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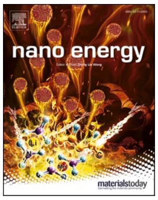

Communication

Enabling bifacial thin film devices by developing a back surface field using Cu_xAlO_y

Kamala Khanal Subedi [1], Adam B. Phillips [1], Niraj Shrestha, Fadhil K. Alfadhili, Anna Osella, Indra Subedi, Rasha A. Awni, Ebin Bastola, Zhaoning Song, Deng-Bing Li, Robert W. Collins, Yanfa Yan, Nikolas J. Podraza, Michael J. Heben, Randy J. Ellingson *

Wright Center for Photovoltaic Innovation and Commercialization, Department of Physics and Astronomy, The University of Toledo, Toledo, OH 43606, USA

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ABSTRACT

Bifacial solar cells have the potential to increase the energy yield per unit area over traditional monofacial devices without significant added cost, driving $/kWh costs lower and accelerating the adoption of solar photovoltaics. However, the performance of bifacial thin film solar cells significantly lags that achieved by crystalline silicon cells. Here we incorporate wide bandgap Cu_xAlO_y as a back buffer layer for CdTe devices and achieve a backside illuminated device with high current density and high fill factor. Moreover, these values remain nearly constant even as the absorber layer thickness changes, indicating that a fully-depleted device is not required for efficient charge collection. We show that this response is indicative of a back surface field, albeit with a persistent high back surface recombination velocity. By managing electron reflection, we achieved a backside illumination conversion efficiency of 7.1% and bifaciality of 0.55 for a 3.3 μm CdTe device and 8.0% and 0.62 for a 2 μm device. Future improvements can be made by identifying and incorporating a passivation material that reduces the back surface recombination velocity.

1. Introduction

As single junction photovoltaic (PV) devices push closer towards the Shockley-Queisser limit, [1] new architectures are being investigated to push the energy yield per area higher. One of the most promising is bifacial PV devices, in which ground-scattered light incident on the back of the device gets absorbed and converted into additional electrical energy. In fact, it is estimated that bifacial PV will make up ~ 40% of the PV market by 2025 [2], and bifacial Si devices are already commercialized. [3].

Development of bifacial thin film (BTF) devices, on the other hand, lags considerably, and the record reported efficiency values of backside illuminated devices are 6.0% for CIGS [4] and 5.0% for CdTe [5]. Much of the reason for the poor performance is due to downward band bending in the absorber layer at the back interface of the device due to a negative initial Fermi level offset (IFLO) and high back surface recombination velocity (BSRV) [6] in addition to highly absorbing back buffer layers [7,8]. From an energetics point of view, if a back buffer layer can induce upward band bending at the back interface, the electrons will be repelled and the backside illuminated device performance will improve [6]. In addition, these types of improvements will be necessary to achieve the highest front illuminated device performance [9], when the front interface is good [10]. Unfortunately, development of such materials has been slow.

Recently, several studies have developed p-type conductive, transparent films that are candidates for use in photovoltaic devices [11–15]. We recently investigated $(CuS)_x(ZnS)_{1-x}$ and $BaCu_4S_3$ as back contact buffer layers for CdTe [16,17]. While incorporation of these materials have led to reasonable front illuminated device performance, conversion efficiency when illuminated from the backside has been poor. This suggests that the IFLO between these back buffer layers and the absorber remains negative, which leads to downward band bending in the absorber at the back interface resulting in high back interface recombination current density [6,9]. $CuAlO_2$, on the other hand, has several potential advantages. It is a p-type wide bandgap material with high visible light transparency, high carrier concentration $> 10^{17}$ cm^{-3}, and expected deep valence band edge, suggesting the potential for positive IFLO [11,18]. To achieve the delafossite crystal structure, though,

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* Corresponding author.
E-mail address: Randy.Ellingson@utoledo.edu (R.J. Ellingson).

[1] These authors contributed equally to this work.

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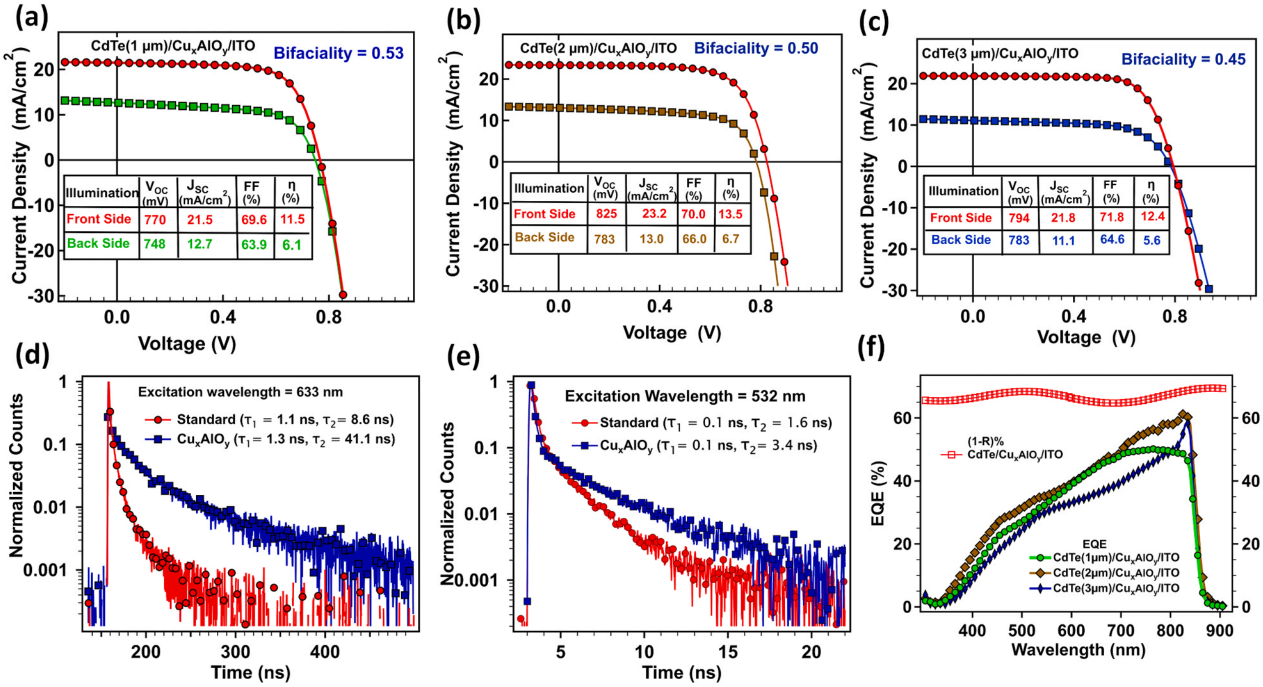
requires annealing the sample at temperatures > 700 °C, which would be detrimental to CdTe devices. However, low temperature (300 °C) annealing of atomic layer deposited Al_2O_3 with a Cu overlay has resulted in a mixed Cu_xAlO_y layer that has led to increased CdTe device performance [19]. This suggests the desired back buffer material properties may be achievable with Cu-Al-O based materials at CdTe-compatible processing temperatures.

Here we investigate Cu_xAlO_y as a transparent back buffer layer to CdTe fabricated by low temperature solution processing. We investigate devices completed with a transparent conducting oxide (TCO) back contact and measure the performance when the device is illuminated through frontside and backside. While front illuminated device performance is consistent with previous devices using these absorber stack materials, back illuminated device performance is high for a BTF device and independent of absorber thickness layer. We use quantum efficiency (QE) and reflection measurements to determine the location of the efficiency loss. Using SCAPS modeling, we identify the likely conditions of the back interface. In addition, we apply an antireflective coating to fabricate an unverified record efficiency back illuminated CdTe device.

The device structure used for these investigations consisted of glass/TCO/CdS/CdTe/Cu_xAlO_y/TCO, where the front glass/TCO was F-doped SnO_2 coated glass substrate with a high resistive transport layer (TEC™ 12D; Pilkington USA) and the back TCO was Sn-doped In_2O_3 (ITO). The CdTe absorber thicknesses studied were 3.3, 2.0, and 1.0 μm. For the devices with thick absorber layer, the ~ 150 nm CdS and 3.3 μm CdTe were deposited using a commercial vapor transport deposition system at high temperature (Willard and Kelsey Solar Group). For the two thinner devices, the 80 nm CdS window layer and the CdTe absorber were sputtered as described elsewhere [20]. Prior to back buffer deposition, all samples were $CdCl_2$ activated at 390 °C for 30 min [20]. No additional Cu was added to these devices. However, devices fabricated using these materials without additional Cu perform reasonably well (~ 10.5%; Supporting Information), indicating that all sets of samples are Cu-doped, though not at an optimized level. The Cu_xAlO_y films were spin coated onto the activated CdTe using a solution of 40 mM aluminum nitrate nonahydrate (98%, Fisher Scientific) and 24 mM copper nitrate trihydrate (99%, Fisher Scientific) dissolved in 5 ml of 2-metoxyethanol followed by a post-deposition annealing treatment at 220 °C for 6 min to convert the precursors into the oxide. Note that the solution preparation and spin coating closely follows the process used to fabricate the $CuAlO_2$ as reported by others.[18,21] The devices were finished by sputtering 250 nm of ITO, and the device area is defined by manual scribing of the one inch substrate to fabricate an array of cells of area 0.09 cm^2. Current density-voltage (J-V) curves were measured using a Keithley 2440 sourcemeter and MiniSol model LSH-7320 solar simulator with digital output control.

Fig. 1 shows the J-V curves for front and back illumination of the three devices. While these graphs show the best performing devices, the characteristics of these curves are representative of the 30 devices measured for the respective absorber thickness. Frontside illuminated device performance is in line with performance of devices made using the same deposition processes when Cu is added [15]. The backside illuminated device performance, though, shows several remarkable characteristics. First is that the device fill factor (FF) at all three absorber thicknessses is relatively high at ~ 64%. There is clearly a slope in the J-V curve at short circuit that sets a limit on the FF. However, this slope appears to be constant before a significant change occurs at ~ 700 mV. This sudden change and high FF value indicates that the recombination mechanism responsible for the slope on the J-V curve near short circuit conditions differs from the mechanism that turns on at ~ 700 mV and dictates the V_{OC}. Our previous modeling of backside illuminated devices shows that when there is negative IFLO and moderate BSRV (10^4 cm-s-^{1}), high J_{SC} can be achieved when the depletion region due to the front junction spans the absorber thickness (i.e., the device is fully depleted) at short circuit [6]. Even for fully depleted devices, though, the FF is low because downward band bending at the back of the device



Illumination	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF (%)	η (%)
Front Side	770	21.5	69.6	11.5
Back Side	748	12.7	63.9	6.1

Illumination	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF (%)	η (%)
Front Side	825	23.2	70.0	13.5
Back Side	783	13.0	66.0	6.7

Illumination	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF (%)	η (%)
Front Side	794	21.8	71.8	12.4
Back Side	783	11.1	64.6	5.6

Fig. 1. J-V performance of front and back illuminated (a) 1 μm, (b) 2 μm, and (c) 3.3 μm thick CdS/CdTe solar cell devices using Cu_xAlO_y/ITO as back buffer layer. Time-resolved photoluminescence response of 3.3 μm thick devices with and without Cu_xAlO_y illuminated from the (d) front and (e) back of the device. (f) Reflection (shown as 1-R) and external quantum efficiency of the devices with different absorber thickness.

emerges with increasing bias, rapidly increasing back interface recombination. The FF results presented here do not follow this pattern, which suggests that the back interface is not dictating device performance as is typically observed.

The second remarkable characteristic of the performance of devices illuminated through the backside is that the short circuit current densities (J_{SC}) are nearly independent of absorber thickness. This suggests that carrier lifetime in the thickest absorber is long, so that the same number of electrons are collected at the front of the device as the absorber thickness is varied. As shown in Fig. 1d and e, the lifetime is greatly improved with the addition of the Cu_xAlO_y buffer layer with a long component decay time (τ_2) that increases from 8.6 ns without the buffer layer to 41.1 ns with the buffer when measured from the frontside, and from 1.6 ns without the buffer layer to 3.4 ns with the buffer when measured from the backside. These measurements not only verify that the carrier lifetime is long, but they also suggest that recombination at the back interface has been significantly reduced.

Additionally, the fact that the J_{SC} is independent of thickness indicates that a common mechanism results in the current loss observed for backside illumination. To determine the nature of current loss, we measured the reflection and QE at zero bias for backside illuminated devices. The reflection data (plotted as 1 - R) in Fig. 1f shows, a ~ 32% reflection loss from the back of the device, but this value does not fully account for the observed J_{SC} loss. The QE, on the other hand, shows a high value at long wavelength and decreases for shorter wavelengths. Nearly all the carriers generated by long wavelength light absorbed closer to the front junction are efficiently collected. At the same time, nearly 50% of the carriers generated by short wavelength (~ 450 nm) light absorbed near the back junction are also collected. While recombination does occur at the back of the device, many carriers generated near the back interface are collected. These results are differ from previously reported backside QE results for thicker CdTe absorber layers. [22,23].

A J_{SC} demonstrating only weak dependence on absorber thickness suggests that the energy band profile at the back of the device is the same for all thicknesses investigated, while a FF that weakly depends on absorber thickness suggests this energy band landscape does not change with bias [6]. That this is the case across the three absorber thicknesses investigated here indicates that the bands may be bending upwards at the back of the device, demonstrating electron repelling behavior. From our previous modeling work, upward band bending achieved through a positive IFLO is consistent with reduced interface recombination and high efficiency devices [6]. This is not the case here, as the measured QE response indicates that recombination does occur at the back interface. Taken together, these results suggest that there may be upward band bending at the back of the device and a higher BSRV than that (10^4 cm-s^{-1}) considered in our previous modeling work.

To investigate the CdTe/Cu_xAlO_y interface in an attempt to better specify the energetic conditions and BSRV, we employed SCAPS modeling [24]. Our studies indicate that achieving a thickness- and bias-independent J_{SC} and FF requires upward band bending at the back of the device. This can be achieved with either a fully depleted absorber layer, fixed charges in the buffer, or a positive IFLO. SCAPS modeling indicates that for fully depleted devices, the frontside illuminated device performance increases as the absorber thickness decreases (SI). As shown in Fig. 1 this is not the case. In addition, devices using the same methods and materials used to fabricate these devices often perform better than expected for fully depleted devices, indicating they contain a suboptimal amount of Cu-doping [16]. Consequently, it is unlikely that these devices are fully depleted.

The alternative option is that Cu_xAlO_y results in upward band bending at the back interface. This can be achieved through fixed positive charges in the buffer layer or a positive IFLO. Al_2O_3 has been shown to improve interface behavior for a number of PV materials [25–27]. In these cases, some evidence suggests that fixed charges in the Al_2O_3 create an electric field that repels electrons, resulting in reduced interfacial recombination [26]. Al_2O_3 has also been used in CdTe devices [28–30], but the exact reasons for performance increases have not been identified [26]. While it may be possible that our solution processed Cu_xAlO_y actually results in the formation of an Al_2O_3 layer, additional experiments discussed in the SI indicate that this is not the case. When only one of the metal precursors in included in the solution processing, the backside illuminated device performance is significantly below the performance when both precursor materials are included. Alternatively, including the Cu source as the first of a two step process designed to dope the absorber layer followed by deposition of an Al_2O_3 film, the backside illuminated device performance is still lower. Furthermore, as the absorber layer thickness is decreased from 3.3 μm to 2 μm, a significant change in the J_{SC} occurs, results that are clearly different from those presented in Fig. 1.

From these results, it is clear that both the Cu and Al precursor material are necessary in the solution to fabricate the highest performing devices. The poor performance of the Cu only sample indicates that it is unlikely that the Cu simply converts into a Cu_xO phase, as others have shown at low temperatures [18,21], to improve device performance [31]. On the other hand, the large increase in J_{SC} for the devices with a Cu-doped CdTe layer with Al_2O_3 buffer as the absorber thickness is reduced indicates that the band bending in the CdTe at the back interface changes with absorber thickness. Furthermore, the decrease in FF for these devices with decreasing film thickness is suggestive of bias dependent band bending. Both of these data points taken together suggest that device has downward band bending with reduced back surface recombination velocity [6] and that the solution processing presented in this manuscript does not result in a Cu-doped CdTe layer with Al_2O_3 buffer layer. Instead, the Cu and Al precursor materials react to from a Cu_xAlO_y, which, while unlikely to form a pure $CuAlO_2$ phase at these temperatures, does exhibit some of the necessary properties required to improve the back interface of CdTe devices.

Thus, the available data suggests that upward band bending is due to the positive IFLO resulting from use of a Cu_xAlO_y back buffer layer. However, to have upward band bending and still lose J_{SC} requires high BSRV. To put a range on the band bending and BSRV for the situation presented here, we used SCAPS to model device performance when varying the IFLO by changing the back buffer acceptor density and varying the BSRV via the number density of the interface defect states. For this modeling work, we used materials parameters used in our previous studies (SI), with the caveat that we used an emitter with band positions of CdS.[6,9,10,32]. For the back buffer layer we assumed the valence band was 0.2 eV below that of CdTe, thereby potentially impeding hole flow, and 32% optical reflection from the back interface in accordance with the measured value. Fig. 2 shows how the PV parameters, including J_{SC} and FF, depend on the IFLO and BSRV for an absorber thickness of 3.3 μm.

From the contour plots, it is clear that there is a wide range of IFLO-BSRV values that will result in a J_{SC} of 11–12 mA cm^{-2}. Similarly, there is a wide range of IFLO-BSRV values that results in a FF of 65%. However, there is only a small range in which both values occur simultaneously, where IFLO is between 0.04 and 0.10 eV and BSRV is between 2×10^6 and 2×10^7 cm-s^{-1}. We note that for our model, we assumed the default ideal series and shunt resistances (0.0 and 10^{30} Ωcm^2, respectively). While nonideal resistance values can affect the FF of the device, we believe the bias-dependent current collection of these devices is dictated by the interface recombination and not the resistance values because the simulated JV curves (SI) show similar characteristics to the measured JV curves. Furthermore, the frontside JV response does not indicate limiting series and shunt resistances in this device. We also note that in this IFLO-BSRV range, the Voc is ~ 790 mV, which is close to the measuredvalues of our devices. Consequently, we believe Cu_xAlO_y back buffer layers have an IFLO ~ 0.07 eV, but the BSRV is high and on the order of 4×10^6 cm-s^{-1}.

To see how high the efficiency can be pushed beyond the 5% record value [4] while using a commercially fabricated device stack, we applied

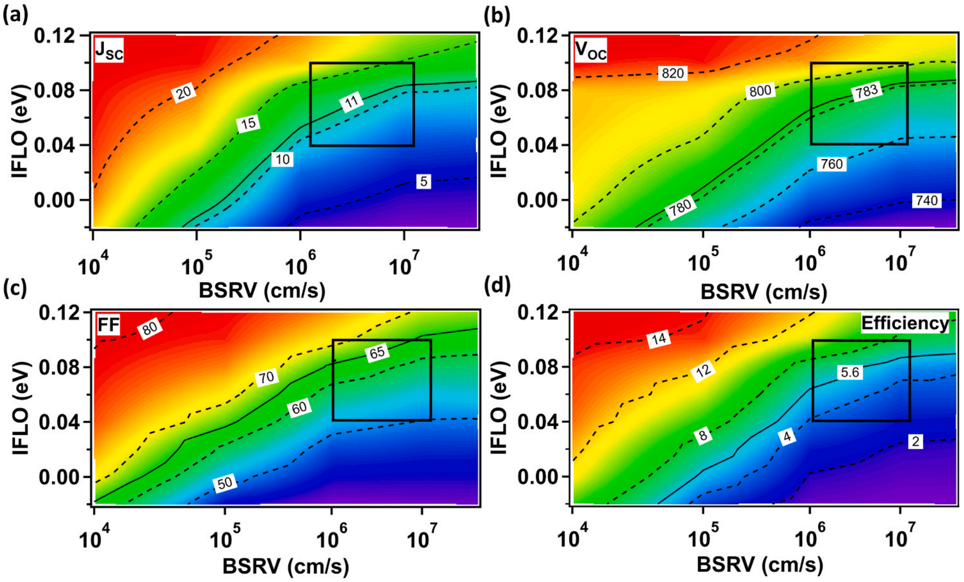
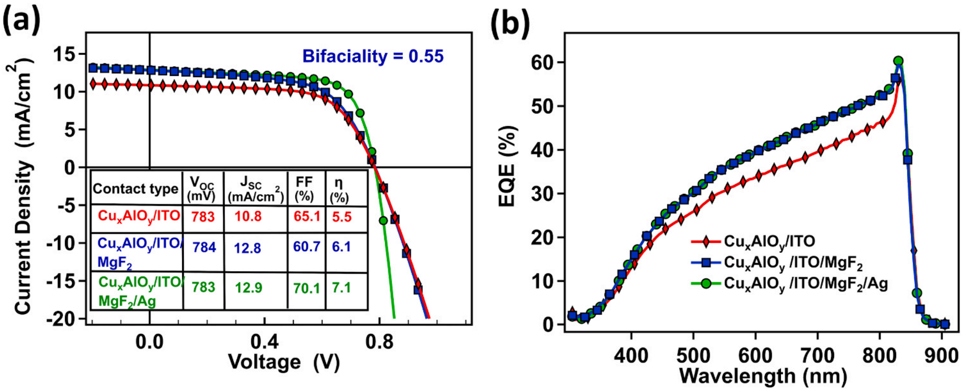


Fig. 2. Contour plots obtained by SCAPS modeling to obtain photovoltaic parameters for a 3.3 μm device illuminated through the backside with varying back surface recombination velocity and initial Fermi level off set. Note the solid contour line is the measured value of each parameter and the box shows the range of the IFLO and BSRV.



Contact type	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF (%)	η (%)
Cu_xAlO_y/ITO	783	10.8	65.1	5.5
Cu_xAlO_y/ITO/ MgF_2	784	12.8	60.7	6.1
Cu_xAlO_y/ITO/ MgF_2/Ag	783	12.9	70.1	7.1

Fig. 3. (a) J-V and (b) EQE curves of the champion devices with Cu_xAlO_y/ITO, Cu_xAlO_y/ITO/MgF_2, and Cu_xAlO_y/ITO/MgF_2/Ag (metal) grid of 3.3 μm thick CdTe device for back illumination.

a MgF_2 antireflective (AR) coating on the Cu_xAlO_y/ITO back contact stack of the 3.3 μm thick CdTe layer device. Fig. 3 shows the JV and QE response of the devices before and after. Note that the results for a 2 μm device with the same treatment is shown in the SI. As expected, the AR coating increased the current collected for all biases by allowing more light to reach the absorber. We note that there is a slight decrease in FF due an increase in series resistance caused by the added AR layer. When a metal grid is added to the device, we see the FF increase, resulting in a backside illuminated efficiency of 7.1% and a bifaciality of 0.55. For the 2 μm device, the backside illuminated efficiency of 8.0% and bifaciality of 0.62 are achieved (SI).

2. Conclusions

Using solution processing, we fabricated a wide bandgap Cu_xAlO_y back buffer layer for CdTe PV devices which shows strong evidence of an electron-repelling upward band bending at the back interface. When ITO was applied as the back electrode, we were able to fabricate devices with backside illuminated efficiency that was nearly independent of absorber layer thickness, and yielded back-illuminated conversion efficiency as high as 7.1% and 8.0% for 3.3 μm and 2 μm devices, respectively. Time-resolved PL results show that the Cu_xAlO_y back contact layer leads to significant gains in carrier lifetime when measured for illumination through either the front or back contacts. The J-V curve, though, shows a photocurrent that is significantly below the expected one sun value. Reflection from the back interface, though significant, does not account for all of the current density loss. The remainder of the loss is due to back interface recombination due to a persistent high BSRV that is offset somewhat by Cu_xAlO_y creating a positive IFLO and a concomitant back surface field.

CRediT authorship contribution statement

Kamala Khanal Subedi: Conceptualization, Data curation, Formal analysis, Investigation, Writing. **Adam B. Phillips:** Conceptualization, Data curation, Formal analysis, Investigation, Writing, Supervision, Project administration. **Niraj Shrestha**: Investigation. **Fadhil K. Alfadhili:** Investigation. **Anna Osella**: Investigation. **Indra Subedi**: Validation, Formal analysis. **Rasha A. Awni:** Validation, Formal analysis. **Ebin Bastola:** Conceptualization, Investigation. **Zhaoning Song:** Validation. **Deng-Bing Li:** Investigation, Validation. **Robert W. Collins:** Conceptualization, Funding acquisition. **Yanfa Yan:** Resources, Funding acquisition. **Nikolas J. Podraza:** Resources, Funding acquisition. **Michael J. Heben:** Resources, Funding acquisition. **Randy J. Ellingson:** Conceptualization, Funding acquisition, Data curation, Project administration, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2021.105827.

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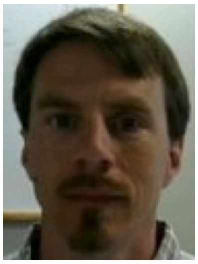
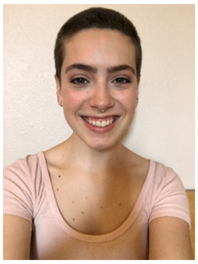
Kamala Khanal Subedi earned her B.Sc and M.Sc degree in physics from Tribhuvan University, Nepal. Currently, she is pursuing a Ph. D at the University of Toledo in physics with a specialization in photovoltaics under the supervision of Prof. Randy J. Ellingson. Her research is focused on the fabrication and characterization of CdTe thin film devices, modeling, and characterization of other semiconducting materials by using laser spectroscopy.



Adam B. Phillips earned his B.S. in Physics from Case Western Reserve University in 1999 and his Ph.D. in Physics from the University of Virginia in 2007. Since 2008 he has been working as research faculty at the University of Toledo where he works on improving photovoltaic device operation and understanding. His current research is focused on understanding, characterizing, and improving the absorber/charge transport layer interfaces of CdTe-based and perovskites devices to enable higher efficiency, lighter weight, or alternative device structures.

Niraj Shrestha received his Ph.D. degree from The University of Toledo, Toledo, OH in 2020. His research interests include study of semiconductor thin films based on their static and dynamic optical properties including techniques of ultrafast pump-probe spectroscopy, steady state photoluminescence spectroscopy and time resolved photoluminescence spectroscopy. At present, he is working as an Optical Scientist at NP Photonics in Tucson, AZ.

Dr. Fadhil Alfadhili obtained his Ph.D. in 2020 from the Department of Physics and Astronomy, the University of Toledo, Ohio, USA. For his achievements in the field of CdTe thin-film solar cells. His research interests are focused on device physics and materials science related to thin film nanomaterials, and photovoltaics.

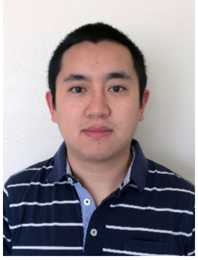


Anna Osella is a graduate student currently studying materials science at the Colorado School of Mines, focusing on semiconductors, fabrication, and material synthesis. She has a Bachelor of Science in physics from Michigan State University, where she worked on ferromagnetic materials for computer memory. She also spent one summer working at the University of Toledo on a bifacial solar cell measurement system, and a semester working at the National Renewable Energy Laboratory synthesizing and measuring battery anode materials.

Indra Subedi is a Postdoctoral researcher at University of Toledo. He received his M.Sc in physics from Tribhuvan University, Kathmandu, Nepal. He obtained both M.S. & Ph.D. degree in physics from the University Toledo, Toledo, OH in year 2016 & 2019 respectively. His current research interest includes fabrication of thin film devices and determination of optical properties of thin films, solar cells, and bulk material by using spectroscopic ellipsometry and used them in devices simulation for optoelectronic devices like solar cells.

Rasha Awni is a postdoctoral researcher at the Wright Center for Photovoltaics Innovation and Commercialization in the Department of Physics and Astronomy at The University of Toledo. She joined Prof. Yanfa Yan's group in 2016. She received her Ph.D. degree in physics from The University of Toledo, USA, in 2020. Her research is focused on understanding defects in II-VI compounds and perovskite materials and electrical characterization, simulations, and modeling of photovoltaic devices.

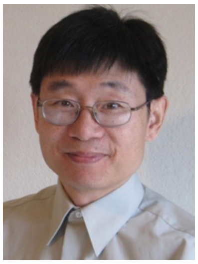
Dr. Ebin Bastola obtained his Ph.D. in Physics from the Center for Photovoltaics Innovation and Commercialization (PVIC), The University of Toledo, Toledo, OH. He is interested in solution-processing of the earth abundant nanomaterials including synthesis, characterization, and device fabrication for photovoltaic application. His current research primarily focuses on cadmium telluride thin film and devices.



Zhaoning Song received his B.S. degree in physics from Xiamen University, China, in 2009 and his Ph.D. in physics from the University of Toledo in 2016. He is currently a research assistant professor at the University of Toledo. His research interests include thin-film photovoltaics, device physics, materials science, and nanomaterials for optoelectronic applications.

Deng-Bing Li received his B.S. degree in School of materials science and engineering from Nanjing Tech University in china in 2009. He received his Ph.D. degree in Institute of Solid state Physics, Chinese Academy of Sciences in 2014. Now he is working in Yanfa Yan's group as a postdoc in the University of Toledo. His research focuses on the fabrication and characterization of high efficiency inorganic photovoltaics.

Robert Collins has held the titles of NEG Endowed Chair of Silicate and Materials Science at the University of Toledo since 2004 and Distinguished University Professor since 2008. He received the Ph.D. degree in Applied Physics from Harvard University in 1982 and served as Professor of Physics and Materials Research at The Pennsylvania State University from 1992 through 2003. He is a National Science Foundation Presidential Young Investigator Awardee, a Fellow of the American Physical Society (APS), and a recipient of the William Fowler Award of the APS Ohio Chapter for his research on the optical physics of materials.



Yanfa Yan has been an Ohio Research Scholar Chair and Professor in the Department of Physics and Astronomy at The University of Toledo, since 2011. Previously, he was a Principal Scientist at the National Renewable Energy Laboratory. He earned his Ph. D. in Physics from Wuhan University in 1993. His expertize includes thin-film solar cell fabrication, defect physics of semiconductors, and nanoscale characterization of microstructures, interfaces, and defects in thin-film photovoltaic materials. He is a Fellow of the American Physical Society.

Nikolas J. Podraza is a Professor in the Department of Physics and Astronomy and Wright Center for Photovoltaics Innovation and Commercialization at the University of Toledo. He earned his PhD in Physics from the University of Toledo and his MS and BS in Physics from the Pennsylvania State University. His research interests include optical properties of materials, spectroscopic ellipsometry, thin film deposition, nanostructured materials, and opto-electronic devices including microbolometers and photovoltaics.

Michael Heben is the McMaster Chair for Photovoltaics, the Managing Director of the Wright Center for Photovoltaics, and a Professor of Physics at the University of Toledo. He earned a BS in Physics from John Carroll University, an MS in Materials Science and Engineering from Stanford, and a PhD in Chemistry from Caltech. He was a postdoc and later a Principal Scientist and Group Leader over an 18 year career at NREL, prior to coming to Toledo in 2008. His interests include the science, manufacturability, and economics of PV materials and devices, and issues relating to building-DER-grid integration.

Randy Ellingson, Professor of Physics and faculty member of the University of Toledo's Wright Center for Photovoltaics Innovation and Commercialization, studies photovoltaic materials, devices, and systems. Research methods include ultrafast and steady-state optical spectroscopies, synthesis and solution-processing of nanocrystalline and quantum-confined semiconductors, device modeling, and PV system modeling and energy production forecasting. He received his B.A. in Physics from Carleton College and his Ph.D. degree in Applied Physics from Cornell University. After 14 very enjoyable years at the National Renewable Energy Laboratory (NREL) in Colorado, he joined the University of Toledo in 2008 to focus on the application of advanced materials and characterization to commercially relevant solar energy technologies. Additional interests include bringing the benefits of clean solar electricity to the underserved communities in and around Toledo by working with the charitable non-profit organization, Glass City Community Solar.