by increasing impurity concentration due to losses in the FF, especially under red illumination. Based on the results for electrical and optical properties of single *i*-layers, oxygen and nitrogen are likely incorporated into the absorber layer (in part) as donor and/or defect states. This model was proposed by Kinoshita *et al.*,[3] who worked on cells and films deposited from pure silane PECVD plasma. Threefold-coordinated oxygen (O_3^+) and fourfold-coordinated nitrogen (N_4^+) are considered to be the doping configurations.[2] The decrease in solar cell performance is mainly attributed to changes in the electric field in the absorber layer.[3] The donors create a positive space charge in the vicinity of the *p*/*i*-interface, strengthening the field at this particular location. This would explain why the solar cell properties under blue illumination do not suffer much from the impurity incorporation until strong changes in the absorber layer structure appear. The created charge carriers can be even better extracted when generated at the *p*/*i*-interface for a moderate contamination. Thus, moderate contamination improves the *p*/*i*-interface of solar cells. As positive as the effect is for the photogenerated charge carriers at the *p*/*i*-interface, so negative is it for the charge carriers created in the bulk absorber layer by long wavelength light. Here, the reduced electric field leads to lower extraction efficiency. This can be observed in the strong sensitivity of the cell properties under red illumination. Nevertheless, the increased subgap absorption (observed by CPM) for contaminated solar cells can, to some extent, originate from defect states leading to a suppression of the bulk carrier collection. A Fermi level shift toward the conduction band will also lead to an increased concentration of charged defects as observed by the CPM measurements. This increase in charged defect density could in turn lead to a lifetime reduction in the generated carriers and deterioration of the solar cell performance. Further aspects supporting this model are the influence of the *i*-layer thickness and the influence of the location of the impurities on the contamination sensitivity.[1,12] For μc-Si:H it was found that for a thin *i*-layer or an *i*-layer contaminated only in the vicinity of the *n*-layer, oxygen and nitrogen contaminations do not decrease the cell properties as much as for thick *i*-layers or *i*-layers contaminated only in the vicinity of the *p*-layer.[12]

It has been shown that a shift in the Fermi level toward the conduction band is accompanied by the effect of a changed electric field by the creation of donor states.[3] This can explain the decrease in activation energy and the increase in dark conductivity observed for single *i*-layers for oxygen and nitrogen incorporation (see Fig. 2). An increase in photoconductivity is also seen, which is typical for doping of *a*-Si:H.[13] Up to an impurity concentration of 10^{20} atoms/cm^{-3}, the variations in dark conductivity, activation energy, and photosensitivity are quite similar for oxygen and nitrogen incorporation. At oxygen concentrations higher than 10^{20} atoms/cm^{-3}, the activation energy is found to increase again. This effect is interpreted as an influence of alloying, i.e., of the formation of Si–O–Si bonds leading to an increase in the band gap as observed by CPM. Corresponding effects, namely, the incorporation of threefold coordinated nitrogen, are likely to occur for nitrogen concentrations exceeding 10^{20} atoms/cm^{-3}, but such samples cannot be prepared with the present deposition conditions. It is well known that the optical gap of Si–N alloys increases strongly when the nitrogen concentration exceeds a few atomic percents.

The electrical properties of the *i*-layers corresponded closely to the solar cell parameters of the cells deposited with the same conditions for the *i*-layer. For example, at the impurity concentration of 10^{19} atoms/cm^3 (see Fig. 1), the FF and cell efficiency are lower for nitrogen ($\mathrm{FF}=70\%$, $\eta=8\%$) than for oxygen ($\mathrm{FF}=74\%$, $\eta=8.7\%$).

Since the impurity concentration was found to influence the solar cell performance, a deposition regime in which the oxygen and nitrogen impurity incorporation is suppressed was sought. Indeed, as seen in Fig. 4(a), by changing from LFLP to HFHP deposition, an increased base pressure by a factor of almost 10 leads to nearly identical oxygen concentrations. For this less efficient incorporation of impurities in the HFHP regime, several reasons are conceivable. In particular, a higher flow rate of the process gases (SiH_4 and H_2) can lead to a decreased impurity incorporation for a given impurity flow rate (base pressure). In addition, the heater temperature decrease from 260 to 200 °C may reduce the amount of impurities originating from outgassing at the chamber walls.[7] This effect may also be responsible for the enhancement of the baseline contamination for the LFLP regime (see Fig. 4). Comparing the incorporation of nitrogen and oxygen in the HFHP regime, the results show that oxygen is more efficiently incorporated than nitrogen. For example, at a contamination related base pressure of 10^{-5} Torr, a concentration of 3×10^{18} atoms/cm^3 of nitrogen was found in the solar cells whereas the amount of oxygen was 3×10^{19} atoms/cm^3. A reason for this difference may be the higher energy necessary to break up nitrogen molecules (compared to oxygen molecules) into atoms, in order to incorporate them (as atoms) into the growing film. There may also be a higher sticking coefficient of oxygen compared to nitrogen on the growing *a*-Si surface. Together with the lower critical contamination level for nitrogen compared to oxygen (due to the higher nitrogen doping efficiency), it is seen that for air leakages or poor vacuum systems, the oxygen incorporation is the more crucial factor determining the solar cell quality.

V. CONCLUSION

The results show that the electrical properties of *a*-Si:H solar cells deteriorate when the critical contamination levels in the *i*-layer of 2×10^{19} atoms/cm^{-3} for oxygen and of 4×10^{18} atoms/cm^3 for nitrogen are reached. These levels are similar to those observed for μc-Si:H solar cells. The decay in efficiency is attributed to a shift in the Fermi level toward the conduction band, related to an incorporation of a fraction of oxygen and nitrogen atoms in doping configurations. Under equal deposition conditions, the critical contamination level is at lower impurity flows for oxygen than for nitrogen. The oxygen incorporation efficiency was decreased significantly by applying changed deposition parameters (higher total gas flow, pressure, deposition rate, and lower heater temperature). Applying the HFHP regime in state-of-the-art solar cell production, the vacuum requirements may be reduced because a higher base pressure could be tolerated. This, in turn, could reduce overall production costs.

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