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Room temperature deposition of homogeneous, highly transparent and conductive Al-doped ZnO films by reactive high power impulse magnetron sputtering

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Abstract

Aluminium doped zinc oxide (AZO) films have been deposited using reactive high power impulse magnetron sputtering (HiPIMS) and reactive direct current (DC) magnetron sputtering from an alloyed target without thermal assistance. These films have been compared in terms of their optical, electrical and structural properties. While both DC and HiPIMS deposited films show comparable transmittance, their electrical properties are significantly improved by the HiPIMS process. The HiPIMS deposited films show a low resistivity down to the order of $10^{-4} \Omega\text{cm}$ with a good homogeneity across the substrate, making them potential candidates for electrodes in solar cells. The density of electrons reached up to $11 \times 10^{20} \text{ cm}^{-3}$, making ionized impurities the main scattering defects. This improvement of the film properties can be related to the specific plasma/target interactions in a HiPIMS discharge. This allows the process to take place in the transition mode and to deposit highly conductive, transparent AZO films on large surfaces at low temperature. While the overall oxygen content is above that of stoichiometric ZnO, higher localization of oxygen is found at the interfaces between crystalline domains with substoichiometric composition.

Keywords: Transparent conducting oxide, AZO, Thin films, Electronic properties, HiPIMS

1. Introduction

Thin films of aluminium doped zinc oxide (AZO) are highly interesting as transparent electrodes in applications such as photovoltaics [1] and flat panel

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displays [2]. For these applications a low resistivity of the AZO films in the order of $10^{-4} \Omega\text{cm}$ is required. An increasingly important parameter in the fabrication of solar cells is the deposition temperature. For copper indium gallium selenide (CIGS) solar cells a substrate temperature of less than 200°C is preferred in order to preserve the performance of the absorber layer [3]. Another interest is to deposit such materials on flexible polymer substrates in order to meet the demand of flexible solar cells. For such substrates, low temperature fabrication methods are also needed and synthesis routes avoiding thermal assistance would be ideal in order to simplify the fabrication process. The same applies to the development of flexible electronics. A common technique to deposit AZO films is reactive direct current (DC) magnetron sputtering because of its scalability [4]. But at low deposition temperatures it is difficult to achieve the required properties using reactive magnetron sputtering [4]. Another issue is the homogeneity of the electrical properties. Weltzel and Ellmer reviewed this problem [5] that can be traced back to the initial report by Minami et al. in 1982 that the resistivity of the deposited films increases in front of the target axis [6]. This degradation has been related to the bombardment with negative oxygen ions that introduces point defects such as zinc vacancies and oxygen interstitials that can compensate the donors [7, 8]. It is worth noting that such bombardment could cause damages to any materials in contact with the zinc oxide layer and might be the reason why CdSe buffer layers are required between the absorber layer and the insulating zinc oxide layer (i-ZnO) covered by the AZO transparent electrode of CIGS solar cells [3]. Another possible reason is the change in the local environment of the Al atoms that can lead to an deactivation of the dopants which is more pronounced in more resistive films [9, 10]. An $\text{Al}_2\text{O}_3(\text{ZnO})_m$ homologous phase can be formed where the Al atoms are in an octahedral configuration and do not contribute to the conductivity.

To improve the properties of the films without heating the substrate, high power impulse magnetron sputtering (HiPIMS) could be used [11, 12]. HiPIMS uses short pulses of high current density, that lead to a denser plasma and a large ionized fraction of the sputtered material [13]. The dynamics of the HiPIMS process leads to several effects such as self-sputtering [14], strong gas compression followed by rarefaction [15] and regions of strong ionization (spokes) [16]. It has also been shown that the hysteresis effect can be reduced or completely suppressed using a reactive HiPIMS process [17, 18].

In the present work AZO films were deposited without thermal assistance using reactive HiPIMS from a metallic Zn/Al target. Their optical, electrical and structural properties were investigated as a function of the position of the substrate during the deposition in order to evaluate the homogeneity of the process. The properties were also compared to the properties of a sample deposited by DC sputtering with similar average current. In a second part, the HiPIMS process was investigated, to explain the difference in the properties of the films between the different deposition methods.

Figure 1: Schematic representation of the sputtering chamber. The inset shows the position of the sample in the chamber.

2. Experimental

AZO films were grown without thermal assistance on glass substrates by reactive HiPIMS from a $\text{Zn}_{0.97}\text{Al}_{0.03}$ target. The target had a diameter of 5 cm and the distance between substrate and target was 8 cm. A schematic representation of the sputtering chamber is shown in figure 1. The gas flow rate was fixed at 80 sccm for argon and between 0 and 20 sccm for oxygen. This led to a total pressure of 1 Pa for the highest oxygen flow rate. The substrates were placed on a rotating substrate holder to ensure a homogeneous deposition. The magnetron was placed eccentric to the rotation axis of the substrate holder at a horizontal distance of 8 cm in order to reach good lateral homogeneity of the chemical composition [19].

The HiPIMS pulses were supplied by a Melec Spik 2000A that was connected to an Advanced Energy Pinnacle Plus DC power supply. The discharge current waveforms were recorded using a Pearson Current Monitor model 110 connected to a Tektronix TDS 2024B oscilloscope. For most of the experiments, the pulse length was 100 μs with a frequency of 1000 Hz. This corresponds to a duty cycle of 10 %. Depositions were done at discharge voltages of 540 V, 555 V and 570 V for 15 minutes. The corresponding discharge current waveforms are shown in figure 2. The peak current increased with the discharge voltage from 2 A at 540 V to 3 A at 570 V. This corresponded to an average current of about 200 mA. Some experiments were also performed at 570 V, using the same duty cycle but pulse lengths of 80 and 120 μs . The average current ranged from 0.18 A to 0.26 A as a function of the voltage and pulse lengths. For comparison, another sample was fabricated using DC magnetron sputtering with a current of 0.2 A and a corresponding voltage of 335 V. The oxygen flow rate was set to 8 sccm to adjust to the lower sputtering power of 67 W. The substrate temperature does not exceed 45 $^{\circ}\text{C}$ during the deposition process as measured by thermal level stripes (Thermax).

The optical transmittance of the films was measured in the ultraviolet, visible and near infrared ranges using a Cary 5000 UV-Vis-NIR spectrophotometer. The resistivity of the films was measured using a 4-point probe setup. The mobility and charge carrier density were measured using a HMS 5000 Hall effect measurement setup. Photoluminescence (PL) measurements have been performed with an excitation wavelength of 266 nm using a CryLas FQCW266-50 laser. The spectra were recorded using a Horiba iHR320 spectrometer equipped with a Horiba Sincerity multichannel CCD detector. The structural properties

Figure 2: Discharge current waveforms for three HiPIMS deposited samples using different discharge voltages with an oxygen flow rate of 20 sccm

were measured using $\theta/2\theta$ x-ray diffraction (XRD) with an AXS Bruker D8 Advance diffractometer with a Cu $K\alpha = 0.154$ nm) and high resolution transmission electron microscopy (HRTEM) with a JEM-ARM200F cold FEG Cs probe corrected microscope (point resolution 0.19 nm in TEM mode and 0.078 nm in STEM mode). The TEM samples were prepared by a focussed ion beam (FIB) / scanning electron microscope (SEM) dual beam system (FEI Strata DB235). The oxygen content, characterized by O/(Zn+O) atomic ratio was measured using Rutherford backscattering spectrometry (RBS). RBS measurements have been performed with a 1.5 MeV $^4\text{He}^+$ beam produced by the Van de Graaf accelerator of the ACACIA platform at the ICube Laboratory. Special attention was paid to the charge collection during the analysis because of the non-conductive behaviour of the glass substrates. A thin gold layer (2 nm) was deposited on all the samples together. The RBS contribution of this layer was used to check the efficiency of the ion beam current measurement. This thin layer does not alter the accuracy of the measurements.

3. Results and discussion

3.1. Optical, electrical and structural properties

The optical and electrical properties of the AZO films were investigated as a function of the position on the sample as shown in figure 1. The corresponding extracted values for the optical bandgap and resistivity along with the film thickness are reported in table S1 in the supplementary material. The transmittance of the samples was measured between 200 nm and 3300 nm. The obtained spectra for the sample position of 4 cm are shown in figure 3. The average transmittance in the visible range is generally above 83% for all the films, but slightly higher in the case of the DC deposited film. For the HiPIMS deposited samples the transmittance close to the absorption edge is slightly decreasing with increasing discharge voltage. A decrease in the transmittance may indicate a small oxygen substoichiometry [20]. In the near infrared, the transmittance

Figure 3: Transmittance curves for different AZO films

decreases going from DC to HiPIMS and it decreases further with increasing discharge voltage. This decrease in transmittance is related to the increase of the bulk plasmon frequency that shifts the absorption by free charge carriers towards the visible range as the density of charge carriers increases [21]. This tunable optical absorption could be used in a future generation of solar cells combining conducting oxides and metals of various specific absorption in order to match the solar spectrum [22]. The HiPIMS deposited films therefore have a higher charge carrier concentration of up to $11 \times 10^{20} \text{ cm}^{-3}$, as measured by Hall effect. The mobility of the films was measured in a range of $4 \text{ cm}^2/\text{Vs}$ to $11 \text{ cm}^2/\text{Vs}$ for the sample deposited at 570 V. The other Hall effect measurement results can be found in table S2 in the supplementary material. For the DC sputtered films the charge carrier concentration could not be measured with the Hall effect setup due to the higher resistivity of these films.

Another way to obtain information about the charge carrier concentration is the shift in the band gap due to the Burstein-Moss effect [23] that links an increase in the carrier concentration to an increase of the optical band gap. The band gap can be calculated from the transmittance data in the UV and visible region using the Tauc method [24]. The calculated values for the band gap are shown in figure 4a. Consistently with the conclusions drawn from measurements in the near infrared, switching from the DC to the HiPIMS process, the band gap generally increases. For the HiPIMS samples it increases with the discharge voltage. The band gap also varies with the position and reaches a minimum at the position facing the center of the target.

The band gap shift can be compared to the resistivity of the samples shown in figure 4b. The evolution of the resistivity is inverse to the evolution of the band gap, showing a good agreement with the Burstein-Moss effect. The resistivity is decreasing going from DC to HiPIMS. It is further decreased by increasing the discharge voltage. Also, the homogeneity of the resistivity is improved for the HiPIMS samples. For the DC sample there is a factor of about 2000 between the minimum and maximum resistivity, whereas the factor is only of about 3 for the sample deposited by HiPIMS at 570 V. The maximum of the resistivity is

Figure 4: Band gap (a) and resistivity (b) of the different AZO films as a function of the position on the sample surface

usually at the position facing the center of the target consistently with previous reports [9, 20]. Overall, the sample deposited at 570 V shows the best electrical properties with a minimum resistivity as low as $4 \times 10^{-4} \Omega\text{cm}$. Although a low resistivity was reported off-axis in transparent DC sputtered AZO films grown without thermal assistance, only highly resistive films could be synthesized in front of the target [25]. Hence, reactive HiPIMS represents an advantageous method to produce simultaneously highly transparent and conductive AZO films on large surfaces without thermal assistance.

To obtain information about the point defects present in the films, room temperature photoluminescence measurements were performed on a HiPIMS deposited AZO film. The resulting spectra are shown in figure 5 for two different sample positions. For both positions a near band edge emission is observed in the ultraviolet range with a maximum around 360 nm. In the visible range, point defects can cause a broad emission band [26]. In the present case this emission band has a much lower intensity as compared to the near band edge emission. This suggests a small point defect density in the film. Although this is not the main result of the present study, it is worth noting that such AZO films could be used in applications requiring UV emission since the band edge is significantly shifted into the UV due to the large density of electrons.

In order to track a possible influence from the microstructure on the film properties, the AZO films have been characterized using XRD. In figure 6a XRD patterns are shown for a DC and a HiPIMS deposited sample for the three different discharge voltages at a position of 4 cm. The XRD patterns

Figure 5: Photoluminescence spectra of a HiPIMS deposited AZO film for different positions

are normalized with relation to the film thickness. The full XRD patterns in logarithmic scale are shown in figure S2 in the supplementary material. All the films show an intense peak at $2\theta = 34.3^\circ$, that corresponds to the $\{002\}$ planes of ZnO. This means that the films have a preferential growth along the c-axis of the wurtzite cell of ZnO. The $\{002\}$ peak is most intense for the DC sample and the HiPIMS samples at the lower discharge voltages. The $\{002\}$ peak is less intense for the sample deposited with HiPIMS at 570 V although it exhibits the lowest electrical resistivity among the four films. For the HiPIMS deposited films at 555 V and 570 V there is an additional weak peak at $2\theta = 36.3^\circ$. This peak can be ascribed to the ZnO $\{101\}$ planes.

From the $\{002\}$ peak of the films, the size of the coherent scattering domains perpendicular to the sample surface can be estimated using the Scherrer equation $D = 0.9\lambda/\beta \cos \theta$ with β being the width of the peak corrected for the instrumental peak broadening [27]. The evolution of the coherence length is shown in figure 6b as a function of the sample position. In case of the DC deposited film, the coherence length increases in front of the target. It is nearly stable for samples deposited by HiPIMS at 540 and 555 V and decreases in front of the target for the films deposited at 570 V.

From the position of the $\{002\}$ peak, the lattice parameter c can be calculated. It is shown in figure 6c for the different samples and the different positions. In the case of the HiPIMS samples, the lattice parameter is larger in front of the target. In contrast, for the DC sample the lattice parameter is smaller in front of the target.

In order to further analyse the microstructure for the sample with the best electrical properties, HRTEM images have been collected for the sample deposited using HiPIMS at 570 V at a position of 8 cm. Figures 7a and b show the V-shape columnar structure of the film in bright field and dark field, respectively. This, along with the presence of a preferred orientation, is consistent with growth in the zone II or T if we refer to the structure zone model of Thornton [28] or Mahieu et al. [29], respectively. This indicates that unlimited surface diffusion occurred during growth [29]. The micrographs also show that the

Figure 6: (a) XRD patterns of AZO thin films deposited with DC and HiPIMS. (b) Coherence length of scattering domains calculated using the Scherrer equation from the ZnO {002} peaks. (c) The lattice parameter c for the different samples as a function of the position. The horizontal line marks the bulk lattice parameter of ZnO.

Figure 7: (a) Bright field TEM image of the AZO thin film deposited by HiPIMS at 570 V. (b) Dark field image of the same sample using the {002} diffraction spot. (c) HRTEM image of the same sample. The inset shows the FFT corresponding to the framed area.

columns have a width of about 20 nm. This value is similar to the length of the coherence domains along the columns calculated from the XRD measurements. Figure 7c shows a HRTEM image of a single column. The inset in figure 7c shows a FFT of a selected area. The column is oriented in the $\langle 001 \rangle$ direction, confirming the XRD results.

The results of the structural characterization show that the crystalline quality is better for the DC sputtered films than for the HiPIMS deposited films. The HiPIMS deposited films, however, have a lower resistivity than the DC sputtered films. Moreover, the sample with the best electrical properties exhibits the lowest crystalline quality as can be seen in figure 6a. This suggests that the scattering mechanisms are to a large extent governed by the ionized impurity scattering and little by the grain boundary scattering. This is in line with recent conclusions concerning AZO films with high carrier concentrations [30]. This is also confirmed by the distribution of the coherence length shown in figure 6b. The coherence length is varying for both DC and HiPIMS deposited films with a similar amplitude of variation, whereas the change in the electrical properties is much more pronounced for the DC sputtered film. The differences in the microstructure can therefore not explain the differences in the electrical properties between HiPIMS and DC deposited films.

The inhomogeneity of the DC sputtered films can be explained by the bombardment with negative oxygen ions during the film growth. As commented in the introduction, oxygen bombardment is effectively believed to lead to the deactivation of the Al dopant [9] and to cause the creation of oxygen interstitials and zinc vacancies that compensate the donor defects [7, 26]. The negative oxygen ions originate from the oxidized target surface and are accelerated across the target sheath [31]. The homogeneity of the HiPIMS deposited films suggests that there is less negative oxygen bombardment during the film growth using HiPIMS.

The slightly lower transmittance in the visible range close to the band edge for the HiPIMS deposited samples suggests that these films are slightly substoichiometric. This is also confirmed by the presence of the $\{101\}$ peak in the case of the sample deposited at 570 V. The $\{101\}$ peak has been found to appear in the case of substoichiometric films by Horwat and Billard [25]. The chemical composition has therefore been checked by RBS. The resulting spectra are shown in figure S4 in the supplementary material and the results are summarized in table 1. The oxygen concentration decreases going from DC to HiPIMS and with increasing voltage in the case of HiPIMS. The absolute value of the oxygen concentration is larger than 50 %. However, EDS profiles obtained within the TEM (shown in figure 8) evidence a fluctuation of the oxygen content with higher localization at the interface between crystalline domains. This could be due to the incorporation of molecular oxygen as found using electronic structure measurements at the conduction band of transparent ZnO films [32]. The minimum oxygen concentration is the same at the core of all measured domains and is close to 48 at%, i.e. well below that for stoichiometric ZnO. Considering that grain boundary scattering is not dominating the electrical behaviour, the substoichiometric grains play a major role on the film resistivity. To deposit

Table 1: Results of RBS measurements		
	Position (cm)	O/(Zn+O)
DC	5	0.545
	8	0.532
HiPIMS 540V	5	0.535
	8	0.541
HiPIMS 555V	5	0.535
	8	0.537
HiPIMS 570V	5	0.531
	8	0.529

Figure 8: TEM image and the corresponding oxygen concentration profile measured by EDS for an AZO film deposited by HiPIMS at a position of 8 cm

substoichiometric films it is necessary to sputter in the transition mode. This suggests that in the case of HiPIMS the sputter process takes place in the transition mode, meaning that the target is not fully oxidized. Less target oxidation would also lead to a lower oxygen bombardment as described in the previous paragraph.

3.2. Relation between film properties and target surface state

The reason for the lower target oxidation in the case of HiPIMS could be a partial removal of the oxide layer during a pulse or less presence of oxygen

Figure 9: Discharge current waveforms for different pulse lengths of 80, 100 and 120 μs . The inset shows the corresponding total ion dose per pulse.

near the target due to the rarefaction of the gas phase commonly observed in HiPIMS experiments [15]. As short pulses and relatively low discharge currents are used in the present study in order to maintain the target well below its melting point, we can discard a strong influence of the rarefaction effect on the surface oxidation of the target. Hence, partial removal of the oxide layer appears the most likely phenomenon. This partial removal of the oxide layer during the pulse was also proposed by Ganesan et al. for reactive HiPIMS deposition of HfO_2 and NbO_2 [33, 34]. They observed that a higher pulse length leads to substoichiometric oxide films. To check the influence of the pulse length, AZO films were deposited with a pulse length of 80, 100 and 120 μs while keeping the duty cycle constant at 10%. With a constant voltage of 570 V the average power was about 150 W. The discharge current waveforms are shown in figure 9. The inset shows that the total ion dose per pulse increases with increasing pulse length.

The band gap and the resistivity as a function of the pulse length for the different sample positions is shown in figure 10. The thickness, resistivity and band gap of these samples are also summarized in table S3 in the supplementary material and the transmittance spectra are shown in figure S1. The band gap is relatively high for all positions and independent of the pulse length. The resistivity is slightly higher than the resistivity of the samples from section 3.1, because a different target was used. The resistivity is still quite homogeneous over the sample surface and for all the pulse lengths. Hence, the pulse length has only a small influence on the electrical properties within the explored synthesis conditions. However, there is a difference in the absorption spectra, that are shown in figure 11 for two different positions on the sample. In figure 11a the absorption in front of the target axis is shown. The absorption spectra are relatively similar for this position. For the spectra measured off the target axis shown in figure 11b the films are less absorbent than in front of the target. This means that the films are substoichiometric in front of the target. Off the target axis, however, the films are more absorbent with increasing pulse length. A similar conclusion can also be drawn from the XRD patterns that are shown

Figure 10: Band gap (a) and resistivity (b) of AZO films deposited with different pulse lengths

Figure 11: Absorption spectra of AZO films deposited using HiPIMS with different pulse lengths facing the target axis (a) and off axis (b)

in figure S3 in the supplementary material. The films deposited using higher pulse lengths show a lower crystalline quality. This suggests that these films are also increasingly substoichiometric with increasing pulse length and therefore increasing ion dose. This supports the hypothesis of partial removal of the oxide layer.

The validity of this hypothesis can be tested by calculating the number of

Table 2: Surface atom density for different families of planes in ZnO calculated from the lattice parameters according to PDF card 04-003-2106

Family of planes	Surface atom density (10^{14} cm^{-2})
{001}	10.9
{110}	6.8
{100}	5.9
{101}	5.2

sputtered atoms during a single pulse and to compare it with the surface density of atoms in ZnO. Our calculation (see section 4 in the supplementary material) leads to a maximal number of sputtered atoms per surface unit of $7.2 \times 10^{14} \text{ cm}^{-2}$ during a pulse. This number can be compared to the surface atom density of ZnO which is given in table 2 for different planes. Considering that the target is polycrystalline or even amorphous due to the ion bombardment, an average surface atom density in the order of $8 \times 10^{14} \text{ cm}^{-2}$ can be assumed. This number is in the same order of magnitude as the number of sputtered atoms. This suggests that it is possible to remove an oxide monolayer or at least reduce the oxide coverage at the target surface in the centre of the racetrack and therefore sputter in the transition between the metallic and compound sputtering modes.

This is also confirmed when comparing the deposition rate for the HiPIMS and the DC processes. The deposition rate normalized by the sputtering power was about 0.15 nm/min/W for the HiPIMS deposited sample and about 0.08 nm/min/W for the DC deposited sample. This suggests, that with HiPIMS, the films are deposited in the transition mode.

In reactive DC sputter deposition of AZO films, small changes in the oxygen partial pressure for a fixed discharge current induce a drastic change of the film properties from absorbent and highly conductive to transparent and insulating [20, 25]. This change of properties occurs within a narrow range of oxygen partial pressures within the transition from the metallic to the compound sputtering mode. In HiPIMS, the transition is not observed due to the strong increase of the discharge current as the oxygen flow rate is increased. Hence, increasing the oxygen flow rate induces a very strong rise of the ion dose to the target. This increases the sputtering rate that can remove the oxide layer formed between pulses and hinder the growth of a new oxide layer during the pulse. A detailed discussion between the electrical properties of the two types of discharges is given in section 5 of the supplementary material.

4. Conclusion

Thin films of AZO have been deposited using both reactive DC magnetron sputtering and reactive HiPIMS without thermal assistance. The HiPIMS deposited films show better, and more homogeneous electrical properties than the

DC deposited films, while keeping similar transmittance in the visible range. However, the crystalline quality decreases for the HiPIMS deposited samples. This is due to the main scattering mechanism of electrons by ionized impurities at the very high values of carrier density reached.

A model was suggested in which the oxide layer on the target is partially removed during the pulse on-time. This way, the sputtering process takes place in the transition mode, which can decrease the oxygen content in the films and lead to lower oxygen bombardment during film growth, therefore improving the film properties without the need for substrate heating. Indeed, the oxygen content was found to be substoichiometric at the core of crystalline grains in contrast to interfaces where higher localization of oxygen is observed.

In conclusion, HiPIMS can be an alternative deposition method in the fabrication of transparent electrodes for solar cells because of the good and homogeneous electrical properties, that can be achieved at room temperature.

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Supplementary material

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Room temperature deposition of homogeneous, highly transparent and conductive Al-doped ZnO films by reactive high power impulse magnetron sputtering

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1. Optical and electrical properties

The thickness, resistivity and band gap for four different AZO films deposited using both DC and HiPIMS at different discharge voltages is shown in table S1. The thickness of the films was measured using a Taylor Hobson Talysurf 10 surface profiler. To measure the resistivity of the films, a 4-point probe setup was used. The band gap was calculated using the Tauc method from the optical transmittance spectra. The results are shown as a function of the position on the sample surface with relation to the rotation axis of the substrate holder. Table S2 shows the electrical properties of the HiPIMS deposited sample with a discharge voltage of 570 V measured using a HMS 5000 Hall effect measurement setup. In table S3 the thickness, resistivity and band gap of the AZO films deposited using HiPIMS with three different pulse lengths are shown. The transmittance spectra of these samples are shown in figure S1.

Table S1: Data of AZO films deposited by DC and HiPIMS

	Position (cm)	Thickness (nm)	Resistivity (Ωcm)	Band gap (eV)
DC	3	50	8.77×10^{-1}	3.34
	4	80	7.98×10^{-1}	3.32
	5	100	8.20×10^{-1}	3.32
	6	90	1.11×10^0	3.28
	7	160	5.44×10^1	3.27
	8	80	1.39×10^3	3.26
	9	130	5.13×10^2	3.26
HiPIMS 540V	3	240	1.34×10^{-2}	3.44
	4	330	4.79×10^{-3}	3.45
	5	300	2.79×10^{-3}	3.43
	6	280	5.95×10^{-3}	3.36
	7	320	2.39×10^{-1}	3.3
	8	320	3.41×10^0	3.27
	9	260	2.18×10^{-1}	3.3
HiPIMS 555V	3	270	4.28×10^{-3}	3.46
	4	320	1.94×10^{-3}	3.47
	5	320	1.12×10^{-3}	3.45
	6	300	1.73×10^{-3}	3.42
	7	330	6.88×10^{-3}	3.36
	8	320	1.52×10^{-2}	3.34
	9	260	5.47×10^{-3}	3.37
HiPIMS 570V	3	250	1.46×10^{-3}	3.51
	4	280	7.17×10^{-4}	3.51
	5	240	4.15×10^{-4}	3.5
	6	280	4.64×10^{-4}	3.48
	7	360	7.03×10^{-4}	3.46
	8	340	7.86×10^{-4}	3.45
	9	300	9.04×10^{-4}	3.46

Table S2: Hall effect measurement results of the AZO film deposited using HiPIMS at 570 V

Position (cm)	Resistivity (Ωcm)	Mobility (cm^2/Vs)	Charge carrier concentration (cm^{-3})
3	2.05×10^{-3}	4.09	7.47×10^{20}
4	7.50×10^{-4}	7.38	1.13×10^{21}
5	7.21×10^{-4}	10.5	8.24×10^{20}
6	8.17×10^{-4}	7.07	1.09×10^{21}
7	1.23×10^{-3}	8.84	5.76×10^{20}

Table S3: Data of AZO films deposited by HiPIMS with different pulse lengths

Peak length (μs)	Position (cm)	Thickness (nm)	Resistivity (Ωcm)	Band gap (eV)
80	3	320	1.53E-03	3.55
	4	320	1.57E-03	3.56
	5	320	1.85E-03	3.55
	6	310	1.99E-03	3.54
	7	300	2.21E-03	3.51
	8	325	2.61E-03	3.50
	9	350	3.04E-03	3.49
100	3	360	2.52E-03	3.46
	4	360	2.64E-03	3.50
	5	360	3.01E-03	3.52
	6	320	2.95E-03	3.51
	7	280	2.88E-03	3.49
	8	345	3.92E-03	3.47
	9	410	5.03E-03	3.46
120	3	240	1.51E-03	3.56
	4	260	1.72E-03	3.56
	5	280	2.08E-03	3.57
	6	300	2.52E-03	3.56
	7	320	2.99E-03	3.54
	8	330	3.35E-03	3.52
	9	340	3.72E-03	3.47

Figure S1: Transmittance spectra of the AZO films deposited using HiPIMS with different pulse lengths at a position of 3 cm

2. Structural properties

The structural properties of the AZO films were measured using $\theta/2\theta$ x-ray diffraction (XRD). The instrument used was a AXS Bruker D8 Advance diffractometer with a Cu anode ($\text{Cu K}\alpha = 0.154 \text{ nm}$). The XRD patterns of four different AZO films at a position of 4 cm are shown in figure S2 as a function of the discharge voltage. In figure S3 the XRD patterns of AZO films deposited by HiPIMS with different pulse lengths are shown. All the XRD patterns are normalized with relation to the film thickness.

Figure S2: XRD patterns of AZO films deposited with DC and HiPIMS at a position of 4 cm

Figure S3: XRD patterns of AZO thin films deposited using HiPIMS with different pulse lengths

3. Chemical composition

The chemical composition of a few selected samples was measured using Rutherford backscattering. The obtained RBS spectra are shown in figure S4a for the sample position at 8 cm. Figure S4b shows the RBS spectrum of the sample deposited at 570 V together with the corresponding simulation.

Figure S4: (a) RBS spectra of HiPIMS deposited AZO films at a position of 8 cm (b) Simulation of the RBS spectrum for the sample deposited with 570 V

4. Calculation of density of sputtered atoms per HiPIMS pulse

The number of sputtered atoms is given by the number of incident ions multiplied with the sputter yield. The number of incident ions during a pulse can be determined from the discharge current waveforms given in figure 2. The current from figure 2 contains both the electron and the ion current. The ion current can be calculated by taking the secondary electron yield γ of the target material into account using the equation $I_{\text{ion}} = I_{\text{total}}/(1 + \gamma)$. The secondary electron yield of AZO can be extrapolated from the data measured by Kim et al. [1] as $\gamma = 0.15$. Integrating over the ion current gives the total ion dose per pulse. The total ion dose is not distributed homogeneously over the target surface. It can be assumed that the local incident ion density is proportional to the target erosion profile. From the measured profile of the used target, the distribution shown in figure S5 can be calculated. These results show that the incident ion density in the center of the racetrack reaches 4.5×10^{14} ions/cm². Using the TRIM (Transport of ions in matter) software [2], the sputtering yield of ZnO can be estimated to 1.6 atoms/ion assuming incident Ar⁺ ions with a kinetic energy of 570 eV and a value 3.75 eV/atom for the surface binding energy of ZnO [3].

Figure S5: Local incident ion density on the target, calculated from the ion dose per pulse and the erosion profile of the target and the corresponding distribution of sputtered atoms assuming a sputtering yield of 1.6 atoms/ion

5. HiPIMS discharge parameters

To investigate the plasma parameters as a function of the oxygen flow rate in HiPIMS, discharge current waveforms were recorded using a discharge voltage of 570 V. The current waveforms are shown in figure S6a. The discharge current is increasing with the oxygen flow rate. The peak current for the highest flow rate is higher than the one shown in figure 2 because a different target with a slightly lower Al content of 2.3 % was used. The plasma impedance, as calculated from the peak current, changes from 950 Ω at 0 sccm oxygen flow rate to 130 Ω at 20 sccm. The normalized decrease of the impedance with the oxygen flow rate corresponding to the increase in the peak current is shown in figure S6b. For comparison, also the relative impedance change for a DC process with the same target with a current of 0.2 A is shown. In the case of HiPIMS the impedance decreases about 86 %. In the case of DC the impedance only decreases about 39 %. The plasma impedance decreases much stronger with the oxygen flow rate in case of HiPIMS than in the case of DC sputtering. Calculating the HiPIMS impedance using the average current instead of the peak current decreases the difference to the DC impedance only to a minor degree. This change in the plasma impedance can be explained due to the increase in secondary electron yield in the compound sputtering mode [4] and improved dissociation and ionization of molecular oxygen leading to increase in the discharge current [5]. In HiPIMS discharges Poolcharuansin and Bradley have detected fast electrons at the beginning of the pulses [6]. It has been speculated that these fast electrons can induce an increased dissociation and ionization of molecular oxygen and, thereby, induce an increase in the density of electrons in the plasma phase and an increase in the discharge current [5]. This could explain the larger decrease in the plasma impedance in the case of HiPIMS. As visible on figure 2, the discharge voltage has a similar effect on the plasma impedance. An increased flux of sputtered particles leads to a stronger compression and improved ionization in the gas phase by charge exchanges as the discharge voltage is increased. We have tested higher discharge voltages than 570 V, but it was not possible to synthesize transparent AZO films, because the plasma became unstable and arcing occurred.

Figure S6: (a) Discharge current waveforms as a function of oxygen flow rate for a discharge voltage of 570 V. (b) Normalized decrease of the impedance with the oxygen flow rate.

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