Highly Efficient Monolithic Perovskite Silicon Tandem Solar Cells: Analyzing the Influence of Current Mismatch on Device Performance

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Metal halide perovskites show great promise to enable highly efficient and low cost tandem solar cells when being combined with silicon. Here, we combine rear junction silicon heterojunction bottom cells with p-i-n perovskite top cells into highly efficient monolithic tandem solar cells with a certified power conversion efficiency (PCE) of 25.0 %. Further improvements are reached by reducing the current mismatch of the certified device. The top contact and perovskite thickness optimization allowed increasing the $J_{SC}$ above 19.5 mA cm$^{-2}$, enabling a remarkable tandem PCE of 26.0 %, however with a slightly limited fill factor (FF). To test the dependency of the FF on the current mismatch between the sub-cells, the tandems’ $J$-$V$ curves are measured under various illumination spectra. Interestingly, the reduced $J_{SC}$ in unmatched conditions is partially compensated by an enhancement of the FF. This finding is confirmed by electrical simulations based on input parameters from reference single junction devices. The simulations reveal that especially the FF in the experiment is below the expected value and show that with improved design we could reach 29 % PCE for our monolithic perovskite/silicon tandem device and 31 % PCE if record perovskite and silicon cells single junctions could be combined in tandem solar cells.

Introduction

The well-established technology of silicon solar cells dominates the photovoltaic market. With a current record power conversion efficiency (PCE) of 26.7 % on interdigitated back contacted silicon heterojunction solar cells (SHJ),\textsuperscript{1,2} silicon solar cells are approaching their theoretical efficiency limit of 29.4 %.\textsuperscript{3} To exceed this limit significantly, multiple absorbers with different band gaps can be combined into a multijunction solar cell architecture to exploit the solar light more efficiently than a single junction. Due to excellent optoelectronic quality in polycrystalline films, tunable optical band gap and potentially low-cost fabrication, metal halide perovskites are promising candidates for tandem applications.\textsuperscript{4–7} Besides low band gap Sn-based perovskites\textsuperscript{8,9} and Cu(In,Ga)Se$_2$ (CIGS)\textsuperscript{10–12}, crystalline silicon cells are well suited for tandem integration with perovskite absorbers. The latter combination is the most extensively studied perovskite-based tandem technology, with several groups reporting power conversion efficiencies above 25 %,\textsuperscript{5,4–7} a certified record PCE of 28 %,\textsuperscript{13} and predicted efficiencies above 30 %.\textsuperscript{14–18}

The monolithic integration of a perovskite top cell on a silicon bottom cell is challenging due to material and processing restrictions. So far, mostly silicon heterojunction (SHJ) bottom cells are utilized due to the well-passivated c-Si wafer surface which leads to high open circuit voltages ($V_{OC}$)\textsuperscript{4,5,7,19,20}. Recently, the p-i-n architecture for perovskite top-cells prevailed over the n-i-p architecture, especially due to temperature limitations of the SHJ cell (200 °C), which prevents the use of high temperature process, such as sintering of mesoporous TiO$_2$.\textsuperscript{21–23} Although there are possibilities to deposit the n-type contact at lower temperatures,\textsuperscript{24} and use temperature stable bottom cells,\textsuperscript{25,26} strong absorption of the p-type top contacts was reported for n-i-p architectures.\textsuperscript{14,27} An efficient device design was presented by Bush \textit{et al.}, who mitigated these losses by utilizing a p-i-n top cell architecture with reduced parasitic absorption in the n-type top contact. By implementing an ALD SnO$_2$ buffer layer in the top contact, the sensitive underlying layers were protected from damage induced by the subsequent sputter deposition of the top electrode, enabling a PCE of 23.6 %.\textsuperscript{28} Later on, Sahli \textit{et al.} used the same polarity and a similar top contact and demonstrated the use of a hybrid sequential fabrication enabling a conformal growth of the perovskite on top of a textured silicon bottom cell. The reduced reflection in the tandem solar cell led to a short circuit current density ($J_{SC}$) of 19.5 mA cm$^{-2}$ and a certified PCE of 25.2 %.\textsuperscript{14} At the same time, a tandem cell with planar front side was certified
with similar PCE of 25.2%, also using p-i-n top cells and implementing a n-type nc-SiO$_2$:H interlayer that increases light in-coupling into the Si bottom cell, resulting in a $J_{SC}$ above 19 mA cm$^{-2}$. By grain engineering and additionally adjusting the band gap of the perovskite absorber, a PCE of 25.4% was achieved in the p-i-n top cell configuration by Chen et al. in 2018.\textsuperscript{3} Recently, a PCE of 25.5% was published along with detailed analysis of the influence of textured interfaces, depending on their position in the cell stack, using a well-developed tandem solar cell and an anti-reflective foil attached on top of the cell.\textsuperscript{5} Recently, Oxford PV published a certified PCE of 28% for perovskite/silicon tandem solar cell, however, without giving any details about the materials and the structure used.\textsuperscript{6}

Despite these impressive efficiency improvements of monolithic perovskite/silicon tandem solar cells, the experimentally realized efficiencies are still far behind the predicted maximum values.\textsuperscript{5,14,15} In addition, there is still insufficient insight into how the individual sub-cells influence each other. One example is dependency of the fill factor (FF) on the difference between the $J_{SC}$ of the sub-cells (in the following: mismatch $m$). This effect has already been investigated for other tandem technologies, such as GaInP/GaAs or a-Si:H/a-Si:H, showing a mismatch dependent FF and emphasizing the need of power matching (i.e. same $J_{SC}$ for both sub-cells) instead of current matching (i.e. same $J_{app}$ for both sub-cells).\textsuperscript{30–33} The issue was also theoretically addressed for perovskite/silicon tandem cells,\textsuperscript{18} but for this device layout, detailed investigations and experimental results are lacking. Despite the difference between power and current matching conditions, the latter can be used as a first approximation for maximum power and is easier to extract from standard EQE measurements. The current matching conditions can be affected during outdoor operation by temporal and weather spectral changes. A tandem cell being current matched at AM1.5G illumination does not necessarily lead to the highest energy yield over time.\textsuperscript{30} Furthermore, different degradation of the sub-cells could lead to a varying mismatch over time. Therefore, it is important to analyze and understand the performance of a monolithic tandem solar cell as a function of the current mismatch.

**Results and discussion**

In this work, the development of an optically and electrically optimized n-type front contact with reduced reflection and parasitic absorption is demonstrated. This is achieved by the following adjustments: first, a good balance between conductivity and transparency of the indium zinc oxide (IZO) top electrode is found by fine-tuning the oxygen concentration during sputtering. Second, a SnO$_2$ buffer layer that is deposited via ALD, enables a good electron-selective contact for the p-i-n top cell. This top contact optimization leads to a certified PCE of 25.4% at a high FF approaching 80%, but with highly unmatched current densities of the sub-cells. Therefore, it is important to analyze and understand the performance of a monolithic tandem solar cell as a function of the current mismatch.

\textsuperscript{19} mA cm$^{-2}$, the perovskite absorber, and the IZO front electrode are fine-tuned to achieve photocurrents well above 19 mA cm$^{-2}$. In addition, reducing the ALD processing temperature increases the FF in the perovskite top cell. With both optimizations, a stabilized PCE of 26.0% is achieved which is higher than the highest reported two-side contacted c-Si single junction PCE.\textsuperscript{1,34} Further reduction of the front IZO thickness enabled a $J_{SC}$ of 19.77 mA cm$^{-2}$ for the limiting sub-cell and a cumulative photocurrent $I_{ParaSi} > 40$ mA cm$^{-2}$, even for planar front sides. In addition, we investigate the behavior of the highly efficient monolithic tandem solar cell as a function of current mismatch. Varying the incident spectrum using a LED based sun simulator, we show that the FF of the tandem solar cell is significantly affected by the current mismatch when reducing or enhancing the intensity of the blue wavelength range of incident light. The FF reaches its minimum when the sub-cells are close to current matching. This is highly important for precise energy yield analysis as the FF enhancement under non-matching conditions mitigates the PCE loss that would be expected on the basis of $J_{SC}$ loss.\textsuperscript{18} Electrical simulations validate the change in FF by using parametrized single junction reference parameters with a single diode equivalent circuit. The simulations predict a higher FF than obtained experimentally in our tandem solar cell, which highlights that further understanding and optimization of the recombination contact is needed to achieve higher efficiencies. In our case, the ideal serial connection would lead to a PCE of 29%. In addition, over 31% could be realized if performance metrics from record perovskite and silicon cells single junctions could be combined in a tandem solar cell.
Figure 1a and b display a stack of a typical monolithic perovskite/silicon tandem solar cell used in this work. The cross sectional SEM image shows the textured backside of the bottom cell (lower panel) and the top cell (upper right panel) recorded with the in-lens detector. The image obtained with the energy selective backscattered (EsB) detector (upper left panel) especially highlights the PTAA layer, which is not resolvable with the in-lens detector. As bottom cell, a rear-junction Shi solar cell with a textured rear side and a planar front side is used. To improve light in-coupling into the bottom cell, n-doped nc-SiO\textsubscript{x}H is utilized to create an electron-selective contact with proper refractive index interfacing with a 20 nm thin indium tin oxide (ITO) as recombination layer to interconnect both sub-cells. The perovskite top cell with p-i-n architecture is utilized in the following layer sequence with the light entering the top cell (upper panel) and back side of the bottom cell (lower panel) of a typical monolithic tandem solar cell used in this work. The perovskite top and silicon bottom cell, respectively. This displays the J-V characteristics reported by the certification lab; a PCE of 25.0 % was measured, stable during 400s of maximum power point power tracking (see Figure S3). The device exhibits virtually no hysteresis and the deviation between the maximum power point parameters obtained from the MPP tracking and those from J-V measurement is negligible. The Voc of 1.78 V is close to the cumulative sub-cell Voc\textsubscript{s} as expected from single junction devices and to the Voc\textsubscript{s} found recently with similar device architectures.\textsuperscript{6,7} A FF of 78.6 % is one of the highest measured for a perovskite/silicon tandem solar cell and will be discussed in more detail below. On the other hand, a Jsc of 17.81 mA cm\textsuperscript{-2} indicates one of the limiting factors of the certified tandem solar cell. This is further confirmed by the EQE measurements, presented in Figure 1d along with the total reflectance of the device presented as 1-R. The photocurrent densities are J\textsubscript{ph} \approx 20.69 mA cm\textsuperscript{-2} and Jsc = 17.85 mA cm\textsuperscript{-2} for the perovskite top and silicon bottom cell, respectively. This shows that the certified tandem device is strongly current-mismatched with the silicon sub-cell being the limiting one. Although the Jsc of a tandem solar cell can be higher than the minimum Jsc\textsubscript{cc} of the sub-cells,\textsuperscript{41,42} for good solar cells the tandem Jsc is expected to be very close to the minimum Jsc\textsubscript{cc} of the sub-cells (limiting sub-cell). This is valid when the limiting sub-cell (here: silicon) has a high shunt resistance. Assuming this, the integrated current density Jsc of 17.85 mA cm\textsuperscript{-2} measured in-house is in very good agreement with the certified Jsc of 17.81 mA cm\textsuperscript{-2} measured at Fraunhofer ISE. The mismatch m
between $J_{\text{pero}}$ and $J_S$ is more than 2.8 mA cm$^{-2}$, which needs to be reduced in order to achieve higher PCEs. Figure 1d displays the sum of the EQEs (black line) and the parasitic losses as the blue area between the sum and 1-R. The parasitic losses are high in the UV and near infrared (NIR) wavelength range and are below 1% on average between 550 nm and 1000 nm. The integrated reflection losses amount to an equivalent photocurrent of 4.65 mA cm$^{-2}$ in the complete wavelength regime; between 750 nm and 1050 nm, there are distinct reflection features with 1.9 mA cm$^{-2}$ reflection losses only in this region. In order to extract parasitic losses for each individual layer, an optical simulation was carried out using GenPro4 (Figure S4).44 We find, that in the UV region, mostly IZO, SnO$_2$ and C$_6$H$_{12}$O$_6$ absorb light, whereas in the infrared region the photons are absorbed by the IZO at the front side, by the recombination ITO, and by the aluminum doped zinc oxide (AZO) and silver (Ag) at the backside of the cell. The rather high reflection losses and the strong current mismatch indicate that further optical optimizations are required.

In order to further improve the tandem solar cell, a lower temperature of the ALD SnO$_2$ process was first considered. Exposing the perovskite for 1 h to 100 °C (in addition to the perovskite annealing) might reduce the perovskite top cell performance and, thus limiting the tandem efficiency.44,45 Therefore, we analyze the SnO$_2$ layer in terms of optical properties as a function of deposition temperature. Figure 2a shows that changing the deposition temperature has an effect on the optical properties of the deposited layer, as evidenced by the shift in extinction coefficient and refractive index. At higher temperatures, a higher absorption in the UV is measured and the absorption onset becomes steeper. Simultaneously, the refractive index increases. However, by using optical simulations including the measured differences in optical data, we find that changing the deposition temperature in the range of 80 °C to 200 °C does not have a major effect on $J_{\text{pero}}$ and $J_S$ in our tandem device (Figure S5a). For higher deposition temperatures, the EQE of the perovskite improves for wavelengths above 370 nm but also reduces below 370 nm as the absorption of the SnO$_2$ increases in that wavelength range. Overall, the cumulated current density $J_{\text{pero}+\text{Si}}$ is constant in the practically relevant range of 80 °C to 120 °C (Figure S5b). More importantly, changing the deposition temperature can have a strong effect on the electrical performance. Therefore, semitransparent perovskite solar cells with SnO$_2$ deposited at moderate temperatures of 80 °C, 100 °C and 120 °C are fabricated and analyzed. The results, depicted in Figure 2b and c, indeed show a beneficial effect on the FF by reducing the temperature from 120 °C to 80 °C, which directly transfers to the change in PCE. The $V_{OC}$ and $J_{SC}$, however, are hardly affected by the different deposition temperatures of the ALD SnO$_2$ (see Figure S5a and b).

Additionally, the thickness of the top TCO in the tandem stack, here IZO, has a significant influence on parasitic absorption throughout the entire wavelength range. To analyze the impact of top electrode thickness, an optical simulation of the tandem stack with thicknesses of IZO between 60 nm and 130 nm in 10 nm steps is performed. Figure S7 shows the simulated gain in current density, which amounts to $\sim$0.32 mA cm$^{-2}$ when reducing the thickness from 130 nm to 90 nm. Further reducing the IZO thickness would be ideal to increase $J_{\text{pero}}$ and $J_S$. However, reducing the thickness also leads to a higher sheet resistance and might have a negative impact on the cell performance, especially reduction of FF. Therefore, 90 nm was used in the optimized device.

Apart from being the electron-selective contact layer for the bottom SHJ solar cell the use of n-type nc-SiO$_2$-H between the perovskite and silicon absorber has two benefits: First, the refractive index at 633 nm of around 2.7 is in between those of the neighboring layers, which reduces reflection; second, this reflection can be spectrally tuned when the layer thickness is adapted for destructive interference in a particular wavelength range. For the n-type nc-SiO$_2$-H, a thickness of around 95 nm was found both in simulations and experimental results to be ideal for monolithic perovskite/silicon tandem solar cells.15,20 Thus, this thickness is implemented in the further development. Finally, the thickness of the perovskite absorber is fine-tuned by adjusting the spin coating speed to enable conditions closer to current matching.47 Figure S8 presents optical simulations with varying perovskite thickness between 390 nm and 520 nm. While $J_{\text{pero}}$ hardly changes, a thinner perovskite absorber layer will transmit more light into the silicon bottom cell, enhancing the $J_S$. As the sub-cell with the lower current approximately determines the $J_{SC}$ of the tandem cell, a maximum tandem $J_{SC}$ is expected for a perovskite thickness of around 470 nm for our design.

All above described optimizations are implemented into tandem solar cell devices and the results are shown in Figure 3. Indeed, the optical losses are reduced and the sub-cells enable short circuit current densities closer to current matching. The $J_{SC}$ improved by 1.4 mA cm$^{-2}$ to a value of 19.22 mA cm$^{-2}$. The $V_{OC}$ remained almost unchanged (1.77 V) while the FF is slightly lower (76.6%). Overall, a stabilized PCE of 26.0% is achieved, confirmed by a 5 minute MPP-track as shown in the inset in Figure 3a. Figure 3b highlights the EQE spectra together with total reflectance depicted as 1-R for the optimized tandem design. Due to improved optics, interference patterns in the 800 nm to 1050 nm wavelength range are smoothed,
For testing the stability of these optimized tandem devices, a device was stored in ambient air and light conditions (i.e. 19-21 °C and 50-90% RH) and regularly MPP-tracked for at least 1000 h of ambient storage. Compared to the device shown in Figure 1c and d, the improved device is much closer to current matching, with only 0.9 mA cm$^{-2}$ mismatch. In Figure S9, EQE spectra of similarly fabricated single junction silicon and perovskite cells are shown. For testing the stability of these optimized tandem devices, a similar tandem solar cell with comparable performance as shown in Figure 3 was glass/glass encapsulated with edge-sealant and stored in ambient conditions. Due to different optics after encapsulation, the PCE dropped from 26.0 % to 24.6 % mostly because of additional reflection from the air/glass interface, as compared to air/LiF. During 1000 h, the device was stored in ambient air and light conditions (i.e. 19-21 °C and 50-90% RH) and regularly MPP-tracked for at least 30 min at each data point. The measurements were performed at 25 °C, ambient humidity and under full simulated, 1 sun AM1.5G illumination. The exact spectrum is shown later in Figure 4a. The PCE as function of time is presented in Figure S10 and found to be remarkably stable with a relative PCE drop of less than 1 % for the last measurement after 1000 h of ambient storage.

As the optimized tandem device in Figure 3 still shows current mismatch and the $J_{\text{Pero}}$ stays below 40 mA cm$^{-2}$, the optical benefit of further reducing the front IZO thickness is tested. As described above, Figure S7 shows simulation results for thinner front IZO. A $J_{\text{Pero}}$ above 40 mA cm$^{-2}$ is only realized with IZO thicknesses of around 60 nm. Consequently, a tandem solar cell with this thin IZO layer is fabricated and the experimental results are presented in Figure S11. Integrated current densities from the EQE spectra up to 20.27 mA cm$^{-2}$ for the top and 19.77 mA cm$^{-2}$ for the bottom cell are measured, which indeed results in a $J_{\text{Pero}}$ of more than 40 mA cm$^{-2}$. This current density is comparable to values reported by Sahli et al. for a fully textured tandem cell but is still lower than the best $J_{\text{Pero}}$ of silicon single junction cells (42.87 mA cm$^{-2}$). If the $J_{\text{Pero}}$ would be equally distributed between $J_{\text{Pero}}$ and $J_{Si}$, the tandem cell with thin IZO would exceed a $J_{SC}$ of 20 mA cm$^{-2}$, a remarkable result for a planar front side tandem design. Due to a lower FF, which is mainly a result of lower shunt resistance and slightly higher series resistance - the latter due to higher ohmic losses in thinner IZO - the stabilized PCE during MPP-tracking is only 25.3 % (see Figure S11). For the optimal performance with thin IZO, metal fingers are necessary and to optimize the resistive versus shading losses, opto-electrical simulations are required, which is a subject of future work.

Comparing the two tandem solar cells, presented in Figure 1 and Figure 3, we observe a lower FF for the device that operates closer to current matching. Besides the possibility of a cell-to-cell variation, operating the sub-cells under strong non-matching conditions should improve the tandem FF, as reported for other tandem technologies. As a thorough understanding of the influence of non-current matching conditions is important for a) device understanding and b) energy yield analysis, the FF of the best performing tandem solar cell shown in Figure 3 is measured here for various illumination spectra, leading to non-matching conditions. Using a LED-based sun simulator, the intensity of individual LEDs can be adjusted while leaving the others unchanged. Thus, the device can be measured under different non-matching conditions. As basis, the simulated AM1.5G spectrum is used. To increase or decrease the irradiance in the blue region, the intensity of two blue LEDs (center emission peaks at 420 nm and 440 nm) is changed, while leaving all other LEDs to simulate the AM 1.5G spectrum constant. This way, only the current generation in the perovskite top cell is affected. Figure 4a shows the utilized spectra including the AM1.5G reference spectrum. The measurement series starts with the highest intensity of blue light well above the blue intensity in AM1.5G spectra. Then the power of the two blue LEDs is decreased and a tandem J-V-curve for each spectrum is measured. The raw data of the 26 measurements are presented in Figure S12, plotted against the spectrum (measurement) number. The first and last J-V curve in the series are measured under AM1.5G illumination and ensure that there is no degradation of the cell and no drift of the spectrum during the course of the series. As the intensity of the blue light (i.e. current generation in the perovskite) decreases,
the hysteresis of the tandem cell starts increasing, revealing that the hysteresis of the perovskite sub-cell is influenced by current mismatch. Interestingly, the hysteresis is decreased after the series is done. However, in the following, only the \( V_{OC} \) to \( J_{SC} \) (reverse scan) measurements will be considered. The currents of the sub-cells are calculated by integrating the sub-cell EQE multiplied by the measured spectra. From these, the mismatch \( m = J_{Si} - J_{Pero} \) is calculated as the difference between \( J_{Si} \) and \( J_{Pero} \). Figure 4b, c and d show the tandem \( J_{SC} \), FF and PCE as a function of mismatch. Due to the logarithmic dependence on \( J_{SC} \), the \( V_{OC} \) is only marginally affected by the spectrum change (Figure S12). As can be seen from the comparison of Figure 4a and b, a stronger photogeneration in the blue wavelength range, i.e. a higher \( J_{Pero} \), does not affect the tandem \( J_{SC} \), as this metric is limited by the unchanged bottom cell current. This confirms that the silicon sub-cell is limiting for the AM1.5G illumination conditions (see Figure 4b) and high intensities of blue light, as seen in the EQE measurement shown in Figure 3b. The blue intensity is lower than in the AM1.5G spectrum, the tandem cell becomes top cell limited, where the \( J_{SC} \) is reduced in accordance to the reduced intensity (see positive current density mismatch values \( m = J_{Si} - J_{Pero} \) in Figure 4b).

Changing the spectrum and therefore changing the mismatch does not only have an effect on the current density, but also on the FF (Figure 4c). It is typically reported that the FF is the lowest when the tandem solar cell is operated close to the current matching point,\(^{30,33,47} \) but this is not necessarily exactly at the matching point and depends on the individual performance of the sub-cells.\(^{31} \) Interestingly, the FF minimum occurs here when the silicon bottom cell is the limiting sub-cell. With a slope of \(-1.31 \% \mathrm{FF per \ mA cm}^{-2} \) mismatch, a FF of 79 % is expected for a mismatch of \(-2.8 \ mA cm}^{-2} \), which is the mismatch value of the certified tandem cell discussed above. As this mismatch-predicted FF value is close to the FF of the certified cell, it strongly supports the assumption that the reduced FF after decreasing the photocurrent mismatch is mainly due to the device physics instead of a cell-to-cell variation. The benefit of ALD optimization and enhanced resistive losses from thinner front IZO counterbalance here. Although, the FF can change with the photocurrent due to e.g. reduced collection losses, we can exclude this effect to be dominant in our tandem solar cell as the FF of a similar fabricated cell does not vary with \( J_{SC} \) in the relevant photocurrent range utilized for mismatch analysis.\(^{6} \) Figure 4d shows the PCE of the tandem solar cell as a function of the current density mismatch. The intensity of each spectrum is calculated as stated in Supplementary note 3. Although the \( J_{SC} \) decreases for a mismatch \( m > -0.3 \ mA cm}^{-2} \), the PCE increases due to the increasing FF (colored data points). The quadratic fit of the PCE calculated with the measured FF reveals of the current density mismatch. The intensity of each spectrum is adjusted in order to get a spectrum with a higher or lower intensity in the blue wavelength range compared to the AM1.5G. b) shows the short circuit current density, fill factor and PCE of the tandem solar cell from Figure 4a as a function of the mismatch in calculated photogeneration currents between the sub-cells. Additionally, the power density is shown when assuming that the FF = FFmin and does not change with changing mismatch.
Furthermore, the PCE is shown for a constant FF = FF\textsubscript{min} with FF\textsubscript{min} being the minimum FF measured in the series (white data points). The comparison between the constant and non-constant FF highlights that the changing FF mostly compensates the drop in J\textsubscript{SC}, thus leading to a broader peak around the maximum PCE. For a mismatch of m = 1 mA cm\(^{-2}\), the PCEs differ by 0.49 % while for m = -1 mA cm\(^{-2}\) the PCEs differ by 0.26 % between constant and measured FF. The PCE of a slightly mismatched monolithic tandem solar cells suffers only marginally from the decreased J\textsubscript{SC}. This is highly important for energy yield analysis, especially when comparing monolithic 2-terminal with 4-terminal tandem solar cells. One of the arguments in favor of 4-terminal devices is to avoid decrease in performance due to current reduction if current mismatch occurs. However, we show that in 2-terminal, monolithic devices the FF increases with mismatch (see colored data points in Figure 4c). Thus, the drop in J\textsubscript{SC} is mostly compensated, leading to a PCE, which is less sensitive to current density mismatch. Assuming an ideal current redistribution with J\textsubscript{SC,matched} = 0.5 J\textsubscript{Pero} + 0.5 J\textsubscript{Si} = 19.73 mA cm\(^{-2}\) for both sub-cells in Figure 3b, using the measured values for FF\textsubscript{matched} and V\textsubscript{OC,matched} extracted for m = 0, a PCE of 26.6 % would be reached.

To validate the experimental results in Figure 4c, we perform electrical simulations using the electronic design automation software LTspice (for more details and parameters see Supplementary note 4).\(^\text{48}\) An equivalent circuit diagram consisting of two series connected single diode models was built as shown in Figure S14a. In the first step, we prove that the FF minimum is not necessarily at the current matching point for any kind of monolithic tandem solar cell. For this, two identical solar cells are connected in series in this simulation. When sweeping the J\textsubscript{Top} and J\textsubscript{Bottom} while maintaining a constant J\textsubscript{Top}/J\textsubscript{Bottom}, the FF indeed changes as a function of current mismatch (see Figure S14b, upper graph) with a FF minimum for current matching conditions. The same procedure is carried out for a reduced shunt resistance R\textsubscript{Sh} of either the top or the bottom cell. Now, the FF minimum is not in the current matching point anymore but shifts to the bottom cell limited side for a low R\textsubscript{Sh,bottom} and to the top cell limited side for a low R\textsubscript{Sh,top} (Figure S14b). Although the experimentally detected FF minimum is present for silicon limiting illumination conditions (Figure 4c), we do not expect to have a lower R\textsubscript{Sh} in the silicon bottom cell compared to the perovskite top cell. The difference between the simulation and experiment is discussed below. The bottom graph in Figure S14b shows that a mismatch between J\textsubscript{Pero} and J\textsubscript{Si} might be necessary for achieving the highest power. As stated earlier in this paper, the highest power is achieved if both sub-cells have the same J\textsubscript{MPP}. The necessity of current matching depends on the shunt resistance and with that overall of the individual sub-cells (i.e. the difference between J\textsubscript{SC} and J\textsubscript{MPP}). If the difference between J\textsubscript{SC} and J\textsubscript{MPP} of both sub-cells is similar, J\textsubscript{SC}-matching leads to the highest power output. If the difference between J\textsubscript{SC} and J\textsubscript{MPP} is unequal for both sub-cells, a J\textsubscript{SC}-mismatch is needed to achieve J\textsubscript{MPP}-matching and with that, the highest power output. In a second step, we attempt to reconstruct the measured tandem J-V-characteristics using electrical simulations. For this, single junction silicon and perovskite (opaque) cells are fabricated with similar device layout as in our tandem solar cell and parametrized using a single diode model. Figure S15a and b show the measured J-Vs at different intensities of the single junction solar cells as well as the simulated J-Vs. We use the parameters of the fitted single junction cells to reconstruct the tandem solar cell and simulate the experiment. Solely the series resistance R\textsubscript{s} and saturation current of the perovskite J\textsubscript{SC} are adjusted slightly to match the tandem conditions (see Supplementary note 4). The simulated J\textsubscript{SC} and FF as a function of mismatch m are shown in Figure S15c and Figure S15d and reveal a higher and shifted FF minimum but a similar variation of J\textsubscript{SC}. Using the FF, V\textsubscript{OC} and J\textsubscript{SC} from simulated results would lead to a PCE of 27.6 % for AM1.5G illumination for the tandem device presented in see Figure 3, mostly due to higher FF in simulation. Comparing the J-Vs with most negative and most positive mismatch, the difference between the simulated and experimental measured J-Vs appears to be just around the MPP (Figure S15e). For unmatched sub-cells, charge carriers accumulate at the recombination contact. This could affect the recombination behavior. Moreover, the electric fields and with this, the charge collection of the sub-cells is affected by the charge accumulation. This might also be the reason for the dependency of the hysteresis on the mismatch. Additionally, the recombination layer might behave non-ohmic, thus another electrical component needs to be implemented in the simulation. Further investigation is needed to understand the device physics in more detail, to exactly reconstruct the monolithic tandem solar cell in an electrical simulation and with this, increase the PCE. If we assume an ideal contact design in the tandem solar cell for example by implementing grid fingers, leading to a series resistance of R\textsubscript{s} = 0 Ω cm\(^{-2}\)(for this, we additionally assume a negligible series resistance of the bulk), the simulated tandem device could reach a PCE of 29 % (Figure S16b). Here the J\textsubscript{Pero} is maintained constant at 39.46 mA cm\(^{-2}\), the same J\textsubscript{Pero} as for the device shown in Figure 3. Furthermore, we simulate a monolithic tandem solar cell by using electrical parameters from record p-i-n perovskite\(^\text{49}\) and both side contacted c-Si cells\(^\text{34,50}\). By neglecting the series resistance, which mostly stems from the perovskite sub-cell, the FF increases and therefore a maximum PCE of 31.0 % is expected due to improved V\textsubscript{OC} (Figure 14d).

Conclusions

In summary, we implemented a transparent n-type top contact layer stack in monolithic silicon/perovskite tandem solar cells in order to achieve a certified PCE of 25.0 % at highly unmatched photocurrents. Further improvements of the rear junction silicon bottom cell with adjusted n-type nc-SiO\(_2\)H layer thickness, the n-type top cell contact with proper ALD SnO\(_2\) deposition temperature and IZO thickness, as well as adjusted perovskite thickness led to a remarkable tandem PCE of 26.0 %. Further reducing the front IZO thickness allowed J\textsubscript{SC} over 19.7 mA cm\(^{-2}\) and cumulative current densities J\textsubscript{Pero} over...
40 mA cm$^{-2}$ in tandem cells with a planar front side. As the FF was found to depend on the mismatch condition $m = J_{sc}/J_{phmax}$, the sub-cell mismatch for the best tandem device was analyzed by varying the illumination spectrum. We show that the FF improves under unmatched current conditions of the tandem solar cell and that the loss in tandem PCE due to lower $J_{sc}$ at unmatched conditions is less pronounced as it would be with a constant FF. This dependence is highly important for energy yield analysis, especially when comparing 2-terminal and 4-terminal tandem solar cells. Electrical simulations based on input parameters from reference single junction devices parametrized with a single diode model confirmed the increasing FF for unmatched monolithic tandem solar cells. The simulation of the tandem performance shows that the FF in our experimentally realized device is lower as the FF expected from simulations, leaving room for improvement. An ideal series resistance can lead to a PCE of 29% for our tandem architecture. The simulation of a monolithic tandem solar cell with record perovskite and silicon cells from literature and a negligible series resistance reveals a PCE of 31%. Therefore, our work provides a detailed device understanding under different current matching conditions. This is mandatory to improve the PCE potential of silicon/ perovskite tandem solar cells to predicted values above 30%.

Materials and methods

Perovskite materials. Anhydrous DMSO (dimethyl sulfoxide), DMF (dimethylformamide), and toluene were purchased from Sigma Aldrich. PTAA (poly (bis (4-phenyl) (2,5,6-trimethylphenyl) amine)), $C_60$ (purity = 99.9 %) and lithium fluoride (purity $\geq$ 99.99 %) purchased from Sigma Aldrich. FAI (formamidinium iodide) and MABr (methylammonium bromide) were purchased from Dynamo. PbI$_2$ and PbBr$_2$ were bought from TCI. CsI was purchased from abcr GmbH. The Tetrakis(dimethylamino)tin(IV) (TDMASn) was used as the Sn precursor and was held at 60 °C in a stainless steel container. Water was used as oxidant, and was delivered from a stainless steel container without intentional heating, whereas the precursor delivery manifold was heated to 115 °C. For the deposition at 100 °C, the TDMASn/purgel1/H$_2$O/purge2 times are 1s/7.5s/0.2s/10s with corresponding nitrogen flows of 30sccm/90sccm/90sccm/90sccm. With this, 167 cycles leads to 20 nm tin oxide. For the deposition at the lower temperature of 80 °C, the purging times were slightly increased (i.e. times of 1s/10s/0.2s/15s) to account for less efficient purging of reactants and their reaction products at lower temperatures.

140 cycles were needed to deposit 20 nm at 80 °C. IZO was sputtered in a Roth&Rau MicroSys 200 PVD. The 2 inch ceramic target consisted of 90% In$_2$O$_3$ and 10% ZnO. A RF-power of 70 W the cells oscillated under the target to have a uniform deposition. To add additional oxygen to the deposition chamber, a mix of Ar and $O_2$ is added. The added amount of oxygen ranges from 0.0 % to 0.25 % while the total gas flow is the same for all depositions. A 150 nm thick Ag metal film was evaporated through a shadow mask as a top contact. Finally, 100 nm LiF was evaporated to serve as an anti-reflective coating.

Silicon solar cell preparation: The silicon heterojunction (SHJ)-bottom cell was fabricated on a 260 µm thick polished FZ <100> n-type crystalline silicon (c-Si) wafer in a rear junction configuration. The front surface of the wafer was left polished in order to facilitate the perovskite top cell deposition, while the rear surface of the wafer was wet-chemically textured to obtain random pyramid with <111> facets in order to improve the optical response of the bottom cell in the NIR region. After a final RCA clean and a 3 minute HF dip (1 % dilution in water) to strip the SiO$_2$ of the surface, a 5 nm thick, intrinsic (i) amorphous silicon (a-Si:H) layer was grown on both sides of the c-Si wafer in order to passivates the c-Si surface. On the textured back-side, a 5 nm thick, p-doped a-Si:H was deposited on the rear passivating layer to form the junction of the SHJ cell. On the polished front-side, a 20 nm thick, n-doped nanocrystalline silicon oxide layer (nc-SiO$_x$:H) with a refractive index, $n$, of 2.7 at 633 nm was used as a front surface field (FSF) of the SHJ bottom cell and intermediate layer between the top and the bottom cells. For the second optimization, this thickness was increased to 95nm. All the a- and nc-Si layers were deposited with an Applied Materials (AKT1600) plasma enhanced chemical vapor deposition (PECVD) tool. In order to contact the bottom cell a ZnO:Al/Ag layer stack was deposited on the textured back-side and a 20 nm thick ITO layer was deposited on the polished front-side on top of the nc-SiO$_x$:H interlayer, both depositions were DC-sputtered in an in-line sputtering tool from Leybold Optics.

$J_{sc}$...
The contact layers of the silicon were deposited using shadow masks with an opening of 1 x 1 cm².

**Silicon single junction solar cell preparation:** To fabricate a silicon single junction, 80 nm IZO is additionally deposited on the 20 nm ITO of the bottom cell in order to reduce the sheet resistance. As for the tandem solar cell, a metal frame is evaporated as a top contact.

**Semitransparent perovskite single junction solar cell preparation:** The semitransparent solar cells are manufactured as described in the section of the perovskite solar cell preparation on silicon bottom cells. Instead of the silicon bottom cell, an ITO coated glass substrate (25×25 mm, 15 Ω sq⁻¹, patterned by Automatic Research GmbH) was used and cleaned sequentially for 15 min with Acetone, Mucasol (2 %vol in Water), DI-water and isopropanol in an ultrasonic bath. Before the deposition of the HTM, the samples were treated in an UV-ozone cleaner for 15 min.

**Tin oxide layers for optical characterization:** For optical characterization, tin oxide was deposited on a silicon wafer coated with 450 nm silicon oxide. For the deposition temperature of 80 °C, the parameters are mentioned in the section “perovskite solar cell fabrication”. For all other temperatures, the pulse- and purge times and nitrogen flows are equal to the deposition of tin oxide at 100 °C.

**Device characterization:** The current density–voltage (J-V) measurements were performed under standard test conditions (25°C, LED based sun simulator from Wavelabs, class AAA), adjusted with a non-filtered calibrated silicon reference cell (Fraunhofer ISE). The scan rate was 0.25 V s⁻¹ with a voltage increment of 20 mV. The external quantum efficiency (EQE) was measured as a function of wavelength from 300 nm to 1200 nm with a step of 10 nm using a home built small spot QE system. The beam size is 2×5 mm², thus smaller than the active area. When measuring perovskite top cell, infrared (850 nm) bias light was applied along with 0.6 V bias voltage to ensure a measurement in short circuit conditions. Additionally, the cell is slightly illuminated with blue (455 nm) light that partially increases the signal to noise ratio. When measuring silicon bottom cell, blue (455 nm) bias light was applied. The EQE of the silicon bottom cell is not affected by bias voltage, as the shunt resistance of the silicon cell is very high. Reflection was measured as a function of wavelength from 300 nm to 1200 nm with a step of 5 nm using an integrating sphere with a Perkin Elmer Lambda – 1050 UV/VIS/NIR spectrophotometer, calibrated with a white Spectralon. The optical properties of ALD SnO₂ were evaluated by spectroscopic ellipsometry. Measurements were performed using a Sentech SE 850 at angles of incidence 50°, 60° and 70°. The dielectric function was modeled using Tauc-Lorentz oscillators to account for the absorption for energies above the band gap.

**Conflicts of interest**

There are no conflicts to declare.

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**Author contributions**

E.K., M.J., P.T., A.A. and Lu.K. developed and optimized the perovskite solar cell fabrication process. E.K. fabricated the tandem devices, performed the measurements and optimized the top contact. A.B.M.V., La.K. and B.S. developed and optimized the silicon-heterojunction solar cell fabrication process and fabricated bottom cells. B.M. and E.K. optimized the SnO₂ process and performed and analyzed spectroscopic ellipsometry measurements. E.K., P.T. and A.A. performed the optical and electrical simulations. All authors participated manuscript writing and proofreading as well as correcting. B.R., R.S., B.S. and S.A. initiated and supervised the project.

**References**


We present a highly efficient monolithic perovskite/silicon tandem solar cell and analyze the tandem performance as a function of photocurrent mismatch with important implications for future device and energy yield optimizations.