PAPER

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Single-beam ion source enhanced growth of transparent conductive thin films

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Abstract

A single-beam ion source was developed and used in combination with magnetron sputtering to modulate the film microstructure. The ion source emits a single beam of ions that interact with the deposited film and simultaneously enhances the magnetron discharge. The magnetron voltage can be adjusted over a wide range, from approximately 240 to 130 V, as the voltage of the ion source varies from 0 to 150 V, while the magnetron current increases accordingly. The low-voltage high-current magnetron discharge enables a 'soft sputtering mode', which is beneficial for thin-film growth. Indium tin oxide (ITO) thin films were deposited at room temperature using a combined single-beam ion source and magnetron sputtering. The ion beam resulted in the formation of polycrystalline ITO thin films with significantly reduced resistivity and surface roughness. Single-beam ion-source-enhanced magnetron sputtering has many potential applications in which low-temperature growth of thin films is required, such as coatings for organic solar cells.

Keywords: ion source, magnetron discharge, low temperature, thin film, high rate deposition

(Some figures may appear in colour only in the online journal)

1. Introduction

Indium tin oxide (ITO) thin films are the primary transparent conductive materials used in a broad variety of applications, such as solar cells, displays, smart windows, and LEDs [1]. ITO thin films are commonly produced by magnetron sputtering and require elevated substrate temperatures (e.g. >200 °C) to achieve satisfactory electric conductivity and optical transmittance. However, many applications involve heat-sensitive materials that limit the thin-film growth temperature. An example is ITO deposition on polyethylene terephthalate for touch screens, where this substrate material could only stand a temperature up to ∼80 °C. Under low substrate temperatures, ITO thin films have an amorphous microstructure, which leads to high resistivity, low optical transmittance, and poor stability [2]. Producing polycrystalline ITO thin films under the off-phase equilibrium temperature is a fundamental challenge.

Ion source assisted deposition has the potential to produce high-quality ITO films at low substrate temperatures. Ion sources are plasma generation devices that enable ion beams to interact with the materials at the atomic level as they

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are deposited to effectively produce dense films with tunable morphology and superior stability [3–5]. Two major types of ion sources have been widely used for surface treatment—filament and racetrack (anode layer) types [6]. Filament-type ion sources can produce ions with controllable energy over a wide range. Some processes require the use of reactive gases, such as oxygen, which could be detrimental to the filament. Racetrack-type ion sources are compatible with reactive gases. The closed-loop drift of the electrons leads to circular or racetrack beam patterns, while some applications would require the ions to be focused onto a small area. Furthermore, the ion sources used to enhance thin-film growth must be compatible with magnetron discharges and can stably operate over an extended period.

A single beam ion source has been recently developed to address the needs described above. This single beam ion source combines several desired features:

- Focused single beam of ions generated without a filament;
- Widely tunable discharge voltage (e.g. 0–250 V) for optimal ion-surface interactions;
- Wide range of operation pressure (0.13–60 Pa) compatible with magnetron sputtering and chemical vapor deposition in inert and reactive gases; and
- Hidden anode suitable for long-term operation in the thin-film manufacturing environment and easy to maintain.

This paper reports the initial study to validate the basic characteristics of the single beam ion source enhanced magnetron sputtering. The goal is to demonstrate the feasibility of low-temperature high-rate deposition of ITO thin films that have the desired microstructure and properties, which could only be obtained at elevated temperatures in conventional magnetron sputtering.

2. Material and methods

The single-beam ion source used in this study is illustrated in figure 1 (model SPR-10, Scion Plasma LLC). It consists of an anode with a center cavity and a closed bottom. A cathode cover with a center opening is located above the anode, which is not directly exposed to the atoms sputtered off the magnetron target. A magnetic field is generated by a magnet assembly and forms a closed loop inside the anode cavity to confine the electrons. Details of this single beam plasma source can be found in [7].

A circular magnetron (model TORUS TM3, KJ Lesker) was used for sputtering deposition of ITO thin films. The ITO target was 76.2 mm in diameter and ~3.2 mm in thickness. The target was 99.99% purity with a composition of $In_2O_3/SnO_2 = 90/10$ wt%. The ion source and the magnetron were arranged at an angle of 45° with their center lines crossing at the substrate surface (see details in section 3). The center-line distances from the ion source to the substrate and from the magnetron target to the substrate were ~100 mm.

Before the ITO film deposition, the system was pumped down to a base pressure below 1.3×10^{-4} Pa. Ar gas mixed with 0.1% oxygen was used in all the depositions. The process pressure was 0.4 Pa. All the ITO films were deposited at room temperature. The magnetron sputtering power was 60 W of pulse DC in all the depositions. The pulse frequency was 100 kHz (10 μs duty cycle) with 1 μs reverse time. However, the ITO film deposition rates strongly depended on and increased almost linearly with the voltage of the ion source. Therefore, the deposition time under each ion source voltage was adjusted to produce the ITO films of 100 nm thickness. The substrates were a soda-lime glass of 25 mm × 25 mm × 0.7 mm.

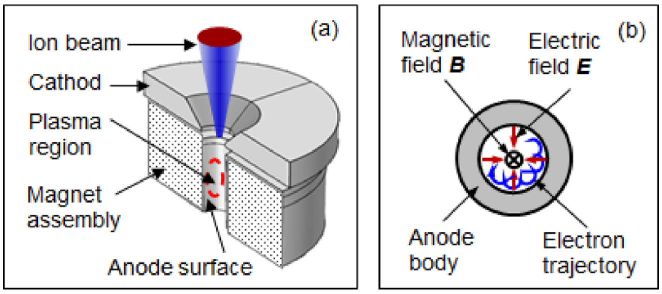

Figure 1. (a) Profile size view and (b) top view of the single beam plasma source.

The film thickness was measured using a Dektak 150 profilometer. The ITO film transmittance was characterized using a spectrophotometer (F20, Filmetics). The sheet resistance was measured using a four-point probe (SRM-232-1000, Guardian Manufacturing). The film microstructure was determined using x-ray diffraction (SmartLab, Rigaku). The surface morphology was characterized using atomic force microscopy (AFM5000, Hitachi).

3. Results

The sputtering magnetron and the single beam ion source could be arranged at any angle, such as parallel, 45°, and 90°. Although all these configurations showed similar discharge characteristics, the preferred arrangement is 45° or larger angle for focusing the beam on the substrate area. Figure 2 shows a typical discharge image of the magnetron operating simultaneously with the ion source. The magnetron was set at 45° from the ion source in this case. The magnetron and the ion source can be excited by various combinations of power sources. For example, the magnetron can be powered by DC, pulsed DC, or RF, while the ion source can be powered by DC, RF, or DC + RF. The discharges were stable in all these combinations. In consideration of the practical application, this study used pulse DC to power the magnetron and DC + RF to power the single beam ion source. The RF power was 0–10 W with a maximum peak–peak voltage of ~400 V.

When the single beam plasma source simultaneously operates with a sputtering magnetron, it can discharge over a wide range of voltages, as illustrated in the I–V curve in figure 2. The DC bias voltage could be varied from 0 V. The ion source current increased almost linearly with the voltage.

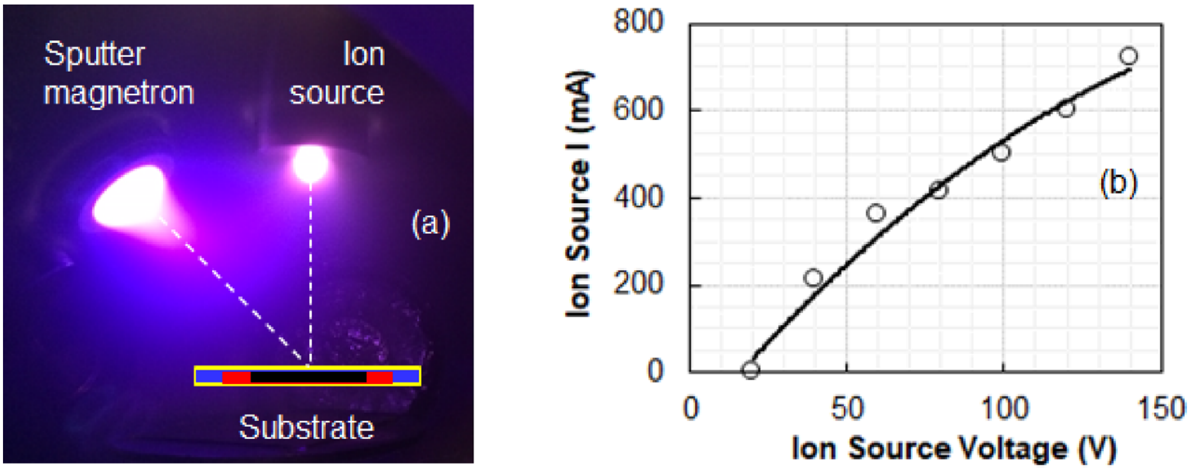

Figure 2. (a) Discharge image of the single beam plasma source operating with a sputtering magnetron. The black area in the substrate is the effectively treated region and the blue area is untreated, while the red area is a transition region partially treated. (b) I–V characteristics of the single beam plasma source operating with a magnetron power of 60 W.

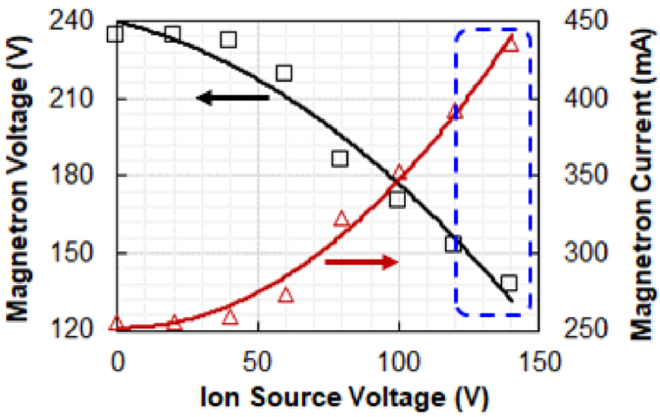

Figure 3. Variation of the magnetron current and voltage with the voltage of the ion source.

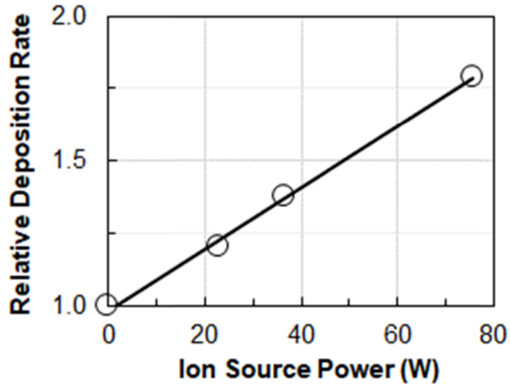

Figure 4. Relative deposition rate dependence on the ion source power. The magnetron power was fixed at 60 W. Ion source power of 0 W means magnetron sputtering alone.

The single-beam ion source could drastically modulate the magnetron discharge. As the voltage of the ion source increased from 0 to 140 V, the magnetron discharge voltage dropped from 234 V to 138 V, while the magnetron current increased accordingly from 256 mA to 435 mA as illustrated in figure 3. Hence, the beam plasma source enables a low-voltage and high-current sputtering mode, marked by the dotted blue line in figure 3. We call this 'soft sputtering mode'. In comparison with magnetron sputtering alone (0 V ion source voltage), the 'soft sputtering mode' led to higher deposition rates of ITO films. It is worth noting that the current of the single beam ion source became larger than the magnetron current at an ion source voltage above 40 V. This result indicates that the single beam ion source can 'amplify' the current from the magnetron discharge by creating additional electrons and ions. Therefore, the ion source and the magnetron mutually enhance each other.

An immediate effect of the ion source enhanced magnetron discharge is the increase of the film deposition rates. Figure 4 illustrates the relative ITO film deposition rates as a function of the ion source power. In this set of tests, the magnetron sputtering power was fixed at 60 W and the deposition rate with the magnetron alone was ~14 nm min^{-1}, which is taken as a relative rate of 1. It can be deduced that the ITO deposition rate increases ~1.6 times with 60 W ion source power. It is worth noting that the increased deposition rate was achieved at a low magnetron discharge voltage (e.g. ~150 V). This low voltage could not be attained in conventional magnetron sputtering, which would require higher power and thus higher voltage to increase the deposition rates. The reduced magnetron discharge voltage resulted in desired film properties as discussed below.

Figure 5 shows the sheet resistance of ITO films deposited at different ion source voltages with the magnetron power fixed at 60 W. The ITO film sheet resistance dropped from 118 to 56 Ω Sq^{-1} as the ion source voltage increased from 0 to 120 V. Further increasing the ion source voltage led to an increase in the ITO sheet resistance. Therefore, there is an optimum voltage that reflects the proper energy of the ions interacting with the ITO films as they were deposited. It is worth noting that the process conditions were not fully

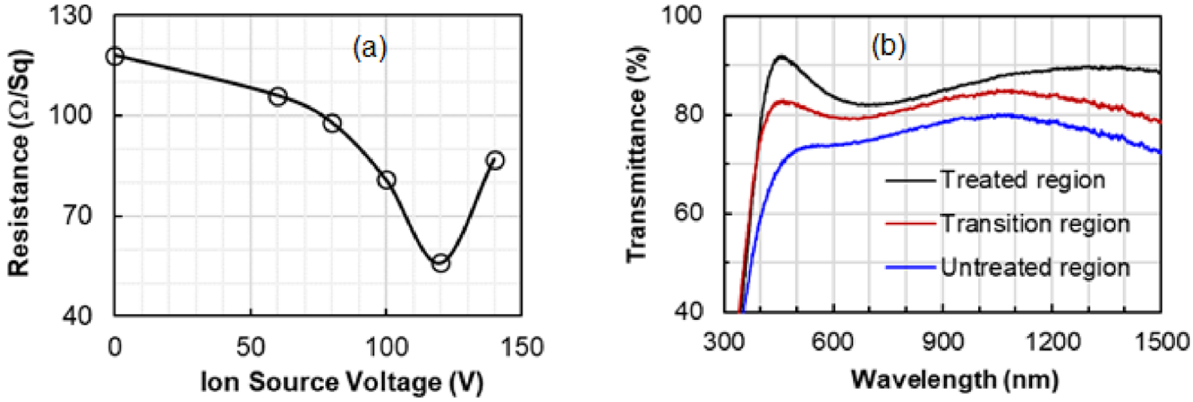

Figure 5. (a) Sheet resistance of 100 nm ITO films deposited at different beam plasma source voltages with the same magnetron power. (b) Optical transmittance spectra of ITO films in different regions of the same substrate under an ion source voltage of 100 V.

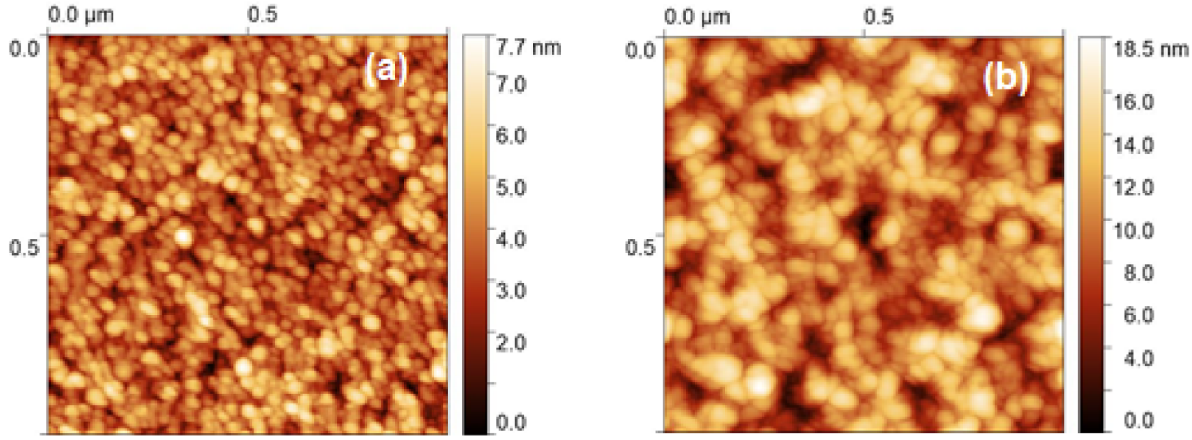

Figure 6. AFM images of ITO film deposited by magnetron sputtering with the ion source (a) on (100 V) and (b) off (0 V).

optimized. The results only show one typical set of process parameters (e.g. pressure, oxygen fraction, and magnetron power) with different ion source voltages.

The optical transmittance of the ITO film varies with the location on the substrate as shown in figure 5(b). The transmittance is the highest in the central region where the ion beam effectively interacts with the film. This center region was about 38 mm in diameter. Away from this region, the transmittance dropped greatly. Therefore, the ion beam 'amplified' the effect of oxygen by effectively delivering oxygen species to the film, while the oxygen concentration was still not sufficient without the ion beam treatment. It is worth noting that ITO films with transmittance close to the ion beam treated region could be made by magnetron sputtering alone if higher concentrations of oxygen were introduced. However, this is usually at the cost of increased resistivity.

The optical transmittance of the ITO films in the treated center region also varied with the ion source voltage. The transmittance spectra appeared to be similar once the ion source voltage was above ~80 V. Below 50 V ion source voltage, the film transmittance appeared to be similar to the one with magnetron sputtering alone.

Atomic force microscopy (AFM) scanning revealed distinct surface morphologies of the ITO films deposited with and without the ion source treatment, as illustrated in figure 6. The ion source led to a much smoother ITO film with a root mean square roughness of RMS = 1.09 nm, while the ITO film deposited with magnetron alone has a roughness of RMS = 2.97 nm.

The significant effects of the single beam ion source on the ITO film properties are closely related to the film microstructure. X-ray diffraction patterns shown in figure 7 indicate that the ion beam led to much improved ITO film crystallinity once the plasma source discharge voltage was above a threshold value (e.g. 100 V). Below this threshold (e.g. 0–80 V), the ITO films appeared to be amorphous. Interestingly, a plasma discharge voltage of 100 V resulted in preferentially oriented crystals, while higher voltages led to random orientations. These effects can be understood in terms of the ion energy delivered to the ITO films as the atoms were deposited on the substrate. There is optimum ion energy. It is worth noting that all the ITO films were deposited at room temperature, which is expected to yield an amorphous structure.

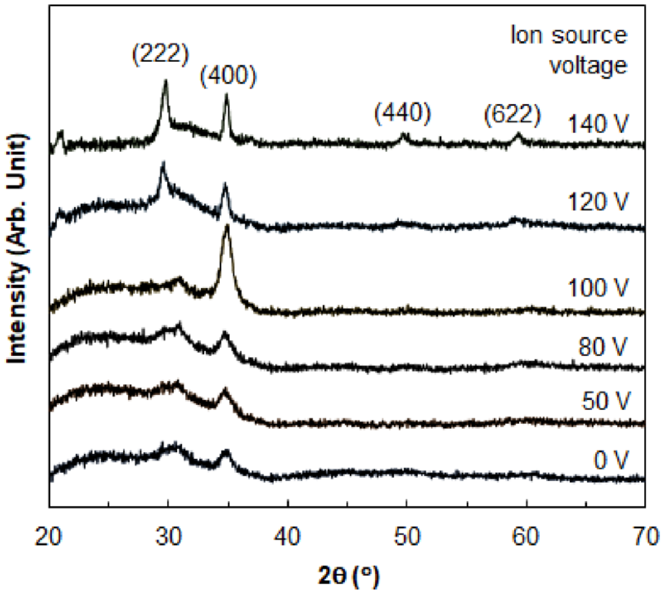

Figure 7. X-ray diffraction patterns of ITO films deposited at different beam plasma source voltages.

4. Discussion

4.1. Beam plasma source enhanced magnetron discharge

The discharge voltage is coupled with the current in conventional magnetron sputtering. This means the voltage and current would increase simultaneously with the excitation of DC power. On the other hand, it has been well recognized that RF sputtering generally results in better film quality than DC. One of the reasons is the DC bias on the cathode (target) during RF discharge is in the range of 100–140 V, which is much lower than the DC discharge voltage which is usually above 250 V. A low discharge voltage eliminates the high-energy tail of the sputtered atoms. According to Thompson's Law [8, 9], a fraction of the sputtered atoms has energy close to the magnetron discharge potential energy (e.g. 250 eV). These energetic atoms could induce disordered film microstructures and create rough surfaces.

Although a low sputtering voltage is generally preferred, RF sputtering and DC sputtering at low voltages suffer from low deposition rates and are practically difficult to be adopted into coatings manufacturing. The single-beam ion source enhances magnetron discharge, leading to low target voltage and high current sputtering that yields an even higher deposition rate than magnetron sputtering alone. This soft sputtering mode opens a new thin-film deposition regime that has many potential applications. The soft sputtering mode cannot be achieved in conventional DC magnetron discharges.

Previous research has indicated that there is a significant potential drop between the bulk plasma and the substrate in RF discharge, while this potential is negligible in DC magnetron sputtering [10, 11]. Therefore, pronounced ion bombardment to the substrate is expected in RF magnetron sputtering. This is another reason that RF sputtering generally produces better film quality than DC sputtering at low temperatures. However, the potential between the bulk plasma and substrate has little tunability in RF magnetron discharges. On the other hand, the single-beam ion source delivers ions with controllable energy to the film, while it enables a soft sputtering mode on the target side. Hence, it can modulate the film microstructure and properties even at practically high deposition rates and low temperatures.

4.2. Electron and ion energies

The energies of the electrons and ions created by the single beam plasma source depend on the excitation power source (DC and/or RF) and voltage, as well as the substrate being a conductor or insulator. Figure 8 shows the simulated electron energy probability function (EEPF) and ion energy distribution function (IEDF). The simulation was performed using an established particle-in-cell Monte Carlo collision code ASTRA. The detail of this simulation scheme is described in previous work [12]. The EEPF includes two regions: inside and outside the anode cavity of the single beam plasma source. The results indicate that higher RF peak voltage and DC bias lead to increased energetic tails that could enhance the plasma density of the magnetron discharge.

The IEDF shows that the ion energy is proportional to the RF and DC voltages on an electrically conductive substrate. Therefore, this result confirms the previous observation (see figure 4) that there is optimum ion energy that leads to the lowest sheet resistance of the ITO films. On the other hand, the ion energy is much reduced if the substrate is an insulator. In this case, the RF excitation plays an important role in modulating the ion energy.

4.3. Scalability of the single beam ion source

This study used a round-shape single-beam ion source that can effectively treat a substrate area about 38 mm in diameter when

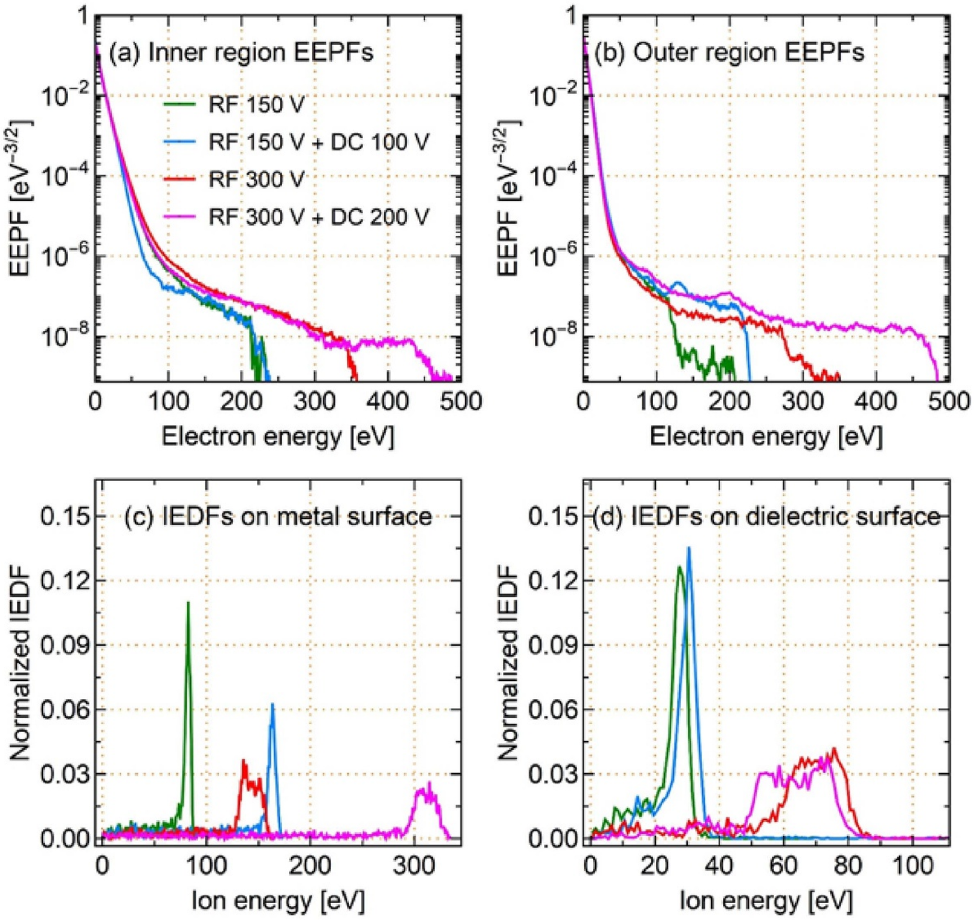

Figure 8. (a), (b) Electron energy probability function (EEPF) inside and outside of the anode cavity. (c), (d) Ion energy distribution function (IEDF) on substrates.

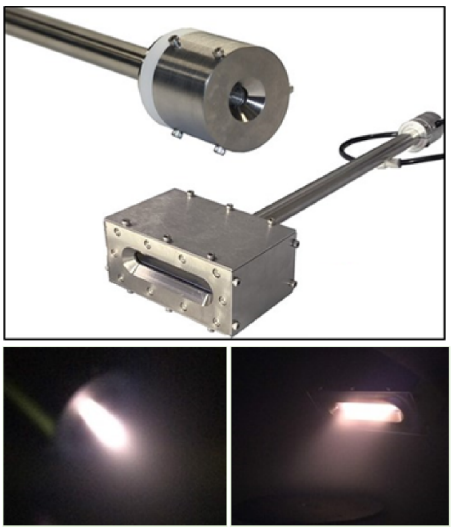

Figure 9. Round and linear (78 mm effective length) single beam plasma sources and discharges.

the source was set at about 76 mm away from the substrate. The single-beam plasma source can be designed into a linear structure of any custom length, as illustrated in figure 9, for treating larger areas. It can treat a rectangular area of about 38 mm wide and the effective length of the source.

5. Conclusion

This research demonstrates the use of a single beam ion source to enhance magnetron discharge and thin-film growth at low temperatures. The single-beam plasma source enables a soft sputtering mode that features low magnetron discharge voltage and high current, which can potentially produce high-quality thin films without sacrificing the deposition rates. The soft sputtering in combination with ion beam interactions with the film leads to high-rate deposition of ITO films with tunable microstructures. Polycrystalline ITO thin films can be produced at room temperature once the ion energy reaches a threshold value. The single-beam ion source enhanced sputtering leads to greatly reduced ITO film resistivity and surface roughness. The single-beam ion source is scalable to a linear structure of any length for large-area coatings.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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