

Article

Development of Sn Oxide Hetero-Junction Band Alignment via Oxygen Plasma Treatment Suitable for Photo-Sensing Applications

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Abstract

We report on results referring to the growth and characterization of Sn-oxide-semiconductor thin films (SnO₂, SnO and intermediate Sn₃O₄) on silicon substrates forming Type II heterojunction band alignment. The samples were produced by a two-step procedure: (a) growth of a metallic Sn layer by RF magnetron sputtering deposition followed by (b) post-growth treatment of the Sn films with oxygen plasma etching for the formation of the oxides in various time steps. Various annealing steps were considered. The structural and chemical properties of the prepared thin films were determined by means of X-Ray Diffraction (XRD) and X-Ray Photoelectron Spectroscopy (XPS). The results demonstrated the presence of SnO₂ and SnO in a tetragonal structure and the intermediate Sn₃O₄ in a triclinic structure. The electric properties of thin films were investigated with four-probe I–V characteristics under various conditions. The evaluation of their photo-sensing properties was performed by means of photocurrent J–t curves using a solar simulator. We found that the sample with equal concentrations of SnO₂ and SnO exhibited superior responsivity and detectivity values as well as a responsivity of 12.5 A/W and detectivity of 1.1×10^{10} Jones for V = 0 under yellow light illumination and an intensity of 2 mW/cm². These excellent values, in combination with the low-cost manufacturing, indicate that the method is promising for future applications.

Keywords: tin oxides (SnO₂/SnO/Sn₃O₄); RF magnetron sputtering; oxygen plasma treatment; electric properties; responsivity; detectivity(D*)



Academic Editors: Xue Yong, Jing Wang and Yuxiang Cai

Received: 15 June 2026

Revised: 11 July 2026

Accepted: 13 July 2026

Published: 14 July 2026

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1. Introduction

Tin oxides, including SnO and SnO₂ and intermediate oxides like Sn₃O₄ [1,2], have attracted scientific interest recently as research subjects due to their unique optoelectronic properties and cheap fabrication costs. Moreover, tin (IV) oxide (SnO₂), which is the most stable main oxide, is an n-type semiconductor characterized by a direct bandgap varying from 3.3 eV to 3.6 eV that exhibits excellent optical transparency [3–7]. In contrast, tin oxide (SnO) is a p-type semiconductor with a direct bandgap around 2.8–3.0 eV [8] and an indirect bandgap at 0.7 eV [6].

Numerous investigations have focused on them, especially SnO_2 , due to its properties as a Transparent Conducting Oxide (TCO) [9]. SnO_2 -based TCOs are widely used in optoelectronic applications due to their optical transparency [10–13]. Among these is ITO (indium–tin oxide), comprising $\text{In}_2\text{O}_3:\text{SnO}_2$, which is particularly prominent; it is an n-type semiconductor characterized by a bandgap ranging from 3.5 to 4.3 eV [10]. Despite the impressive performance of ITO, its relatively high cost has prompted the exploration for more economical alternatives, such as zinc–tin oxide (ZTO) and fluorine–tin oxide (FTO). ZTO presents a cost-effective option capable of supporting flexible device fabrication [14], while FTO serves as a lower-cost alternative to ITO, offering improved thermal stability, despite its relatively lower conductivity [15,16].

Nanostructures such as heterojunctions formed from SnO_2/SnO are promising for photocatalytic [17,18] and gas/photon-sensing applications [19–23] according to the literature. Given these advances, the potential utilization of these Sn oxide heterojunctions in solar cell applications is particularly noteworthy. The photoelectrical properties of Sn oxide heterojunctions are already under thorough investigation. A key approach to determining their utilization as photovoltaics is conducting research on their role as photo-response devices using various methods [24]. The findings suggest that these oxides could also be adequate for creating photodetection devices.

Besides SnO_2 and SnO , metastable intermediate oxide phases have been documented during the decomposition reaction of SnO ever since the first reports of the thermal instability of this compound were made. Several oxide compositions, including Sn_5O_6 , Sn_3O_4 , and Sn_2O_3 , have been referenced in the scientific literature [25].

Triclinic Sn_3O_4 is a good example of a non-stoichiometric structure. It was first identified by Lawson [26] as a result of the oxidation of another non-stoichiometric phase of SnO . It has two oxidation states, in which 1/3 of the Sn atoms are in an octahedral coordination with oxygens with an oxidation number of +4, while the remaining 2/3 are in a tetrahedral coordination with an oxidation number of +2. The energy gap of the intermediate Sn_3O_4 is 2.6 eV, which is an interesting characteristic for technological applications. According to the work of Berengue et al. [27], there are empty vacant oxygen positions, creating an additional energy state within the energy gap and lying 1.8 eV below the conduction band. Sn_3O_4 is an n-type semiconductor due to the new oxygen positions. It is an excellent material for photocatalysis due to the energy gap lying in the visible spectrum, and it has been successfully used as a photocatalyst for various dyes, including methylene blue, methyl orange, various phenols, etc. [25,28–30].

The band alignment of the two main Sn oxides forms a Type II heterojunction (a staggered gap) from the point of view of the transport of charge carriers [31]. In addition, the metastable oxide Sn_3O_4 can also form heterojunctions or other types of nanointerfaces with the main oxides. The proposed band alignment of $\text{SnO}_2/\text{SnO}/\text{Sn}_3\text{O}_4$ is schematically illustrated in Figure 1, as chosen from the literature values for a potential heterojunction. The schematic proposes that the multiple phases that form are dependent of each other. A decrease in the number of heterojunctions of the intermediate oxide is expected to positively affect the total results. The staggered-gap heterojunction offers better charge carrier separation compared to the other heterojunction types, resulting in bigger exciton lifetimes [32].

Sn oxides can be synthesized by utilizing multiple techniques for the fabrication of powder and thin films. The most common methods of heterojunction creation are single-step, such as sputtering [33–35], thermal evaporation [36], and chemical methods like hydrothermal synthesis [20,24,37] and sol–gel [7,38,39]. These usual production methods provide an increased opportunity for successful growth but cannot always be used to attain specific selective properties. In the present study, we suggest an attractive alternative

approach based on the utilization of a two-step growth process that contains a second post-growth step after deposition [40,41].

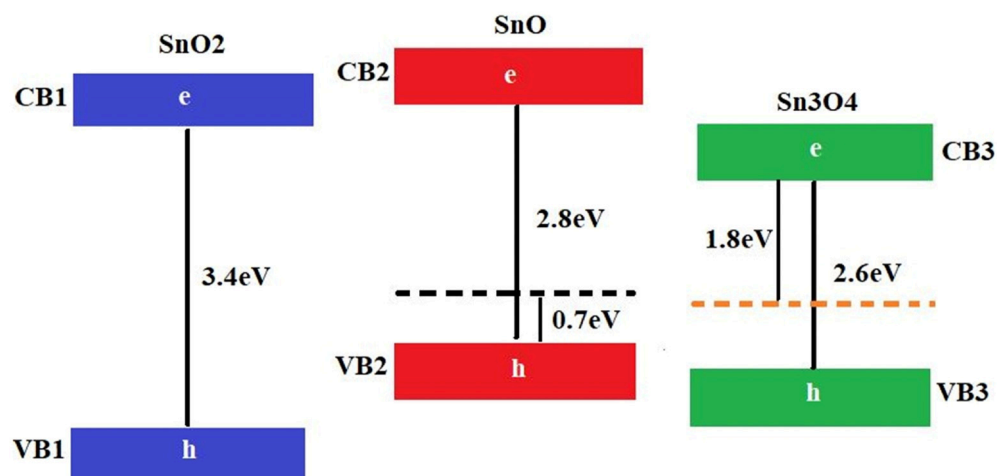


Figure 1. Schematic representation of band alignment in the Sn oxide (SnO_2 , SnO and Sn_3O_4) interfaces. Dashed lines stand for the additional states introduced by the oxygen vacant sites (orange line) and the indirect bandgap of SnO (black line).

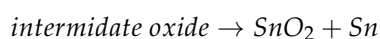
The objective of this study is to implement a method for the fabrication of Sn oxide heterojunctions through the plasma oxidation of metallic Sn sputtering targets. The two-step growth method involving thin film deposition and plasma oxidation allows for the broad parametrization of many characteristics, like the thickness of the thin film and the estimation of the phase's concentration of crystal structures in the oxides, as shown in similar works of Panagiotopoulos et al. [40,41]. This present study, based on the mentioned works, introduces an empiric relationship for the prediction of the second-step (plasma treatment process) phase concentrations and the total results in potential applications. Variations in oxidation times can lead to distinct crystal structures, including notable phases such as the intermediate oxide Sn_3O_4 , which can also play a role in the formation of heterojunctions [1,42–45]. Additionally, due to the relatively short plasma treatment durations, this method is economically advantageous while still creating high-quality final products.

2. Materials and Methods

2.1. Synthesis Process

The Sn thin films were synthesized using a two-step procedure: (a) the magnetron sputtering deposition technique was used for the growth of Sn thin films on Si(001) n-type substrates and (b) the films were oxidized in an oxygen plasma chamber. The magnetron deposition system consisted of dual-cathode unbalanced RF/DC sputtering at an angle of 45° concerning the sample holder. A high-purity Sn target (99.999%), purchased from the Kurt J. Lesker company (Dresden, Germany), was powered by an RF source (13.5 MHz frequency) working at 5 W during the deposition. The constant flow of Ar_2 gas (purity 99.999%) in the chamber at 50 sccm (Aalborg Instruments & Controls, Inc., Orangeburg, NY, USA) was used to sustain the working pressure in the vacuum chamber during deposition at 3×10^{-2} mbar (the base pressure was 2×10^{-5} mbar). Pre-sputtering in the Ar^+ condition for 5 min was applied before the deposition. The overall deposition procedure lasted 25 min in total, characterized by an alternative sequence of 5 min deposition-interruption steps to avoid the heating and melting of the target.

The oxidation process was performed in a home-made oxygen plasma chamber of a borosilicate glass tube under vacuum conditions, under a base pressure of 9×10^{-2} mbar. The setup was supplied with high-purity (99.999%) O_2 gas, with a fixed working pressure of 2×10^{-1} mbar during the process; the oxygen plasma in H mode was formed by applying Radio Frequency (RF) with power of 550 W. The established geometry and annealing times for the plasma treatment of thin films, and therefore the heating of the sample, has been studied in similar works [31,40], suggesting that the process could be used to control electrical properties as well. The Sn-oxide thin films were oxidized for a total time of 70 s, following different duration steps of oxidation (starting from 3 s and up to 13 s with a step of +2 s) and a pause duration of 90 s, to avoid melting and to implicitly control the oxidization temperature. Given the rapid temperature fluctuation in the process of plasma etching, crystallization in intermediate oxides is also possible. While temperature is considered an important parameter, a limitation of the etching procedure in the chamber is the lack of the in situ temperature during the oxygen plasma treatment. The proposed reaction for the formation of intermediate oxides follow a two-step mechanism:



Due to air exposure, we expected possible carbon contamination at the surface [46]. The final sample is semi-transparent in the area where the plasma was formed, as expected, showing a small change from the gray-like color of the Sn buffer layer.

2.2. Characterization Techniques

The structures of the oxygen-plasma-treated films were examined by X-Ray Diffraction (XRD) measurements with a Bruker D8 Advanced diffractometer in Bragg–Bertrano geometry for the 5 s, 7 s and 9 s annealing step samples. The measurements were performed with Cu K α radiation ($\lambda = 1.5406$ Å) and a secondary monochromator, operating at 36 kV and 36 mA. The diffractogram was collected in a step of 0.01° , with a 0.3 s dwell time for each step measurement (and 0.1 s for the 5 s sample). Data were acquired over a 2θ range of 15 – 60° .

For the validation of oxide phases and the evaluation of the surface's chemical composition, X-Ray Photoelectron Spectroscopy (XPS) was utilized under ultra-high vacuum conditions, maintaining a base pressure of 5×10^{-9} mbar. The apparatus employed a Mg K α (1253.6 eV) anode, facilitated by a SPECS GmbH system equipped with the Phoibos HSA-Phoibos 100 hemispherical analyzer. The energy resolution was calibrated to 1.18 eV, with a beam incidence angle of approximately 45° and an analyzed surface area of ≈ 1 μm . High-resolution spectra were collected using a 0.05 eV energy step and a dwell time of 1 s. Gaussian deconvolution of the spectra was conducted with CasaXPS v2.3 software, utilizing the GL(30) fitting function and the Shirley background type. Energy shifts in the spectra were corrected by referencing the C-C bond at 284.8 eV.

To investigate the electrical characteristics of the thin films, I–V curve measurements were performed in a four-probe configuration utilizing the SUSS Microtek PM5 probe station in conjunction with the Keithley 2400 Source Measurement Unit (SMU). The measurements were performed under three distinct conditions, covering a voltage range from -8 V to $+8$ V.

Additional photocurrent J–t measurements under a constant voltage of 1 V were conducted using 2-probe contacts with a solar simulator from Sciencetech Inc. (model 550–200-PS), which was equipped with an Osram XBO 150 W/CR Xenon lamp. This setup produces illumination of 100 mW/cm^2 and employs an AM 1.5 filter to analyze the sample behavior during light activation and deactivation.

3. Results and Discussion

3.1. X-Ray Diffraction

The structural properties of the oxygen plasma-treated thin films were studied by means of X-Ray Diffraction in Bragg–Bertrano geometry. The results for three annealing step times are shown in Figure 2a–c. The various peaks correspond to known crystal structures, being in very good agreement with literature. For SnO₂, a tetragonal structure was identified, matching with the (101), (111), (211), (220), and (002) directions [37,47–50]. The SnO₂ phase was confirmed by the (101) phase, although it is partly “buried” from the other oxides. As for the formation of SnO, the tetragonal structure was shown as well, with the diffraction peaks corresponding to the (001), (101), (110), (002), (200), (112), (211) directions [37,51,52]. In SnO, the main peak of (101) is clearly shown. The presence of SnO and SnO₂ oxide peaks indicates the successful plasma treatment of the sample. In addition, metallic Sn in the tetragonal structure (301) plane was also present [2]. Moreover, we observed the intermediate Sn₃O₄ (003), ($\bar{1}\bar{1}\bar{2}$), and (112) reflection planes [26]. The intermediate tin oxide was found in the triclinic crystal structure, and the main (003) plane is centered at 32.95°. Additively, an intense peak at 33.05° corresponds to Si (002), and this is due to the Si wafer used as substrate for deposition. The identification of the patterns around 33° is rather complicated because of the interference of the Si substrate. The (002) diffraction peak is considered a “forbidden” diffraction peak attributed to the distortion of the lattice of the Si substrate [53]. It totally overlapped the (110) plane of SnO at 33.15°, which is “buried” in the pattern. Lastly, the peaks at 33.64°, attributed to the Sn₃O₄ (112) reflection of the intermediate oxide and the (101) reflection of SnO₂ at 34.83°, are also depicted in the inset for the 7 s sample, as depicted in Figure 2d, for making the identification clearer. Although the samples were all scanned in the same procedure, the sample oxidized with a 5 s step is observed to have broader SnO-related peaks and noise, indicating that for smaller etching time steps, the formations are more particle-like. As the step increases, the peak width decreases, indicating the formations of larger crystallites.

The first estimation of the average grain size (*D*) was calculated by means of both Scherrer’s formula [54] and the Williamson–Hall method for the 5 s annealing step’s SnO characteristic peaks. No detailed Williamson–Hall plot was created for the other samples, since the broader peaks disappeared due to the increase in crystallinity. Using Scherrer’s formula for Equation (1), the average grain size was estimated to be around 21 nm. Moreover, from the W–H plot (Figure 3), the average grain size, determined by extrapolating the linear fit according to Equation (2), was estimated to be 37 nm, and the difference from that determined using Scherrer’s method indicates the presence of micro-strains in the thin film.

$$D = \frac{\kappa\lambda}{b\cos(\theta)} \quad (1)$$

$$b\cos(\theta) = \frac{\kappa\lambda}{D} + 4\epsilon\sin(\theta) \quad (2)$$

For the other two samples from Figure 2a–c, we observe that the broad SnO peaks disappear and only the sharp high-crystallinity peaks remain. In Table 1, the details of some main peaks are shown. Focusing on the main peak of SnO in the (101) plane the Scherrer method calculations, a huge change in size is observed, showing an increase in crystallinity, which can be explained by the grain size drastic boost.

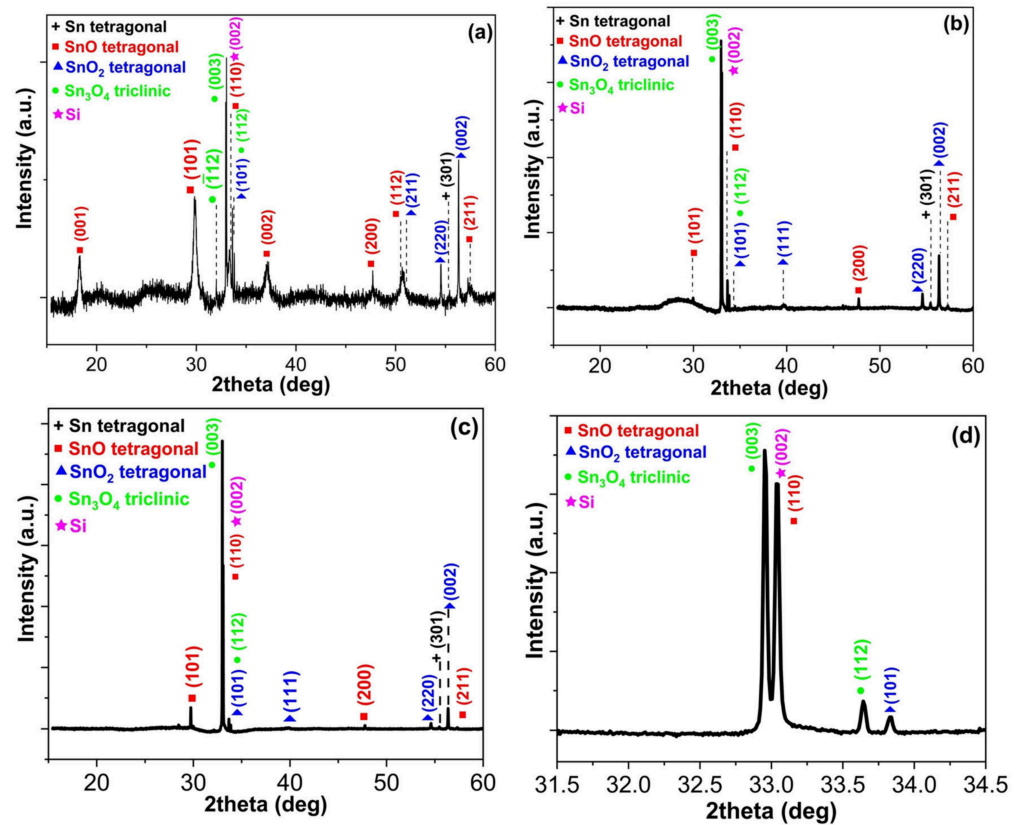


Figure 2. The XRD pattern of the oxygen-plasma-treated Sn thin films with a (a) 5 s annealing step, (b) 7 s annealing step and (c) 9 s annealing step. In (d), a close-up inset of the 7 s annealing step is depicted.

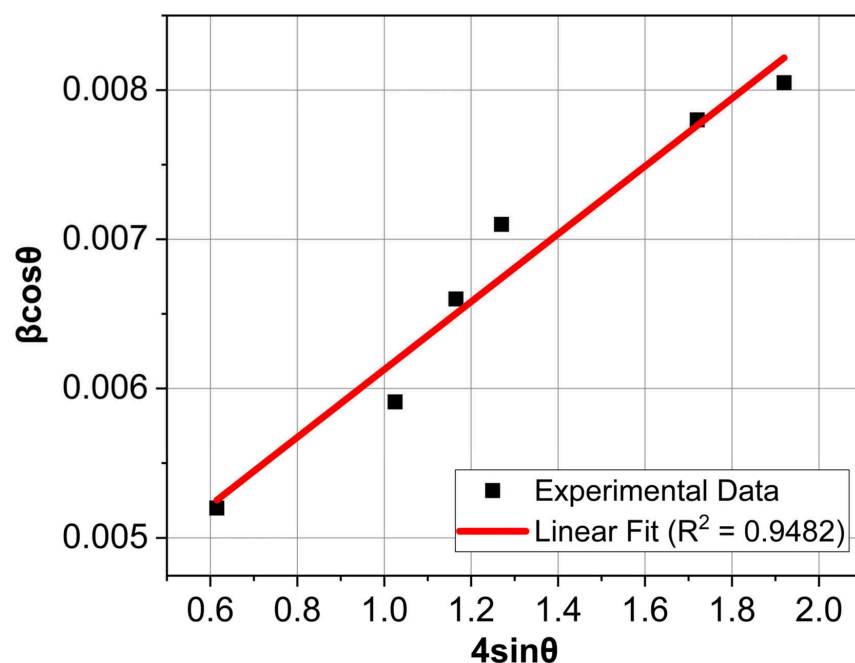


Figure 3. Williamson–Hall plot for the XRD analysis of the 5 s step sample. The black squares are the experimental points taken from the peaks. The red line is the linear fit.

3.2. X-Ray Photoelectron Spectroscopy

For the study of the elemental composition of the thin film's surface, we conducted X-Ray Photoelectron Spectroscopy (XPS) measurements. The measurements were performed

with the use of a Mg anode with an electron energy of 1253.6 eV. The spectra for the Sample with the 9 s step are shown below. For the other samples, the same pattern is applied. From the survey scan (Figure 4a) performed in the samples, a total of three atoms were matched from the peaks: Sn, O, and C. The quality of the thin film can be certified by the lack of impurities. The existence of carbon in the spectrum can be attributed to adventitious carbon atmospheric exposure [55]. Additionally, high-resolution spectra of the Sn 3d, O 1s and C 1s photoelectron peaks were collected, as shown in Figure 4b–d.

Table 1. Evolution of XRD peak values for some of the main peaks.

(101) SnO Tetragonal				(002) SnO ₂ Tetragonal			
Ox. Step	2 θ (deg)	FWHM	D (nm)	Ox. Step	2 θ (deg)	FWHM	D (nm)
5 s	29.79	0.401	20.5	5 s	56.31	0.065	138.6
7 s	29.93	0.042	195.8	7 s	56.33	0.064	141.5
9 s	29.73	0.047	174.9	9 s	56.37	0.064	141.9
(001) SnO Tetragonal				(003) Sn ₃ O ₄ Triclinic			
Ox. Step	2 θ (deg)	FWHM	D (nm)	Ox. Step	2 θ (deg)	FWHM	D (nm)
5 s	18.28	0.295	27.3	5 s	32.95	0.032	258.9
7 s	-	-	-	7 s	32.94	0.035	236.7
9 s	-	-	-	9 s	32.99	0.037	223.9

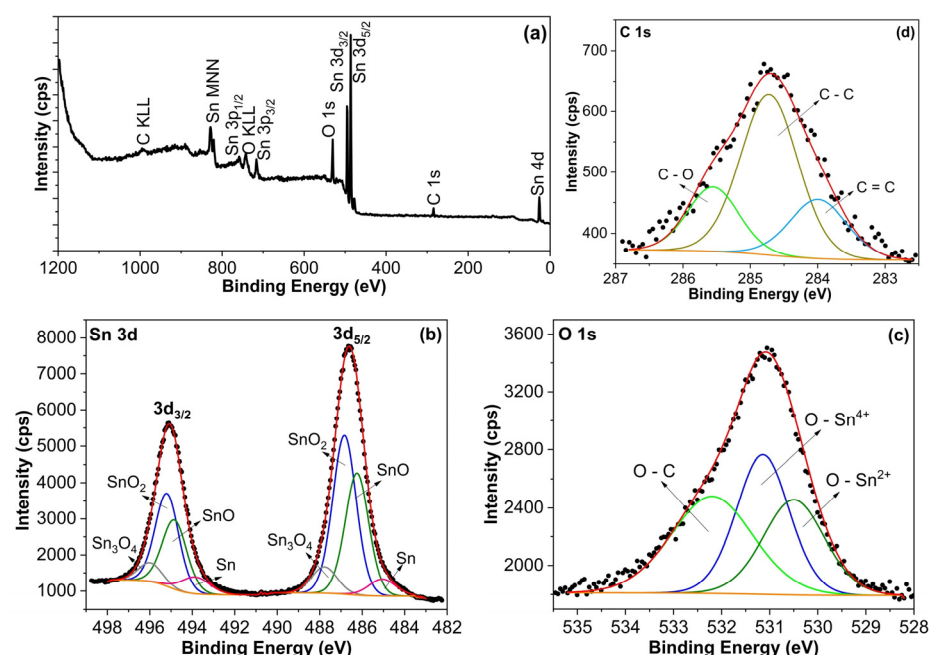


Figure 4. XPS spectra of Sn-oxide thin films. (a) Survey scan and high-resolution scan of (b) Sn (tin) 3d, (c) O (oxygen) 1s, and (d) C (carbon) 1s peaks. The experimental data are shown in dotted patterns and the deconvoluted data in dashed lines.

For the main Sn 3d peak after the Gaussian deconvolution, a total of four extra peaks were fitted in each photoelectron peak, as shown in Figure 4b. At 485.0 eV and the 493.4 eV, the peaks correspond to the metallic Sn phase [56], at the 486.2 eV and 494.6 eV, the peaks correspond to the SnO phase [57–59], and at the 486.7 eV and 495.1 eV, the peaks correspond to the SnO₂ phase [58–61]. In addition, the two peaks at 487.8 eV and 496.2 eV [59,62] could be attributed to the Sn₃O₄ intermediate tin oxide, consisting of both Sn²⁺ and Sn⁴⁺ electron

states. It is assigned as a discrete peak due to the influence of the tetrahedral coordination, which is dominant in the geometry of the intermediate phase. The peaks of the Sn 3d_{3/2} peak are in complete agreement with those of the Sn 3d_{5/2} peak, with the precalculated energy difference of around 8.4 eV. Therefore, the phases of the XPS fitting are in complete agreement with the XRD patterns.

As for the fitting of the O 1s photoelectron peak (Figure 4c), three different peaks were determined. The first peak, 532.2 eV, is the O-C bond, which is formed due to the existence of adventitious carbon from air exposure [59,63]. There are two other deconvoluted peaks with binding energies at 531.1 eV and 530.5 eV that correspond to the metallic oxides' photoelectron peaks (O-Sn⁴⁺ and O-Sn²⁺) [61,63]. Lastly, the deconvolution of the C 1s peak (Figure 4d) is also designated three Gaussians that can be attributed to the C=C bond at 284.0 eV, C-C bond at 284.8 eV and C-O bond at 285.6 eV [64].

In addition, for the other samples, no contamination from other elements was detected. The different annealing steps could have resulted in composition changes in the resulting oxides. Figure 5 depicts the evolution of the concentrations of the tin oxides and suggests their strong dependence on the annealing step during the post-growth treatment. The intermediate oxide concentrations decrease in the bigger steps, while the main oxide concentrations increase. Therefore, for small treatment times, there are fewer interfaces between SnO and SnO₂. As the annealing step is increased, the main oxides' concentrations are increased, while that of Sn₃O₄ is decreased, an at a step of 9 s, the SnO₂ and SnO concentrations are equal, showing that the evolution in Figure 5 seems to follow a non-linear dependence rule. Because the temperature is implicitly controlled and not controlled in real time, no clear relationship can be determined.

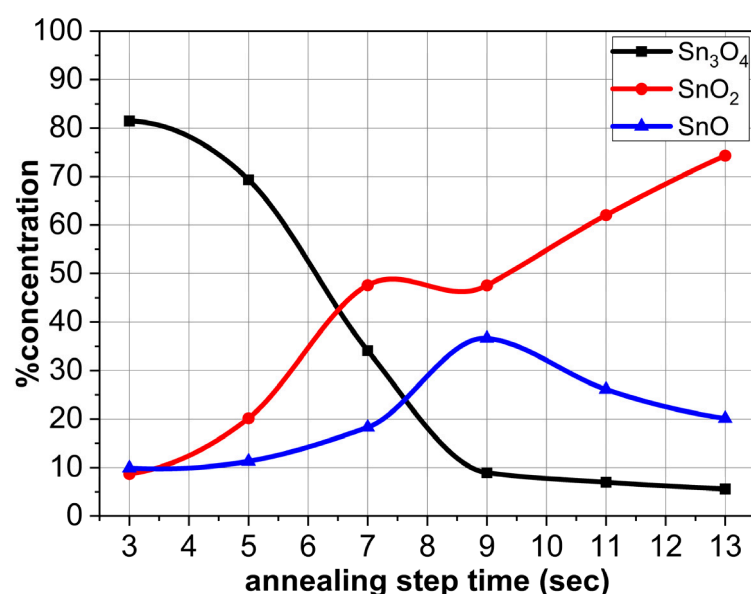


Figure 5. The evolution of the concentrations of the Sn oxide phases with respect to the annealing step time. The black, red and blue dots/lines correspond to the Sn₃O₄, SnO₂ and SnO phases, respectively.

3.3. I–V Characteristics

To elucidate the role of the different concentrations of the phases in the performance of the samples (in accordance with the annealing step), we performed four-probe current–voltage (I–V) measurements in the voltage range of −2 V to 2 V under various illumination conditions, i.e., dark, yellow and white light. We note here that the two illumination sources we used provide spectra in the range of low-light photovoltaic applications (Figure 6a–c) [65]. The yellow light source refers to the characteristic halogen lamp spec-

trum, delivering a radiation intensity of 2 mW/cm^2 , while the white light source to the spectrum of cool white LEDs delivered an intensity of 45 mW/cm^2 .

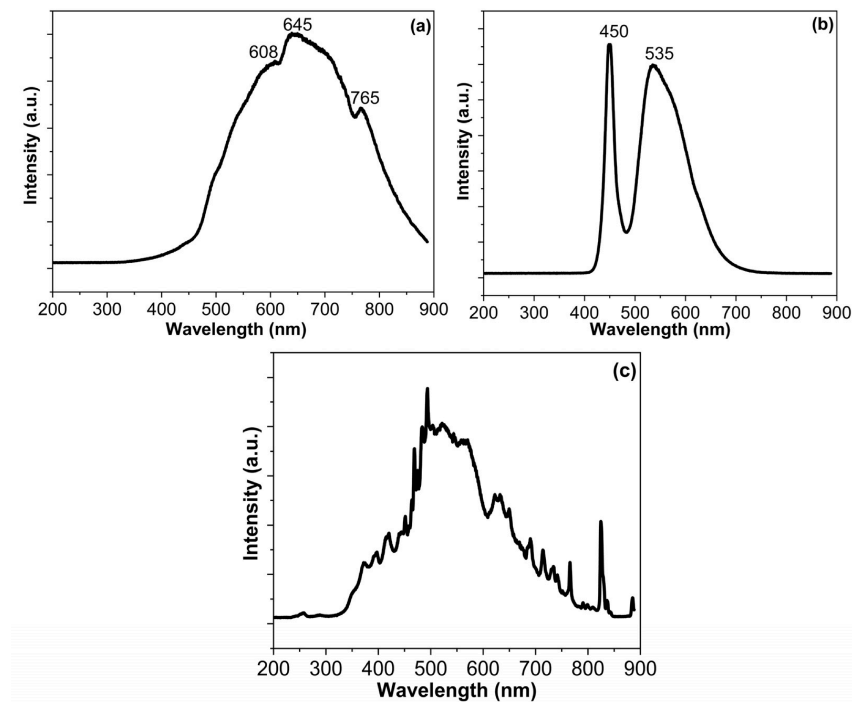


Figure 6. The lamp emission spectra for the (a) yellow light and (b) white light (c) solar simulator xenon lamp.

The analysis of the I–V curves of all samples exhibits nearly linear behavior in the smaller voltage range, suggesting an Ohmic relationship similar to that of a photoresistor [66]. After a certain voltage, which is different for each illumination condition, the curve deviates from the Ohmic behavior. The resulting I–V characteristics over a voltage range of -2 V to 2 V are depicted in Figure 7a–c.

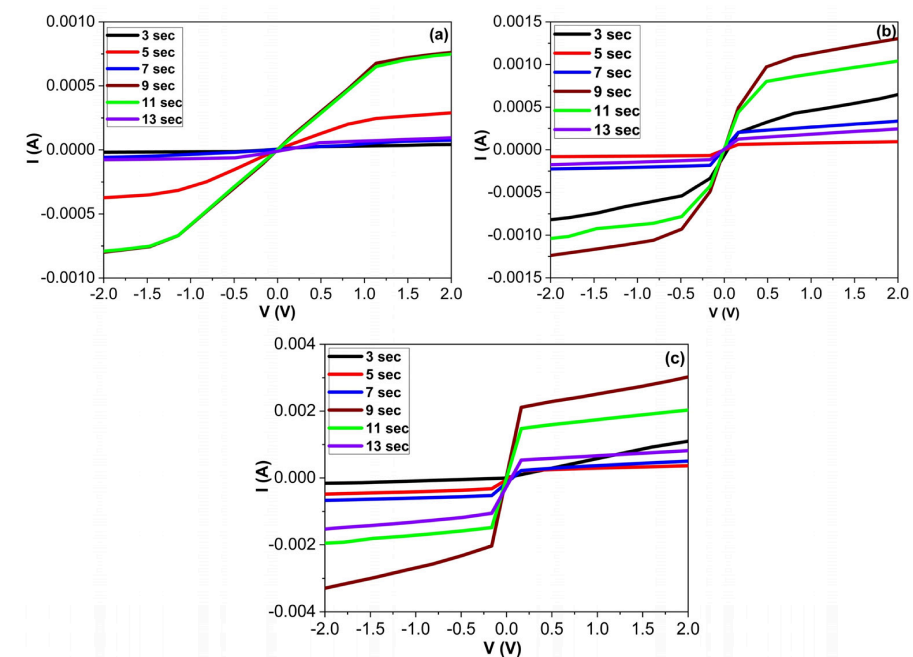


Figure 7. The I–V characteristics of the Sn oxides thin films samples for (a) darkness, (b) yellow light, and (c) white light.

The samples referring to 9 s and 11 s annealing steps exhibit the highest current for all illumination sources. The variations in current can be correlated with the oxides' concentrations, which can be found from the XPS spectra, showing that the 9 s annealing step sample demonstrates the best resulting photocurrent as it attains the highest current values. Specifically, by examining the concentrations shown in Figure 5, it becomes clear that lower concentrations of Sn_3O_4 , along with the higher percentages of the main oxides, improves significantly the resulting current in the electrical measurements, as shown in Figure 7. Specifically, the evolution curve of the concentration for all samples indicated that for the concentration of $\text{SnO}_2/\text{SnO} \approx 1$, the maximum performance is observed, followed by the sample of 11 s, where $\%\text{Sn}_3\text{O}_4 < 10\%$.

Additively, to determine the samples' behavior under greater voltages, the evolution relationship for the maximum current additional I–V curves were acquired at $V = \pm 8$ V. In Figure 8, an analog to the concentration relationship shows that the maximum and minimum current values are also reliant on a non-linear relationship, just like the concentration's evolution. Therefore, the annealing time step control can allow for the potential prediction and predetermination of the total current and also of surface concentrations occurring even before the etching process.

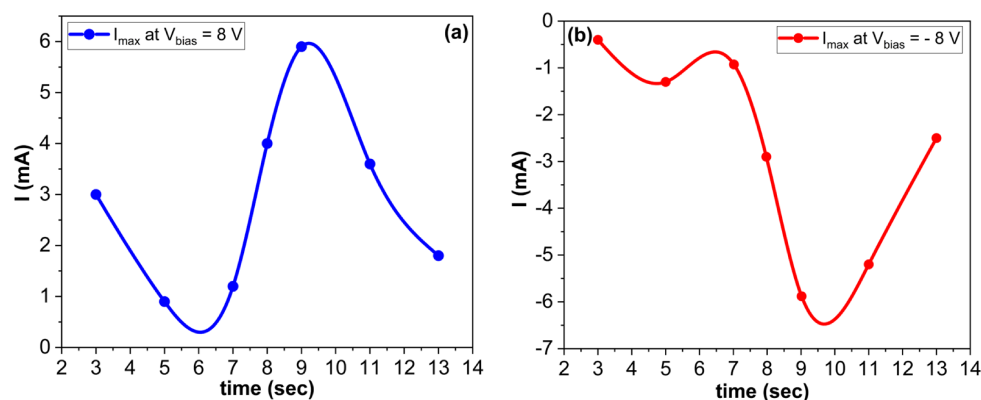


Figure 8. (a) Annealing step time-dependent evolution of maximum current values for $V = 8$ V. (b) Annealing step time-dependent evolution of the minimum current values at $V = -8$ V.

3.4. Photo-Sensing Property Evaluation

Photocurrent measurements were conducted to assess the responsivity and reversibility of the Sn oxide thin films. These measurements are usual in various contexts for applications at room temperature [67,68]. The photocurrent measurements of devices like photoresistors represent the process by which the optical signal from a light source is transformed into electrical energy [69].

The time-dependent photocurrent J – t plots were obtained for all samples (Figure 9a) for a fixed voltage of 1 V under illumination of $100 \text{ mW}/\text{cm}^2$ with the AM1.5 filter equipped. The emission spectrum of the lamp is illustrated at Figure 6c. We recorded three ON/OFF cycles after 30 s of darkness in the beginning, with 3 min duration of each cycle. All samples exhibited instant responses in the photocurrent density upon illumination and the deactivation of light. The samples with smaller annealing times were characterized by smaller photocurrents and longer times for reaching saturation. Correlating these findings with the XPS spectra, we can realize the importance of the percentages of SnO_2/SnO in the photodetection process. It turns out that in the samples that exhibit weaker photocurrents the excessive presence of Sn_3O_4 acts more as an electron trap, thus prolonging the overall detection time. For greater annealing steps, the increased concentrations of the main oxides greatly affect the resulting photocurrent density.

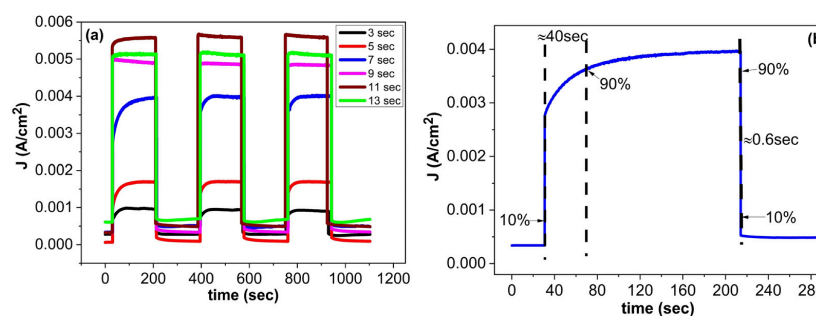


Figure 9. (a) Photocurrent J–t plots for the samples under the supply of 1 V in the solar simulator. (b) Characteristic photocurrent times shown as a schematic example for the 7 s annealing step sample.

To estimate the effectiveness of the samples, the rise and decay characteristic times [66] were determined for each sample. The rise time is defined as the duration required for the current to ascend from 10% to 90% of its maximum value, as depicted in Figure 9b. Conversely, decay time is defined as the duration for the current to decrease. Among the as-grown samples, those subjected to an annealing process exceeding a time step of 7 s demonstrated exceptional photocurrent properties.

For further evaluating the performance of the potential Sn oxide photodetectors, the responsivity (R) was calculated by means of the following equation:

$$R = \frac{J_{ph} - J_{dark}}{P} \quad (3)$$

where J_{ph} represents the photocurrent density, J_{dark} denotes the current density in darkness, and P is the illumination source intensity [66,70]. The most characteristic parameters are summarized in Table 2 in a comparative analysis for all samples under a solar simulator with an applied voltage of 1 V. Based on Equation (3), the responsivity of the sample with an 11 s annealing step reaches a maximum value of 0.73 A/W, indicating an effective photon-to-electron/hole conversion efficiency. This indicates that this sample is the best overall in terms of photoresponsivity under a solar simulator. Its superior values over the 9 s sample are probably due to the maximum presence of SnO_2 , as shown in the evolution curve of Figure 5. This indicates that the solar simulator light stimulates the SnO_2 -rich samples more efficiently even with decreased SnO concentrations due to a small edge in the emission spectra in the UV region, as shown in Figure 6c.

Table 2. The characteristic rise, decay times and responsivity values for the photocurrent measurements of the Sn oxides.

Annealing Step (s)	3	5	7	9	11	13
J_{ph} (A/m ²)	89.62	254.13	470.12	681.47	770.45	568.58
J_d (A/m ²)	37.31	9.40	40.09	38.59	45.22	66.09
R (A $\times$ W ^{−1})	0.05	0.25	0.43	0.64	0.73	0.50
Rise Time (s)	42.00	34.00	40.00	0.09	0.04	0.11
Decay Time (s)	4.60	4.00	0.60	0.07	0.06	0.07

Moreover, another parameter for assessing a photodetector's performance is the specific detectivity (D^*). This quantity evaluates the device's ability to detect optical signals effectively in low-light conditions, and it is described by the following formula [71]:

$$D^* = \frac{R}{(2eJ_{dark})^{1/2}} \quad (4)$$

where e is the electron charge and J_{dark} is the dark current density. By applying Equation (4), the specific detectivity was calculated at 2.0×10^8 Jones (1 Jones = $1 \text{ cm Hz}^{1/2} \text{ W}^{-1}$), showing excellent detecting properties [72].

The responsivity and the detectivity of the samples were also calculated under yellow and white light for the sample with a 9 s annealing step and an applied voltage of 0 V in order to compare the properties of the thin films under different illumination conditions. Applying Equations (3) and (4), for white light with an intensity of 45 mW/cm^2 , a responsivity of 2.2 A/W was calculated, with a specific detectivity of 1.9×10^9 Jones. As for its exposure to the yellow light with an intensity of 2 mW/cm^2 , a maximum responsivity of 12.5 A/W was calculated, as was a specific detectivity of 1.1×10^{10} Jones. From the results of the parameters, we observe better detection under yellow light, despite the significantly lower current, as shown in the I–V characteristics, indicating better light absorption in the near-IR regions.

A comparative analysis of these characteristic results with other Sn-based photodetectors from the bibliography is presented in Table 3 [71,73–76]. The measured responsivity and specific detectivity values suggest that the materials produced with our two-step method, especially those treated with 9 s and 11 s annealing steps, hold substantial potential for photo-electric applications due to their excellent photoresponsivity parameters, relatively fast process and low overall manufacturing costs.

Table 3. Photoresponse values of this work for different illumination conditions and comparison with other Sn-based photodetectors from the bibliography.

Materials	Bias (V)	λ (nm)	R ($\text{A} \times \text{W}^{-1}$)	Response Time	Reference
GNWs/SnO ₂	1	365	8.6×10^{-3}	-	[73]
Ag/SnO ₂ NSs/Ag	1	267	539.60	80/1217 s	[74]
Au/SnO ₂ NWs/n-Si	0	UV	3.6×10^{-4}	0.72/1.78 s	[75]
Gr/SnO ₂ MW/GaN	0	360	0.22	18 μs /580 μs	[71]
ITO/SnO ₂ MW/GaN	0	360	3.0×10^{-3}	53 ms/71 ms	[71]
SnO ₂ /TiO ₂	0	370	0.15	0.14 s/0.06 s	[76]
Sn Oxides (solar)	1	Figure 6c	0.73	0.04 s/0.06 s	This work
Sn Oxides (white)	0	Figure 6b	2.20	-	This work
Sn Oxides (yellow)	0	Figure 6a	12.50	-	This work

4. Discussion

The formation process of the Sn-oxide thin films via the two-step method was successfully implemented. The XRD pattern demonstrated the presence of the three metal oxides, SnO₂, SnO and Sn₃O₄, as confirmed by XPS. The good agreement between the two techniques shows that there exists consistency in the surface properties found with XPS, as corroborated with the bulk material scanned from XRD data. Moreover, we confirmed the absence of extra impurities. Together, these complementary techniques provide strong evidence for the successful formation of the intended material and confirm the consistency between its bulk crystal structure and surface chemical composition. The samples with a low Sn₃O₄ concentration in the outer surface and relatively close main oxides concentrations were found to exhibit better I–V characteristics upon illumination. The recorded electric measurement curves demonstrate diode-like Ohmic behavior for small voltages, showing signs of sensitivity upon different sources of intensity and illumination conditions. The correlation relationship between the surface oxide concentrations and maximum current and therefore photodetection values was thoroughly investigated. A

non-linear relationship predicting the dependence of the surface oxide concentrations and the maximum photocurrent on oxidation was shown to exist.

The evaluation of the samples' photo-sensing abilities was performed by recording the characteristic photoresponse J–t plots. Under both lights, they showed excellent parameters, which prove that the photocurrent is strongly dependent on the annealing step. For the 9 s annealing step under yellow LED light with an intensity of 2 mW/cm^2 at $V = 0 \text{ V}$, a maximum responsivity of 12.5 A/W and a detectivity of $1.1 \times 10^{10} \text{ Jones}$ were determined. Similarly, for the 11 s annealing step under the solar spectrum at $V = 1 \text{ V}$, a responsivity of 0.73 A/W and detectivity of $2.0 \times 10^8 \text{ Jones}$ was found, with characteristic rise/decay times of 40/60 ms. As determined for the samples with a 9/11 s annealing step, the best results were found for almost equal concentrations of the two main oxides (SnO/SnO_2). The first results are promising but further photo-sensing evaluation with EQE (external quantum efficiency) and spectral responsivity are important in future plans.

This work first showed evaluation of the applied two-step growth method over traditional processes that utilize a single step mechanism. The focus was placed on the effect of the surface concentration of the thin films and its direct relationship with the total sensing mechanism and its direct association with the heterojunction effect. Another important factor of the samples is that even after six month, the properties of the film did not seem to decrease, while its structure looked the same. Still, the long-term stability of the samples under ambient conditions is one aspect that needs to be further considered for potential device fabrication. Some future aspects remaining to be studied in follow-up projects include the exact in situ temperature in the borosilicate chamber during the plasma treatment, which needs to be determined in order to fully control the formation of phases and the crystal structure even more precisely. Also, key factors like the total thin film thickness, the surface morphology and the effect of a potential non-oxidized metallic Sn buffer layer between the substrate and the film are important concerns for the exploration of further potential applications.

5. Conclusions

Sn-oxide ($\text{SnO}_2/\text{SnO}/\text{Sn}_3\text{O}_4$) thin films were produced by a two-step process of oxygen plasma treatment of metallic Sn thin films deposited via RF magnetron sputtering. The XRD diffractogram showed the polycrystalline nature of the samples, indicating the tetragonal and triclinic structures for the oxides. XPS studies showed the evolution of the presence of Sn oxides and their respective concentrations. The study aligned with the correlation of the electrical properties of the samples with respect to the XPS oxide concentrations. The optoelectronic properties obtained by the I–V characteristics and the photo response J–t plots both showed that the photocurrent is strongly dependent on annealing step. Therefore, those two samples could also hold potential for various other optoelectronic related applications such as photocatalysis, gas sensing, and photoelectric conversion.

Author Contributions: Conceptualization, G.A.E., I.P. and P.P.S.; methodology, I.P., C.P. and G.A.E.; software, I.P. and P.P.S.; validation, I.P., P.P.S., K.A.A., C.P. and G.A.E.; formal analysis, I.P., P.P.S. and K.A.A.; investigation, K.A.A. and C.P.; data curation, I.P. and P.P.S. and G.A.E.; writing—original draft preparation, I.P. and P.P.S. and G.A.E.; writing—review and editing, I.P., C.P. and G.A.E.; visualization, I.P. and P.P.S.; supervision, C.P. and G.A.E.; project administration, C.P. and G.A.E. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

RF	Radio Frequency
DC	Direct Current
a.u.	Arbitrary Units
cps	Counts per Second

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