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Hydrogen related phenomena at the ITO/a-Si:H/Si heterojunction solar cell interfaces

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Properties of thin a-Si:H and indium-tin oxide (ITO) layers as well as properties of interfaces of Si based heterojunction (HJ) ITO/(p)a-Si:H/n-Si structures were analyzed by means of atomic force microscopy (AFM) and scanning spreading resistance microscopy. It is shown that the morphology of thin ITO layers grown on n-type polished crystalline Si or on (p)a-Si:H/n-Si substrates depends on the deposition temperature and has peculiarities on a nano-scale. Formation of highly conductive nano-dots on the surface and in the bulk of ITO layers is found. The observed nano-spots and nano-dots are attributed to the influence of hydrogen initiated reduction process, which occurs upon deposition of ITO films on an a-Si:H layer during the fabrication process of a HJ solar cell. This fact is confirmed by investigation of morphological properties of ITO surfaces after treatment by hydrogen plasma. It is shown that formation of conductive nano-particles on the ITO surface initiated by hydrogen does not change essentially transparency of an ITO layer. It is concluded that conductive nano-dots at the ITO/a.Si:H interface can be considered as local conductive channels, which provide a current flow through the ITO/(p)a-Si:H interface without essential shadowing of the solar cell structure. This finding opens an interesting way for the optimization of properties of the ITO/Si-based HJ solar cells.

1 Introduction Investigations of Si based heterojunction (HJ) solar cells, which consist of transparent conductive oxide (TCO)/Si structures started a long time ago [1–6] and are still intensive nowadays [7]. Such solar cells are regarded as low-cost photovoltaic devices. The cost reduction is provided by the low-temperature junction-formation step and by the unique properties of the TCO layer, which serves as an emitter, a conductive top electrode and as an antireflection coating. A remarkable progress for such solar cells has been demonstrated by SANYO [8] using an a-Si:H as an emitter as well as a buffer layer between the TCO top electrode and the Si substrate. Nevertheless, a number of problems in this field still remain unsolved [7, 9]. In particular, it is not clear so far why the efficiency of such solar cells is higher for n-type substrates than for p-type ones, and this represents a subject for intensive investigations [9–11]. It is interesting to note that this problem has been discussed earlier with regard to the TCO/Si solar cell efficiency. It was found that in many cases only n-type silicon substrates could provide fabrication of high-efficiency HJ solar cells [6]. It was established that peculiarities of the interfaces between the TCOs, such as indium-tin oxide (ITO), and the Si substrate are responsible for this phenomenon. These peculiarities were attributed to the formation of defects on the ITO/Si interface since the TCO layer growth, which is usually done by a magnetron sputtering, leads to formation of ion damages in the Si subsurface region of the Si substrate. These damages induce a positive charge decreasing the surface barrier height in the case of n-type Si, while increasing the barrier in the case of p-type Si [12]. The interface properties of TCO/Si structures have been intensively studied in the past [13] and recently [14, 15] by means of current- and capacitance–voltage measurements. However, because of nano-size dimensions of the ITO (70–80 nm) and a-Si:H (3–20 nm) layers in ITO/Si and ITO/a-Si:H/Si solar cells these structures have to be investigated by atomic force microscopy (AFM) and scanning spreading resistance microscopy (SSRM) methods, as it was demonstrated in [16, 17]. It is worth noting that these analytical methods are widely used for nano-size systems

and it is reasonable to apply them for Si-based HJ solar cell structures for characterization in addition to the conventional electrical methods. The goal of this work is to investigate the properties of ITO/n-Si and ITO/(p)a-Si:H/n-Si structures and in particular, the interfaces in these structures on nano-scale, by means of AFM and SSRM methods.

2 Experimental details Polished (100)-oriented n-type Si wafers were used as substrates for the fabrication of ITO/(p)a-Si:H/n-Si HJ structures. Prior deposition of a-Si:H layer to all Si substrates the standard RCA cleaning has been applied.

The 5 nm thick p-type a-Si:H layers were deposited in the "p-chamber" of a plasma enhanced chemical vapor deposition (PECVD) set up at following process conditions: 13.56 MHz frequency, 4.3 W power, 106.4 Pa (800 mTorr) deposition pressure, ~200 °C deposition temperature, 40 sccm flow rate for SiH4 and 10 sccm for 2% mixture of trymethylboron in He.

The ITO layers with thicknesses of 70–80 nm were deposited at room temperature (RT), 160 and at 230 °C (substrate temperatures) using an ITO sintered target, with In_2O_3 and SnO_2 in a weight proportion of 9:1. The DC plasma power was 100 W at all deposition temperatures. The base pressure in the sputter system was about 1.33 Pa (10–5 Torr). The total pressure of the sputtering gas mixture was adjusted to 0.399 Pa (3 mTorr) during the film preparation. The argon flow rate was kept constant at 38 sccm.

The AFM and SSRM measurements were performed using a Digital Instrument's Nanoscope Dim 3100 microscope equipped with spreading resistance and capacitance measurement electronics. The AFM measurements were performed in tapping mode using commercial silicon tips MikroMasch NSC35/AlBS with a typical tip curvature radius of less than 10 nm. The following parameters were used for the analysis of the AFM measurements: (i) the root mean square (RMS) roughness (R_q), which gives the root mean square average of height deviations taken from the mean data plane within a given area; (ii) the mean roughness (R_a), which represents the arithmetic average of the absolute values of the surface height deviations measured from the mean plane; (iii) the difference in height between the highest and lowest points on the surface relative to the mean plane (h_{max}); (iv) the average differences of heights (h_{aver}) [18].

The SSRM measurements were performed using commercial conductive B-doped diamond coated silicon tips NanoSensors CDT/NCHR with a microscopic tip radius of 10 nm (on nano-roughness). A DC bias was applied to the tip and the current flowing through the sample was measured by a logarithmic current amplifier. The SSRM measurements for ITO/a-Si:H/Si structures were performed in a "cross-section" configuration: one electrode has been made on the back side of the solar cell structure and the conductive tip was used to perform mapping of the ITO surface conductivity. Thus, the vertical current through the ITO/a-Si:H/Si structure was measured in this case. In case of ITO/glass structure, one electrode was made on the ITO surface and the conductive tip of the SSRM system (second electrode) was measuring the lateral current distribution.

To investigate the influence of atomic hydrogen on the properties of the ITO layers, sample hydrogenation was performed in the PECVD setup at 230 °C for 2 min using a 110 MHz plasma generator. The RF plasma density was 0.04 W cm^{-2} during the hydrogenation process, while the ignition of plasma was done at higher densities (above 0.1 W cm^{-2}). At the stage of the plasma ignition the samples were removed from the plasma area and then reintroduced once the discharge had been stabilized at the density of 0.04 W cm^{-2} with the hydrogen gas flux of 200 sccm.

Direct transmittance of ITO glass samples was measured in the spectral range of 250–3300 nm with a UV/VIS/NIR double channel spectrophotometer (Perkin Elmer Lambda 950).

3 Results and discussions Figure 1 shows the surface morphology (3D AFM image) of (p)a-Si:H layers deposited on (100) oriented Si substrate at a temperature about 200 °C. From Fig. 1 it can be concluded that the a-Si:H layer is rather flat on nano-sclale, without any specific morphological peculiarities.

Figure 2 shows the surface morphology of ITO layers deposited on (p)a-Si:H/n-Si substrates at RT (Fig. 3a), 160 °C (Fig. 3b) and 230 °C (Fig. 3c).

From these images one may conclude that the roughness of ITO layers deposited at 160 and 230 °C on a hydrogen containing a-Si:H layer is higher than for the case of the RT deposition. This means that the morphology of ITO layers, as well as other properties, depend substantially on the deposition temperature, as it was reported in [16, 20] for similar ITO layers deposited by magnetron sputtering on Si and glass substrates at different temperatures [19]. Important to note that trends for ITO/glass structures are different: the surface roughness of ITO layers decreases with increasing of the deposition temperature [19].

Figure 3a shows the SSRM image of the ITO (RT)/(p)a-Si:H/n-Si surface.

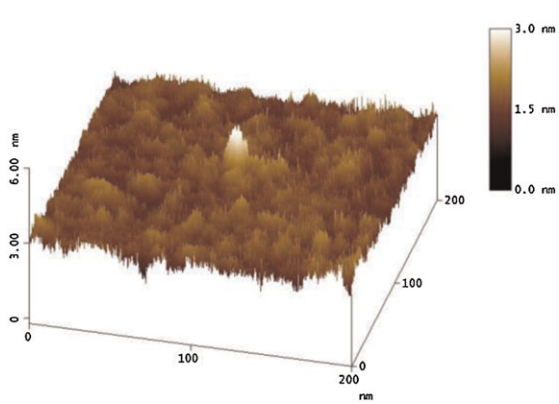

Figure 1 (online color at: www.pss-a.com) AFM 3D image of (p)a-Si:H layer deposited at ~200 °C on an n-type polished Si substrate ($R_q = 0.164$ nm, $R_a = 0.129$ nm, $h_{max} = 1.741$ nm, $h_{aver} = 0.7$ nm).

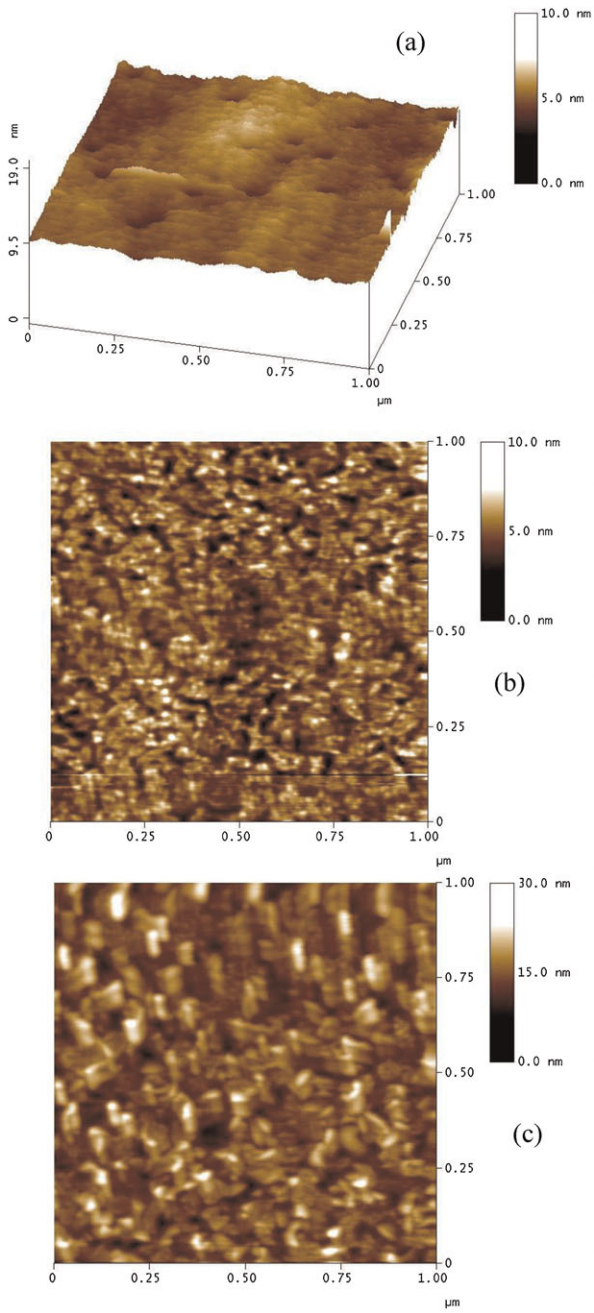

Figure 2 (online color at: www.pss-a.com) AFM 3D image of ITO layer deposited on (p)a-Si:H/n-Si substrate at (a) RT ($R_q = 0.29$ nm, $R_a = 0.23$ nm, $h_{max} = 3.6$ nm, $h_{aver} = 1.3$ nm); (b) 2D images of ITO layer deposited at 160 °C ($R_q = 1.0$ nm, $R_a = 0.74$ nm, $h_{max} = 30$ nm, $h_{aver} = 11.3$ nm); and (c) 230 °C ($R_q = 3.12$ nm, $R_a = 2.43$ nm, $h_{max} = 22$ nm, $h_{aver} = 14.6$ nm).

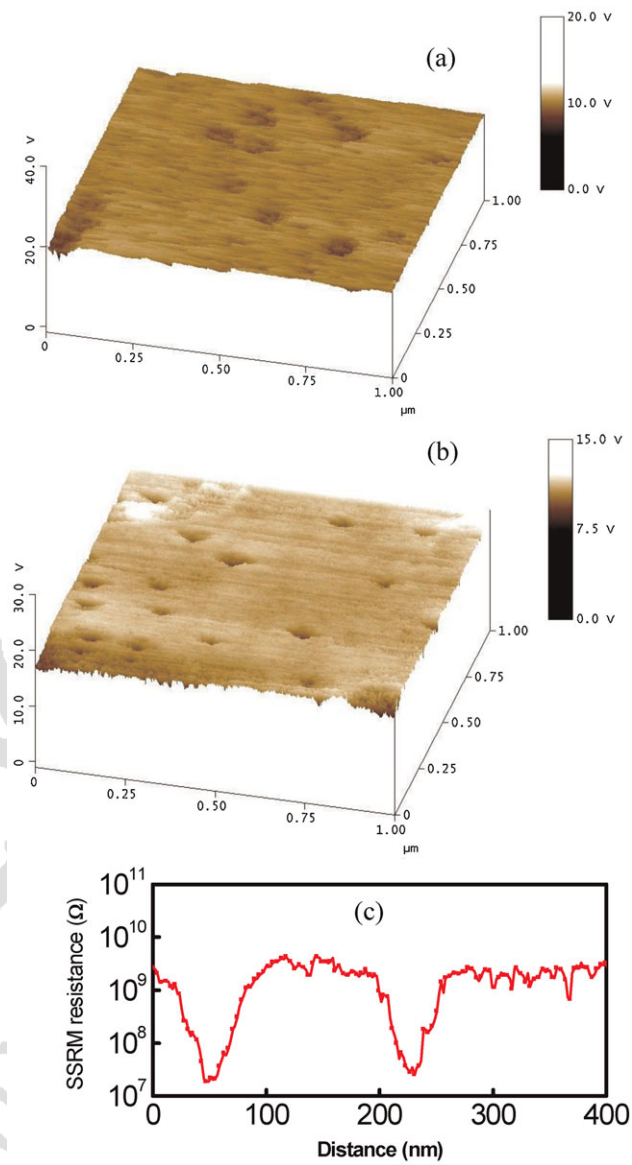

Figure 3 (online color at: www.pss-a.com) (a) SSRM 2D image of ITO (RT)/(p)a-Si:H/n-Si surface; (b) SSRM 2D image of ITO (RT)/(p)a-Si:H/Si surface after 5% HF treatment for 2 min; (c) linear SSRM profile obtained on the surface of the HF etched ITO (RT)/(p)a-Si:H/n-Si structure.

A DC forward bias of 0.1 V was applied to the tip and the current flowing through the sample was measured by a logarithmic current amplifier.

The SSRM data are presented using a voltage scale, and can be converted by a standard procedure to the spreading resistance values [18]. Darker spots (nano-dots) on the SSRM image correspond to local regions with lower resistance. It is interesting to note that these highly conductive nano-dots remain after etching of the ITO layers by 5% HF (Fig. 3b). AFM measurements for etched samples show similar morphology as for a-Si:H/Si structures (Fig. 1), which indicates that ITO layer has been removed completely by the HF etching. The linear spreading resistance profile shown in Fig. 3c indicates that the difference in absolute values of the spreading resistance for different local regions may be as large as two orders of magnitude, approximately. Therefore, it can be concluded that these highly conductive

local regions provide a current flow through the conductive nano-channels at the ITO/(p)a-Si:H interface. Nevertheless it has to be noted that at this stage it is not clear how the observed highly conductive channels have been formed. The following scenario can be considered, as the most probable version: metallic rich clusters with high conductivity can be formed at the initial stage of the ITO growth on top of an a-Si:H layer due to reduction process initiated by hydrogen, released from the a-Si:H layer. Independently on the reasons, which lead to the formation of such conductive local regions at the ITO/Si interfaces, it can be stated that properties of these conductive nano-channels (size, density, conductivity, etc.) are very probably crucial for the properties of Si based HJ solar cells. The validity of this conclusion should be verified by a more detailed examination of the nano-scale properties of ITO/a-Si:H/Si structures. Such analysis is ultimately necessary and extremely important in this case, since conventional techniques like DLTS or CV methods have limited spatial resolution.

It has to be noted that in [9] it was demonstrated that a release of hydrogen from the a-Si:H layer occurs during the ITO deposition process. It can be assumed that the hydrogen release could be responsible for the observed morphological peculiarities due to ability of hydrogen to initiate reduction processes for any oxides.

In order to prove that hydrogen can affect properties of thin ITO layers, a flat 80 nm thick ITO layer was deposited on a glass substrate at 230 °C, and subsequently hydrogenated at 230 °C for 2 min.

Figure 4a shows 2D images for ITO (230 °C)/glass structure before and after (Fig. 4b) the hydrogen plasma treatment at 230 °C for 2 min. Important to note that ITO layer deposited at 230 °C is rather flat and any changes of the morphology for such layers can be detected easy. From Fig. 4b it can be seen that formation of nano-structures occurs as fast as after just 2 min of hydrogenation with a power density as low as 0.04 W cm^{-2}; the structures have a typical size of 20–40 nm with a tendency to agglomerate. From Fig. 4 it can be concluded that hydrogen plasma treatment causes remarkable changes in the morphology of ITO layer, even for rather short times of the oxide reduction process, which can be performed at rather low (230 °C) temperatures.

Figure 5a shows the SSRM 2D image obtained for the ITO (230 °C)/glass structure after a hydrogen plasma treatment at 230 °C for 2 min.

The SSRM measurements were done with a dc bias of −0.1 V applied to the tip, and the results of the SSRM mapping are shown in terms of voltage scale where darker regions correspond to higher conductivity values.

To correlate the spatial variation of the conductivity with topography, a linear SSRM scan with simultaneous data acquisition was performed (Fig. 5b). To ensure reproducibility of the measurement results and their stability against the topography induced noise, both trace and retrace scans were performed. Further, the SSRM data acquired in the

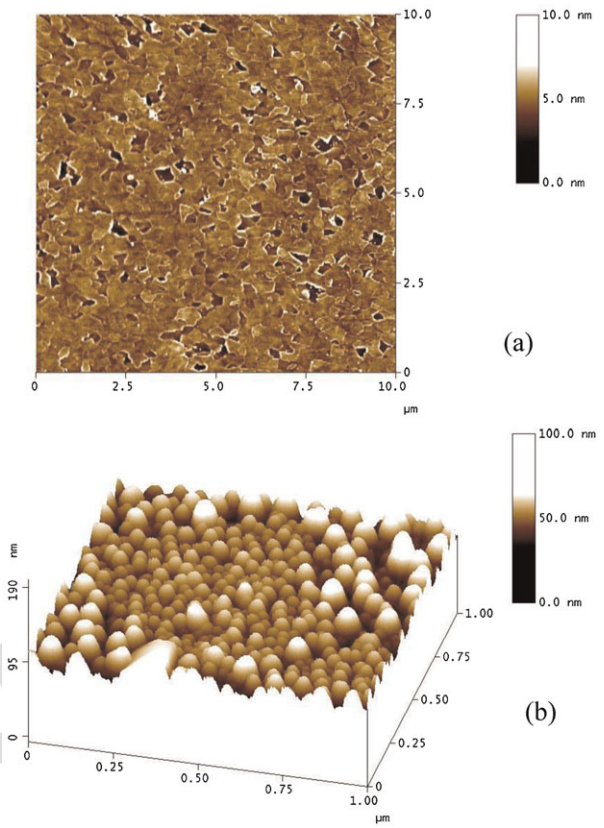

Figure 4 (online color at: www.pss-a.com) AFM images of ITO (230 °C)/glass structures: (a) – before, and (b) – after hydrogen plasma treatment at 230 °C for 2 min.

voltage scale used by the software of the Digital Instrument's Nanoscope Dim 3100 system were converted into SSRM resistance via a standard procedure described in [18]. From Fig. 5b it is seen that the morphology and the resistance do not exhibit a clear correlation. In some cases the maximum height corresponds to the maximum value of the resistance while in other cases the opposite behavior can be observed.

Thus, all nano-structures do not have maximum conductivity at their maximum heights. This indicates that the composition of the observed nano-structures at the initial stages of their formation is not fully metallic and probably consists of partially reduced oxides with high resistivity. Nevertheless, highly conductive nano-structures are clearly revealed from Fig. 5b, and the difference in the spreading resistance can be as high as 5 orders of magnitude, approximately. It should be noted that for the untreated ITO (230 °C)/glass structures, local deviations of the conductivity do not exceed one order of magnitude. The "macroscopic" resistivity of such ITO layer before hydrogenation measured by conventional four probe method is about $2.7 \times 10^{-4}\,\Omega$ cm. After hydrogenation the resistivity is almost un-changed and shows a bit better value $\sim 2.3 \times 10^{-4}\,\Omega$ cm.

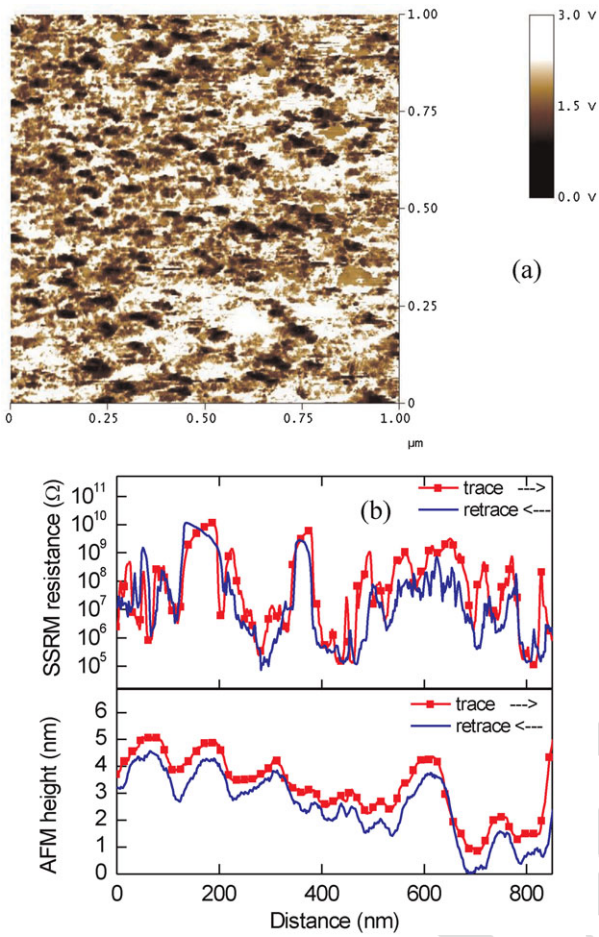

Figure 5 (online color at: www.pss-a.com) SSRM images of ITO (230 °C)/glass structures. (a) – before and (b) – after hydrogen plasma treatment at 230 °C for 2 min. Linear SSRM profile taken at an arbitrary position not indicated on the SSRM mapping image. For SSRM measurements DC bias used was −0.1 V.

Important to note that the lateral current flow in ITO/glass structure after hydrogen plasma treatment is not homogeneous, and show similar peculiarities as in case of ITO/a-Si:H structures – formation of local highly conductive regions/channels.

It can be concluded that morphological and electrical properties ITO layers can be easily modified by atomic hydrogen, which is present in hydrogen plasma. Therefore, it can be assumed that similar processes occur at the ITO/a-Si:H interface upon deposition of ITO layer since atomic hydrogen can be released from an a-Si:H layer upon deposition of ITO on an a-Si:H/Si substrate. Important to note with this regards that hydrogen release from the a-Si:H layer upon ITO deposition is a well established fact, which has been presented in [10]. Thus, although we are considering our explanation concerning the formation of highly conductive regions at the ITO/a-Si:H interfaces as an assumption, this assumption has rather solid background from our point of view.

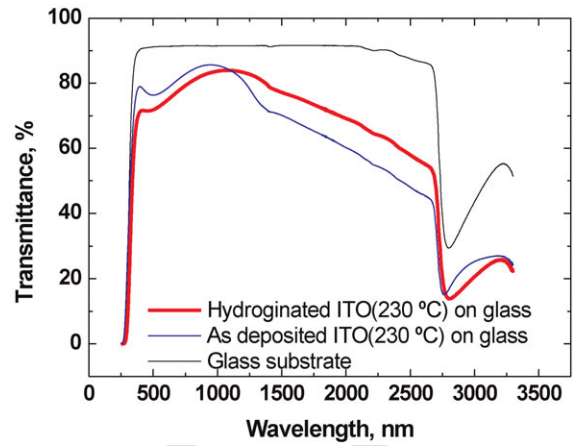

Figure 6 (online color at: www.pss-a.com) Transmittance of ITO (230 °C)/glass structure after hydrogen plasma treatment at 230 °C for 2 min.

Transmittance of ITO (230 °C)/glass structure after hydrogen plasma treatment at 230 °C for 2 min is shown in Fig. 6.

Figure 6 illustrates that despite the formation of nano-particles on the ITO surface initiated by hydrogen, such plasma-treated layer is sufficiently transparent to be used as a transparent front side electrode and at the same time as an antireflection coating for solar cells: it has more than 80% transmittance in the visible spectral range and its transmittance is even improved in the near IR, compared to that of as deposited ITO film.

4 Conclusions The AFM measurement study performed in this work has demonstrated that the morphology of thin ITO layers grown on n-type polished crystalline Si or on (p)a-Si:H/n-Si substrates depends on the deposition temperature and has peculiarities on nano-scale. Formation of nano-wells, which can be attributed to a hydrogen initiated reduction process of ITO in local regions during the ITO deposition process, is observed on the ITO surface in ITO/a-Si:H/Si structures.

Moreover, formation of highly conductive nano-dots/channels on the surface of ITO layers has been observed. These local regions provide current flow through the conductive nano-channels at the ITO/(p)a-Si:H interface.

It is necessary to underline that formation of conductive channels can be attributed to the peculiarities at the ITO/a-Si:H interface, since channels have been observed (SSRM mapping) on ITO or ITO etched surfaces of HJ ITO/a-Si:H structures. Similar peculiarities have been observed for the ITO/glass structures, after hydrogen plasma treatments, which shows that hydrogen initiated ITO structure modification is responsible for the formation of highly conductive in ITO structures.

Therefore, it is supposed that formation of highly conductive nano-channels occurs due to hydrogen initiated reduction process at the initial stage of ITO films formation

in the case of magnetron sputtering on a-Si:H layers. This assumption is supported by investigations of morphological and electrical properties of ITO surfaces in ITO/glass after treatment by hydrogen plasma, which shows that indeed, highly conductive channels in ITO layers can be formed in presence of the atomic hydrogen. Nevertheless, important to note, that the proposed explanation is only an assumption, which has to be verified by further studies of ITO/a-Si:H interfaces on nano-scale.

It has been found that nano-structure formation on ITO surface occurs at hydrogenation temperatures as low as 230 °C and as fast as for only 2 min. Formation of conductive nano-particles on the ITO surface initiated by hydrogen does not change essentially their transparency, and such treated ITO layer can be used as a transparent front side electrode and at the same time as antireflection coating for solar cells.

To optimize properties of ITO/a-Si:H/Si HJ solar cells, an adequate control of such local conductive channels formation should be guaranteed during solar cell fabrication.

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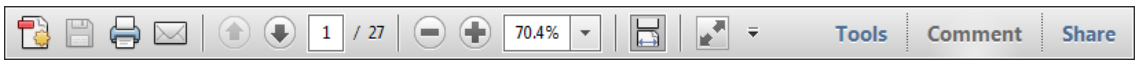

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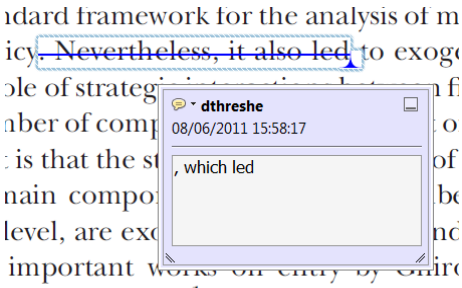

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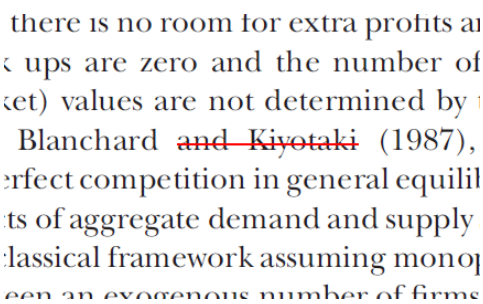

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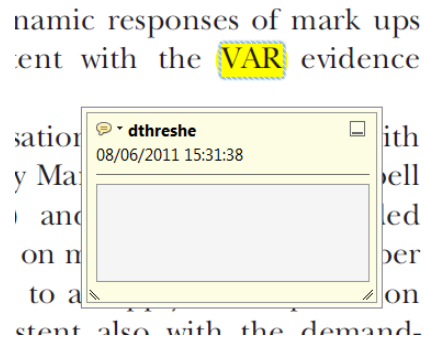

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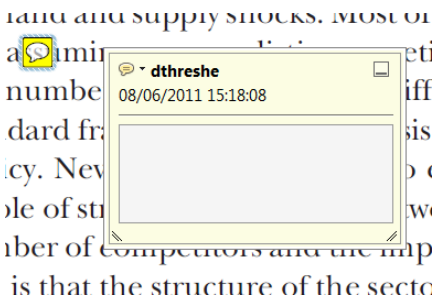
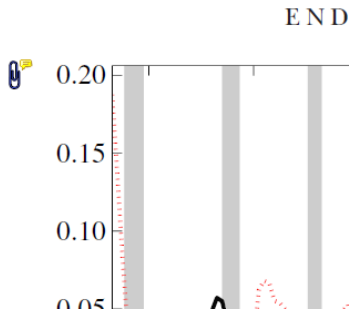


5. Attach File Tool – for inserting large amounts of text or replacement figures.

Inserts an icon linking to the attached file in the appropriate pace in the text.

How to use it

- Click on the Attach File icon in the Annotations section.
- Click on the proof to where you'd like the attached file to be linked.
- Select the file to be attached from your computer or network.
- Select the colour and type of icon that will appear in the proof. Click OK.

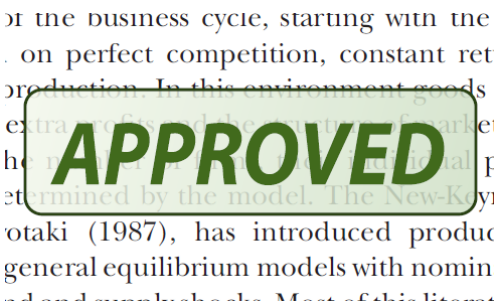


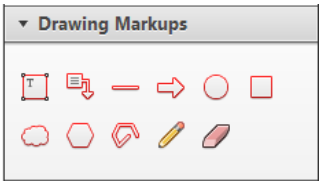
6. Add stamp Tool – for approving a proof if no corrections are required.

Inserts a selected stamp onto an appropriate place in the proof.

How to use it

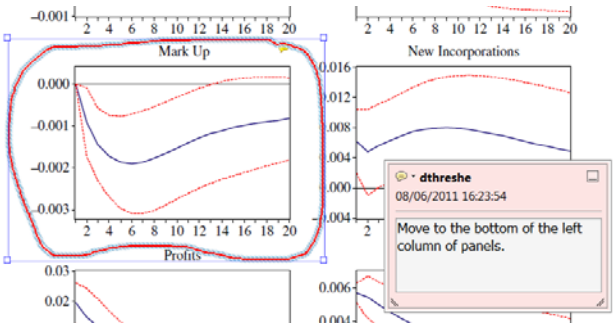
- Click on the Add stamp icon in the Annotations section.
- Select the stamp you want to use. (The Approved stamp is usually available directly in the menu that appears).
- Click on the proof where you'd like the stamp to appear. (Where a proof is to be approved as it is, this would normally be on the first page).





7. Drawing Markups Tools – for drawing shapes, lines and freeform annotations on proofs and commenting on these marks.

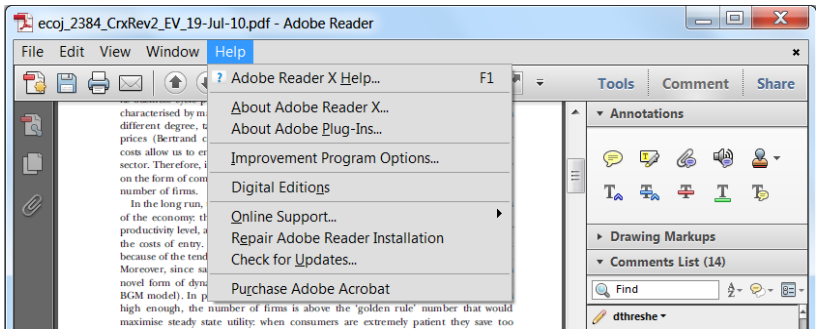
Allows shapes, lines and freeform annotations to be drawn on proofs and for comment to be made on these marks..



How to use it

- Click on one of the shapes in the Drawing Markups section.
- Click on the proof at the relevant point and draw the selected shape with the cursor.
- To add a comment to the drawn shape, move the cursor over the shape until an arrowhead appears.
- Double click on the shape and type any text in the red box that appears.

For further information on how to annotate proofs, click on the Help menu to reveal a list of further options:



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