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Sputtered ITO for application in thin-film silicon solar cells: relationship between structural and electrical properties

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Abstract

Indium tin oxide (ITO) thin films for application in thin-film silicon solar cells with superior electrical and optical properties (resistivity ranging from 1.4 to $8.4x10^{-4}$ Ωcm; transparency of >80%) have been investigated. ITO layers were deposited by radio-frequency (RF) magnetron sputtering process at different argon gas pressures and substrate temperatures ranging from room temperature to 280°C. The main goal was to identify the relationship between structural and electrical properties. Generally, ITO layers were rather smooth with granular topography; electro-optically superior layers exhibited substantially different surface morphology of large, well-organized domain formations. Hall mobility of remarkably high value of 49 cm^2/Vs (resistivity of $2.6x10^{-4}$ Ω.cm) was achieved for the ITO layers, which were deposited at surprisingly low temperature of 125°C. ITO deposition process has been successfully applied, even at room temperature, to fabricate front contacts for microcrystalline silicon solar cells, exhibiting excellent performance on both rigid and flexible substrates.

Keywords: ITO, Surface morphology, Microstructure, Hall mobility, electrical properties, µc-Si:H, nip solar cell.

1. Introduction

Indium tin oxide (ITO) is well established transparent conductive oxide (TCO) material for mass production of front contacts in flat-panel displays, as well as in thin-film silicon solar cells insubstrate configuration and in other types of thin-film solar cells [1],[2] or a-Si:H/c-Si heterojunction solar cells [3]. Further material study on deposition of ITO thin films with outstanding optical properties without compromising their electrical properties is still an important matter of research [4]. Essentially, this goal could be attained by means of increasing film quality, thus securing high mobility of free charge carriers, which would lead to decreased free carrier absorption. Development of room temperature process for front contacts deposition is also of high practical importance, since processes involving no additional heating reduce manufacturing costs and increase throughput. However, the main issue of low temperature processes is the utilization of low-cost, typically temperature sensitive substrates and the deposition onto semiconductor thin films that may change their properties upon annealing.

A vast amount of papers dealing with magnetron-sputtered ITO layers could be found [5],[6],[7],[8]. However, there are not many systematic studies on the relationship between surface morphology, microstructure and electrical properties reported for the ITO layers deposited at various temperatures and gas pressures. Moreover, room and low temperature sputtering process of ITO layers with a focus on achieving high values of Hall mobility has not been up to now intensively investigated.

In this work we investigate the relationship between structural and electrical properties of ITO layers deposited by RF magnetron sputtering process at various substrate temperatures and gas pressure conditions. Furthermore, performance of microcrystalline silicon solar cells in substrate configuration (both on rigid and flexible substrates) with ITO front contacts fabricated at room and low temperature is reported.

2. Experimental

ITO thin films were deposited on the Corning Eagle XG glass substrates by radio-frequency (RF) magnetron sputtering in a small area planar sputtering system (J.K. Lesker, USA), using 6″ target In_2O_3:SnO_2 (95/5 wt.%). Before deposition, the surface of the ITO target was pre-sputtered for at least 5 minutes at process conditions. The base pressure was in the range below 8×10^{-7} mbar and the total pressure of working atmosphere was varied between 2.7 and 6.7 μbar. Total gas flow rate was maintained constant at 10 sccm during the sputtering. The utilized sputtering power density was 1.0 W/cm^2 and the deposition was carried out at substrate temperature of 280°C, 125°C or room temperature.

The thickness of ITO thin films was measured by the surface profiler. The crystal structure was identified with a Theta Theta Diffractometer D5000 with a Goebel mirror in grazing incidence geometry with CuKα radiation. Texture grade (TC) was calculated using Equation 1, where: $I^0_{(hkl)}$ is the theoretical intensity of the particular diffraction peak given in the relevant Powder Diffraction File and $I_{(hkl)}$ is the intensity of that peak as acquired by the actual XRD measurement (k is the number of diffraction peaks considered in the analysis).

$$TC_{I_{(hkl)}} = \frac{\dfrac{I_{(hkl)}}{I^0_{(hkl)}}}{\dfrac{1}{k}\sum_{i=1}^{k}\dfrac{I_{i_{(hkl)}}}{I^0_{i_{(hkl)}}}} \qquad (1)$$

If TC equals one for each peak, that means the sample is an ideal polycrystalline material. The differences from one indicate a texture nature. Values greater than one mean that in this direction a texture can be found. For lattice planes with TC values smaller than one, there is a depletion of grains in the corresponding direction [9], [10]. The surface morphology of the ITO thin films was observed by scanning electron microscope (SEM) and by atomic force microscope (AFM) using *Park Systems XE-100* under normal air conditions, operating in non-contact mode. Average roughness R_a was determined from AFM data of 2x2 μm² scan area. The calculation was done by using Equation 2, where n is the number of equally spaced points along the trace, and y_i is the vertical distance from the mean line to the i^{th} data point.

$$R_a = \frac{1}{n}\sum_{i=1}^{n}\left|y_i\right| \qquad (2)$$

Electrical properties of the ITO thin films were investigated by Hall measurements in van der Pauw geometry at room temperature. Optical properties of ITO thin films were measured using UV/VIS/NIR spectrophotometer PerkinElmer LAMBDA 950.

The RF magnetron-sputtered ITO thin layers were utilized as front contacts in microcrystalline silicon solar cells in substrate configuration, both on rigid (Corning Eagle XG glass) and flexible substrates (polyimide foils). Microcrystalline silicon (μc-Si:H) solar cells were deposited in n–i–p sequence by plasma enhanced chemical vapour deposition (PECVD) in a multichamber UHV system using standard conditions. Additional details on the preparation of the solar cells on glass can be found in [11],[12] and solar cells on foils in [13]. The sputtering of ITO front contacts was carried out in the gas mixture of argon and oxygen. The relative oxygen concentration in the working atmosphere, defined as *gas flow rate*(O_2) / *gas flow rate* (O_2+Ar), was 0.1%. Test cells with the area of 1 x 1 cm^2 were defined by using mask during ITO deposition. Front metal finger electrodes were prepared by silver evaporation. Finally, standard annealing procedure of finished solar cells was performed in air at 160°C for 30 minutes. The complete solar cell structures consist of the following layers: substrate (polyimide or glass) / texture-etched ZnO / silver / ZnO (80 nm) / n-i-p μc-Si:H layers / ITO front contact / Ag bus bar. The silver bus bars, consisting of interconnected finger electrodes spaced approximately 1 mm away from each other, supported the conductivity of front contact. JV characterization of fabricated cells was performed under AM1.5 irradiation, not taking into account the shadowed area by the Ag fingers at the front side. The external quantum efficiency (EQE) of the solar cells was measured by differential spectral response at zero bias.

The aim of this work was to study the relationship between structural and electrical properties of ITO layers applicable in thin-film silicon solar cells. Our investigations were primarily focused on such sputtering conditions, which result in ITO layers with superior electro-optical properties. It was reported that electrical parameters are not further improved at substrate temperatures higher than 300°C [14] or 350°C [15]. Moreover, gas pressure above 6.7 μbar deteriorates electrical properties of sputtered ITO layers [16]. Therefore substrate temperature during ITO deposition was varied between room temperature and 280°C at gas pressures up to 6.7 μbar. In order to reduce errors originating from imprecise film thickness estimation and to ensure possibility to reveal the microstructure of the ITO layers, we have decided to investigate thicker ITO films of ~600 nm. Application to solar cells was carried out using thinner ITO layers of ~80 nm, assuming that observed trends for thicker films are valid also for thinner ones.

3. Results and discussion

3.1 Electrical and optical properties of ITO layers

Electrical parameters of ITO layers, such as resistivity, concentration of free charge carriers and Hall mobility, are shown in Fig.1. Resistivity of ITO layers deposited at 2.7 μbar improved from 7.4 to 1.6×10^{-4} Ωcm and carrier concentration increased from 3.7 to 6.0×10^{20} cm^{-3} due to increasing substrate temperature (see Fig.1). Heating the substrate at surprisingly low temperature of only 125°C resulted in ITO layers with remarkably high Hall mobility of 49 cm^2/Vs (resistivity of 2.6×10^{-4} Ωcm). This fact explains much lower sheet resistance and better optical properties of samples deposited at 125°C in comparison to room-temperature

(RT) sputtered ITO layers. Absorption edge (as noticeable from Fig.2a) of RT sputtered ITO layers is shifted towards greater wavelength due to Burstein-Moss shift effect [17]. Moreover, optical absorption in the visible range is higher for ITO layers deposited at RT, indicating higher density of defects in the films [18].

Electrical properties of ITO layers deposited at 6.7 μbar showed also the tendency of increasing carrier concentration and decreasing resistivity due to substrate heating, same as for those layers deposited at 2.7 μbar. ITO layers deposited at substrate temperatures of 280°C possessed one of the lowest resistivities of 1.4×10^{-4} Ωcm (sheet resistance of 2.4 Ω at thickness of 570 nm) among all investigated ITO layers, thank to the very high carrier concentration of 9.7×10^{20} cm^{-3}, which was also a reason for increased optical absorption in near-infrared wavelength region (NIR). ITO layers deposited at 125°C exhibited substantially lower optical absorption in the whole visible (VIS) and NIR wavelength region than RT ITO, as shown in Fig.2b. The absorption of RT ITO layers in VIS region is mainly attributed to the amount of defect states in the films, which seems to be higher than that for RT ITO layers deposited at 2.7 μbar.

Increasing argon pressure did not influence carrier concentration for RT ITO layers, which was in the range of $3.3\text{-}3.8 \times 10^{20}$ cm^{-3}. However, resistivity dropped from 9.8 to 5.8×10^{-4} Ωcm, thank to the improvement of Hall mobility from 20 to 30 cm^2/Vs. ITO layers deposited at elevated temperatures exhibited bigger variation of carrier concentration, but the variation of resistivity and Hall mobility was smaller, when compared with RT ITO layers.

3.2 Structural properties of ITO layers

Fig.3 depicts a matrix of surface morphologies of 560-610 nm thick ITO layers deposited at pressures ranging from 2.7 to 6.7 μbar pure argon (horizontal) and substrate temperatures from room temperature to 280°C (vertical). Surfaces of all ITO layers were very smooth, not exceeding the average roughness R_a of 9 nm. The surface structures range from small grained features to large domains (lateral length up to 1 μm), that still exhibit small grains inside the domain. Room-temperature sputtered ITO layers deposited at 2.7 μbar exhibited disorganized smooth surface, which moved into domain topography due to elevated substrate temperature. ITO layers prepared at 6.7 μbar showed similar trend of changing surface topography towards larger domain structures due to substrate heating. These ITO layers prepared at RT had organized grains in small domains, which gradually transformed into bigger features in larger domains at 125°C and further into even larger, well-oriented domains of fine features at 280°C. By increasing argon pressure from 2.7 to 6.7 µbar during deposition of ITO layers onto unheated substrates, the surface changed from disordered smooth structure to the surface characterized by domain formations with aligned surface grains.

XRD measurements revealed that grains of the ITO layers crystallized in several crystallographic orientations. All X-ray reflexes were deduced from the body-centred cubic structure of In_2O_3 [19]. Full-width at half maximum (FWHM) values of diffraction peaks and shift of major peaks in relation to corresponding ideal peak positions, as taken from powder diffractogram, are shown in Fig. 4 and Fig. 5 as function of substrate temperature and sputter pressure, respectively. The XRD results of ITO layers deposited at 2.7 µbar showed decreasing trend of FWHM values of major (222) and (440) diffraction peaks (as shown in Fig.4a), as well as shifting peak to smaller 2-Theta angles (closer to their ideal positions in In_2O_3 lattice) due to increasing substrate temperature during sputtering (shown in Fig.4b). Similarly for RT ITO layers, FWHM values of diffraction peaks and peak shifts decreased as the pressure increased from 2.7 to 6.7 µbar (see Fig. 5a, b).

Texture grades of all significant diffraction planes found in the diffractograms were calculated. Fig. 6 shows texture grades of the diffraction planes of ITO layers deposited at 2.7 µbar. Diffraction peak (222) declined as the substrate temperature increased from room temperature (RT) to 280°C. Furthermore, texture grade calculation revealed that during depositions at low heating temperatures, incoming species prefer to crystallize in (222) plane but not in (400) orientation, which is present only for layers deposited at 280°C. Similar findings were reported in [20]. ITO layers deposited at 280°C exhibited more random orientation, supported by the fact that texture grade of all diffraction planes is close to unity. Texture grade calculation of the ITO layers deposited at 6.7 µbar (Fig.7) showed that (400) crystallographic orientation is preferred for high substrate temperature of 280°C, whereas orientation (222) is the most pronounced for the ITO layers grown at 125°C. At the room temperature by increasing deposition pressure from 2.7 to 6.7 µbar, (222) diffraction peak is decreasing (Fig.8). Only RT ITO layers deposited at 6.7 µbar exhibited (400) diffraction peak and at the same time only these layers had all texture grades close to unity, meaning they exhibit random orientation of the crystallites.

3.3 Application to μc-Si:H solar cells in substrate configuration

ITO layers with thickness of ~80 nm have been optimized for the application as front contacts in μc-Si:H solar cells, especially in terms of their sheet resistance, optical properties and thickness in the range of 70-80 nm. Fig. 9 illustrates the influence of oxygen admixture on the optical properties of ITO thin films grown at room temperature. Correspondingly, sheet resistance was increased from 43 to over 3000 Ω, while increasing oxygen concentration from 0% to 0.9% in the sputtering gas. It is well-known that oxygen admixture during deposition improves optical properties of ITO layers and increases sheet resistance [21].

As a final step to elaborate the quality of ITO, selected layers were used in the state-of the-art μc-Si:H solar cells as front contacts. ITO layers deposited at 125°C and room temperature were utilized in the cells deposited on both glass and polyimide substrates. Performance of μc-Si:H solar cells deposited on glass with ZnO front contact (already reported before in [22]) are shown for comparison. The thicknesses of the intrinsic layer were 1.1 μm and 1.8 μm for the cells deposited on glass and polyimide substrates, respectively. The J-V parameters: efficiency (eta), open circuit voltage (V_{oc}), fill factor (FF) and short circuit current density (J_{sc}) of the best solar cells achieved up to now are summarized in Table 1. Maximum efficiency of 8.4% and 7.5% was obtained for a 1cm² cell on a glass and polyimide substrate, respectively. High J_{SC} above 23 mA/cm^2 was achieved for the cells on both types of substrate as evident from EQE results (see Fig. 10).

3.4 Discussion

Pressure variation had smaller impact on structural and electrical properties of ITO than the influence of the substrate temperature. Increasing substrate temperature at both low and high pressures caused the increase of carrier concentration and Hall mobility. However, the mobility of ITO layers grown at 280°C was not further improved in comparison to 125°C. On the other hand, varying argon pressure did not influence the carrier concentration, nevertheless the resistivity was improved for RT ITO thank to slight increase of Hall mobility.

We have observed some general trends concerning the influence of substrate temperature and increasing total gas pressure on the surface morphology and microstructure. The surface of ITO layers generally changed from disorganized grain structure or small grain domains to topography characterized by large, well-organized domains of fine features, as a consequence of elevated substrate temperature during deposition. Increasing pressure at RT also caused that ITO surfaces looked differently: layers deposited at higher pressures had grains better organized as those grown at lower pressures. The shift of diffraction peaks closer to ideal positions of corresponding peak positions in In_2O_3 powder was considered as an indicator of the stress release in the films [23]. Thus, based on XRD measurements, stress has been released and the values of FWHM of the major diffraction peaks declined due to increased substrate temperature as well as argon pressure.

The ITO layers with increased values of Hall mobility were achieved thank to elevated substrate temperature and deposition pressure. At the same time, the surface morphology of such ITO layers is characterized by presence of grains organized in domains. Moreover, taking into account stress release and lower FWHM values due to increasing substrate temperature and deposition pressure, one can conclude that the microstructure of these films was improved in comparison with those deposited at RT and lower pressures. Based on these results, we conclude that high values of Hall mobility are directly correlated with surface morphology, which differs from other ITO layers of lower Hall mobility and is characterized by domain formations. Such morphology is called grain-subgrain structure in the literature [24]. Therefore surface morphology provides information about the microstructure quality correlated with Hall mobility. Thus, the large domains seem to favour high mobility, which is related to the density and maybe the type of grain boundaries seen on the surface by AFM. As the small grains inside one domain are of the same size as the grains in low mobility material, their grain boundary structure does not limit the mobility of free carriers, indicating narrow or well-structured grain boundaries.

4. Summary

The ITO layers deposited by RF magnetron sputtering exhibited a resistivity in the 10^{-4} Ωcm range and a carrier concentration up to 10^{21} cm^{-3}. The ITO layers deposited at elevated substrate temperatures had values of Hall mobility above 41 cm^2/Vs. By heating the substrate at surprisingly low temperature of only 125°C, it was achieved to deposit the ITO layers with remarkably high Hall mobility of 49 cm^2/Vs, exhibiting superior resistivity of 2.6×10^{-4} Ωcm.

In general, surfaces of all ITO layers were very smooth, having grains sometimes organized in domains. XRD investigations of the ITO layers showed that the layers were polycrystalline, but not all of them exhibited strong (222) or (400) preferential orientation.

ITO layers deposited at pressure and substrate temperature of 2.7 µbar, 280°C and those layers deposited at 6.7 µbar, RT had texture grade of all diffraction peaks close to 1, indicating their random nature. Increasing substrate temperature as well as deposition pressure improved the microstructure of the ITO layers, as deduced from decreasing FWHM values of diffraction peaks and stress release.

Relationship between structural and electrical properties of ITO layers on one side and the parameters of RF magnetron-sputtering process on the other has been identified. Transition from disorganized surface morphology to domain-type morphology always improved the Hall mobility. By taking it into consideration and also the relationship between microstructure quality and electrical properties, a correlation between surface morphology and enhanced Hall mobility could be revealed for the investigated ITO layers. To sum up, high values of Hall mobility could be explained by improved microstructure, which might be correlated with the surface morphology characterized by presence of grains in the domains.

The results of the solar cell studies demonstrate that ITO layers grown at low temperatures developed in this work can be successfully used as a front contact in the case of microcrystalline cells deposited on glass or polyimide substrates. High conversion efficiency of 8.4% and J_{SC} above 23mA/cm^2 were achieved. The solar cells deposited on glass with ITO front contact outperformed those with ZnO front contact.

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Table 1
J-V parameters of the µc-Si:H solar cells under AM1.5 irradiation.

substrate type	i-layer thickness (µm)	front contact	η (%)	FF (%)	V_{OC} (V)	J_{SC} (mA/cm^2)
flexible (polyimide)	1.8	**ITO**	7.5	68.0	0.475	23.1
rigid (glass)	1.1	**ITO**	8.4	70.5	0.516	23.2
rigid (glass)	1.1	**ZnO**	8.2	70.9	0.507	22.9

Figure captions

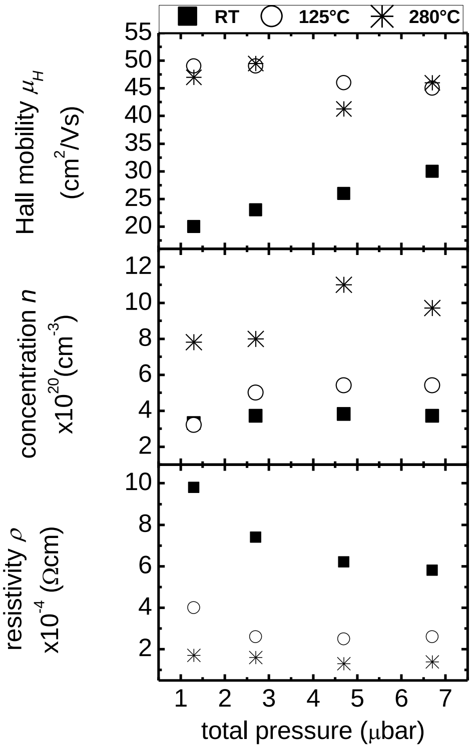

Fig.1. Electrical parameters of ITO layers in relation to the argon pressure and substrate temperature during deposition.

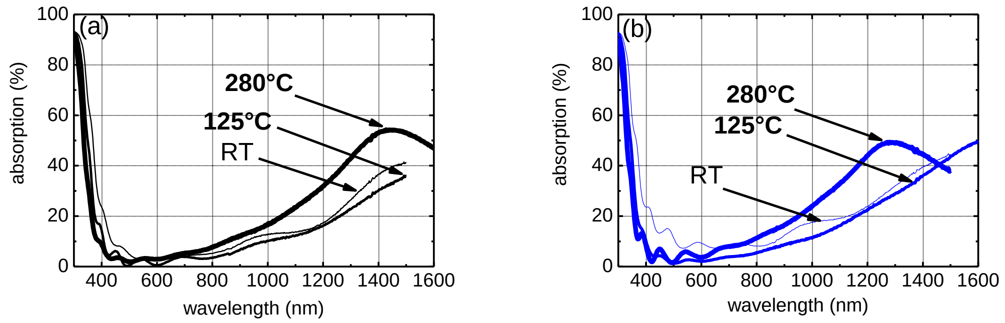

Fig.2. Optical absorption of ITO layers deposited at a) 2.7 µbar and b) 6.7 µbar.

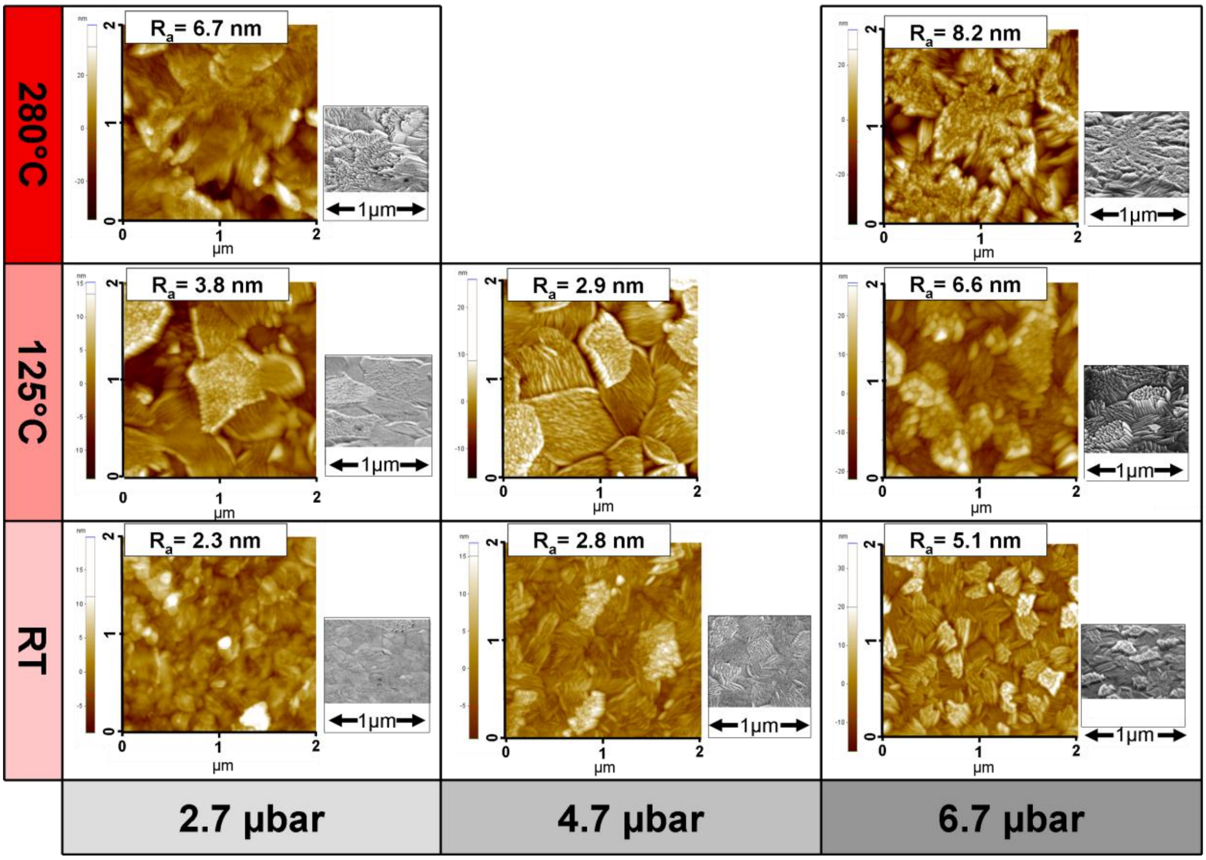

Fig.3. AFM (image size 2x2µm), SEM images and values of average roughness R_a of surface of ITO layers deposited at argon pressures ranging from 2.7 to 6.7 µbar and substrate temperatures from room temperature (RT) to 280°C.

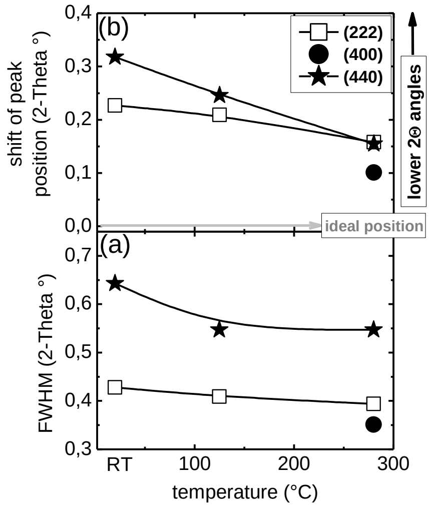

Fig.4. a) Values of FWHM and b) position of (222), (400), (440) diffraction peaks of ITO layers deposited at 2.7 µbar and substrate temperatures ranging from room temperature (RT) to 280°C.

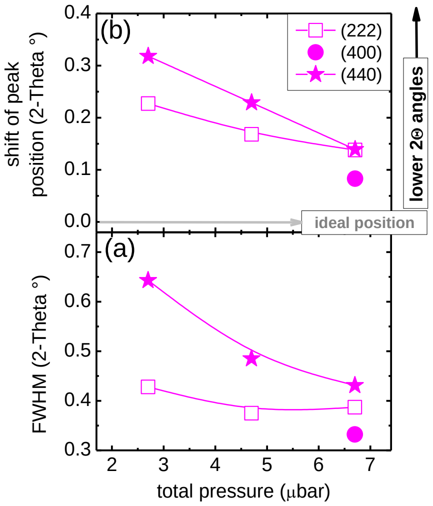

Fig.5. a) Values of FWHM and b) position of (222), (400), (440) diffraction peaks of ITO layers deposited at RT and at pressures ranging from 2.7 to 6.7 µbar.

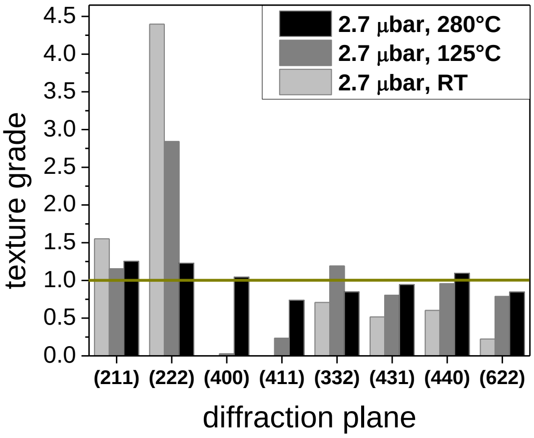

Fig.6. Texture grade of ITO layers deposited at 2.7 µbar and substrate temperatures ranging from room temperature (RT) to 280°C

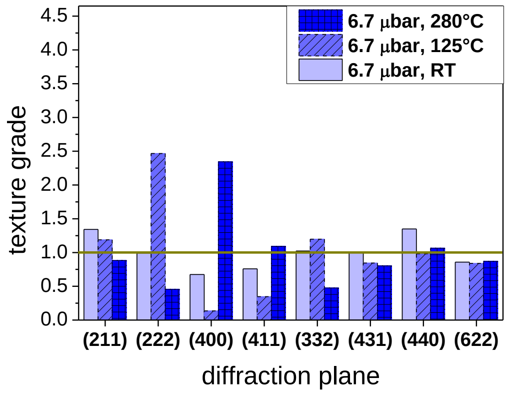

Fig.7. Texture grade of ITO layers deposited at 6.7 µbar and substrate temperatures ranging from room temperature (RT) to 280°C.

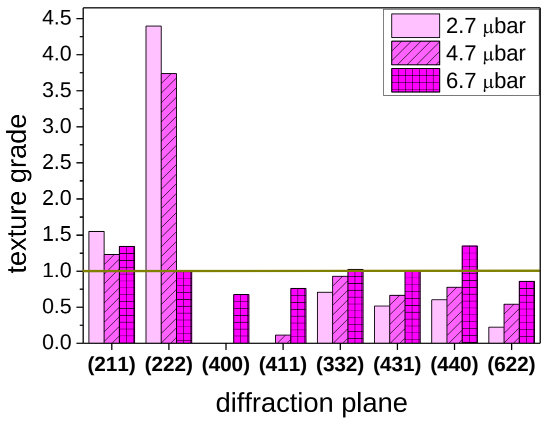

Fig.8. Texture grade of ITO layers deposited at RT and pressures ranging from 2.7 to 6.7 µbar.

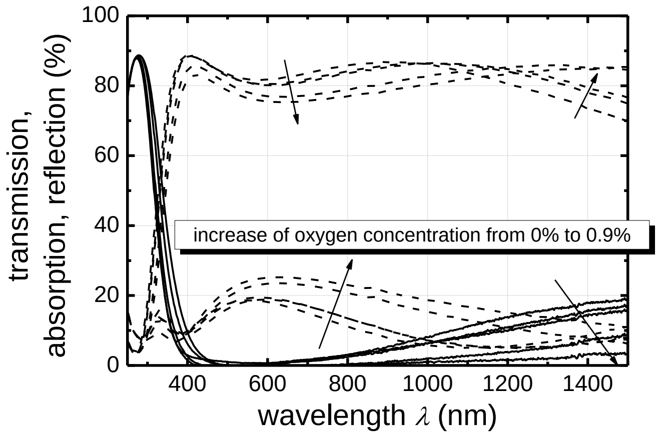

Fig.9. Optical properties of RF sputtered 80 nm thick ITO layers deposited at room temperature.

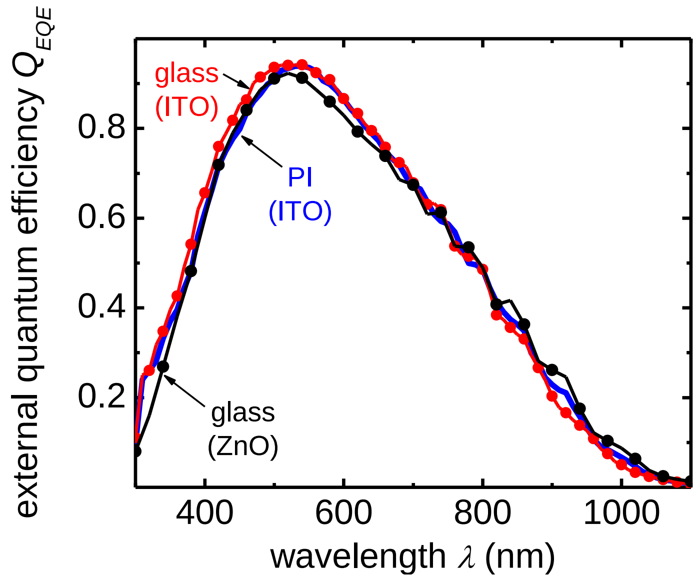

Fig. 10. EQE of µc-Si:H solar cells on glass (red line) and polyimide (PI) substrate (blue line) with ITO front contacts deposited at 125°C and room temperature, respectively. For comparison, EQE of µc-Si:H solar cell on glass (black line) with ZnO front contact is shown.