Nano-optical designs for high-efficiency monolithic perovskite–silicon tandem solar cells

Perovskite–silicon tandem solar cells offer the possibility of overcoming the power conversion efficiency limit of conventional silicon solar cells. Various textured tandem devices have been presented aiming at improved optical performance, but optimizing film growth on surface-textured wafers remains challenging. Here we present perovskite–silicon tandem solar cells with periodic nanotextures that offer various advantages without compromising the material quality of solution-processed perovskite layers. We show a reduction in reflection losses in comparison to planar tandems, with the new devices being less sensitive to deviations from optimum layer thicknesses. The nanotextures also enable a greatly increased fabrication yield from 50% to 95%. Moreover, the open-circuit voltage is improved by 15 mV due to the enhanced optoelectronic properties of the perovskite top cell. Our optically advanced rear reflector with a dielectric buffer layer results in reduced parasitic absorption at near-infrared wavelengths. As a result, we demonstrate a certified power conversion efficiency of 29.80%.

Monolithic two-terminal perovskite–silicon tandem solar cells (PSTSCs) have recently achieved power conversion efficiencies (PCEs) exceeding 31%, thus overcoming the physical limit of conventional crystalline-silicon single-junction solar cells. Such high PCEs were reached by continuous improvements of the optical and electronic properties of PSTSCs. These improvements include, amongst others, switching the cell polarity for enhanced top-contact transmission and fine-tuning of various layers to improve the optical performance. In addition, various publications have addressed the improvement of the electronic properties by optimizing contact layers, utilizing additives and adjusting the perovskite composition or deposition. Numerical studies underline the importance of adequate light management by introducing textured device interfaces for high PCEs. For silicon solar cells, potassium-hydroxide-etched random pyramidal textures with a size of several micrometres are commonly used for light management. However, they are not compatible without further adaptation with solution-processed perovskite absorbers, which result in the highest reported PCEs of perovskite single-junction solar...
In recent years, different approaches to implement light management textures in PSTSCs have been investigated, either by adapting the perovskite deposition technique but leaving the pyramidal texture unchanged24–26, or by adapting the textures such that perovskite solution-processing becomes feasible27–31 (Supplementary Fig. 1). These studies demonstrated remarkable optical improvements in PSTSCs and provided insights into the morphology and optoelectronic properties of textured perovskite top cells. However, it remains a major challenge to develop an appropriate texture, which is able to balance the persistent trade-off between electronic and optical performance of textured PSTSCs with solution-processed perovskite top cells. In recent studies we already introduced gentle sinusoidal nano-textures with submicrometre feature size as promising candidate for PSTSCs: optical simulations indicated that the PCE can be substantially enhanced compared to planar reference tandem solar cells32. We further demonstrated experimentally that such nano-textures provide a feasible light-management approach in both solution-processed perovskite33 and silicon34 single-junction solar cells, without compromising the optoelectronic quality of the respective absorber.

In this work, we present PSTSCs with a gentle sinusoidal nano-texture connecting the advantages of structuring the silicon surface while preserving the material quality of the solution-processed perovskite. We show that the nano-texturing causes a substantial reduction of reflection losses compared to their planar counterpart, strongly improves the fabrication yield enabled by the excellent film formation properties and increases the open-circuit voltage by 15 mV. To meet the challenge of parasitic absorption losses, we further implement a reflector with a dielectric buffer layer (RDBL) at the rear side of the silicon bottom cell. Combining both approaches, the gentle nano-texture at the front side and the reflector with dielectric buffer layer at the rear side of the silicon bottom cell, we demonstrate a monolithic PSTSC with an independently certified PCE of 29.80%.

**Tandem design**

Figure 1a–c shows cross-sectional scanning electron microscopy (SEM) images of the investigated PSTSC device configurations: the reference PSTSC device (Fig. 1a) features a fully planar front side (upper half) and a standard random pyramid texture at the rear side of the silicon heterojunction (SHJ) subcell (lower half). The reference solar cell is similar to that presented in our previous publications35 and consists of a SHJ solar cell with n-doped hydrogenated nanocrystalline silicon oxide (nc-SiO$_x$;H(n)), doped indium oxide as the transparent conducting oxide (TCO), a self-assembled monolayer (SAM) denoted Me-4PACz([4-(3,6-dimethyl-9H-carbazol-9-yl)butyl]phosphonic acid), a mixed-cation and mixed-halide perovskite absorber, lithium fluoride (LiF), C$_{60}$, tin oxide and indium zinc oxide. The perovskite composition is either Cs$_{0.05}$(FA$_{0.79}$MA$_{0.21}$)$_{0.95}$Pb(I$_{0.79}$Br$_{0.21}$)$_3$ or (Cs$_{0.05}$(FA$_{0.77}$MA$_{0.23}$)$_{0.95}$Pb(I$_{0.77}$Br$_{0.23}$)$_3$ with bandgap energies of 1.66 eV or 1.68 eV, respectively.

The sinusoidal nanostructure at the front surface of the silicon subcell (Fig. 1b,c) was manufactured by combining ultraviolet nano-imprint lithography, reactive ion etching and wet chemical etching36. The experimental procedure is detailed in Methods. The resulting nanostructure has a hexagonal lattice with a period of 750 nm and a peak-to-valley height of approximately 300 nm (Fig. 1d). This nano-texture can be completely covered by the perovskite film with a typical thickness of 500–600 nm (on planar surfaces) revealing a flat perovskite front surface on the C$_{60}$ side. At the rear side of the silicon wafer we applied a reflector with an RDBL31 (Fig. 1e). The RDBL comprises a SiO$_2$ buffer layer between the TCO and the silver back-reflector, reducing parasitic absorption losses. A silver grid covering 4% of the active area is screen-printed on top of the TCO before SiO$_2$ deposition to establish the electric contact between the TCO and Ag (Fig. 1f, right side).

**Perovskite film formation and morphology**

Although Me-4PACz improves the electronic properties (for example, compared to 2PACz and poly(triaryl amine)), the suboptimal formation of perovskite films on planar bottom cells/Me-4PACz often leads to macroscopic holes (Fig. 2a). However, when the perovskite is spin-coated onto nanostructured silicon bottom cells covered with Me-4PACz (Fig. 2b), the occurrence of macroscopic holes is strongly reduced. After the perovskite was deposited, we visually inspected the samples and rejected any that exhibited macroscopic holes. As illustrated in Fig. 2c, out of 45 processed nanotextured tandem solar cells, only two had visible holes after perovskite spin-coating (~95% yield). In contrast, out of 30 planar devices, which were processed in parallel to the nanotextured cells, 15 showed macroscopic holes (~50% yield). To obtain a more systematic understanding of this observation, we measured the static contact angle and the roll-off angle of the perovskite solution on the respective surfaces (Fig. 2d,e). We find decreased surface energy and reduced wettability on the nanostructured surfaces compared to the planar surface (see static contact angle measurement as insets). However, reduced wettability does not necessarily lead to a reduction in droplet...
retention: on the nanotextured sample, we observe a larger roll-off angle (25°), which is defined as the tilting angle at which a droplet of perovskite solution starts to roll off the surface\(^28\), than for the planar reference (18°). This indicates the improved ability of the nanotextured surface to retain the perovskite solution. This observation can be explained from the approach of de Gennes and co-workers, in which the resulting droplet retention force is enhanced by surface roughness pinning the three-phase contact line\(^29,30\). Such phenomena are a feature of pseudo-superhydrophobic surfaces, which exhibit very large contact angles, but on which the droplet is pinned by a particular combination of material and texture, resulting in an unusually large roll-off angle\(^31\). We hence regard increased droplet retention on nanotextured surfaces as an important factor for our improved fabrication yield.

To further study the morphological properties of perovskite layers grown on sinusoidal nanotextures, we captured SEM top-view images (Supplementary Fig. 2), and analysed the projected domain size distribution as depicted in Fig. 2f. The domain size distribution is equal with a mean equivalent disk radius of 130 ± 60 nm and 120 ± 60 nm for planar and nanotextured silicon bottom cells, respectively. This is not surprising as it was reported that with the antisolvent method the crystallization is initiated at the top surface, which is not affected by the buried nanotextured interface\(^32\).

To gain further understanding of the influence of nanotexture on the morphological properties of the perovskite layers, we performed depth-resolved grazing-incidence wide-angle X-ray scattering (GIWAXS) measurements: representative GIWAXS maps of planar and nanotextured samples are shown in Supplementary Fig. 3. Figure 2g displays the azimuthal intensity profiles of the (100) reflection of perovskite averaged over 39 incidence angles from 0.1° to 2° as determined from GIWAXS measurements.

Optical analysis
The optical performance of the planar and nanotextured PSTSCs was analysed by measuring the external quantum efficiency (EQE) and the reflectance (R). Representative EQEs and 1 – R spectra for planar and
nanotextured PSTSCs are shown in Fig. 3a; no notable difference can be observed in the EQEs of the perovskite. In contrast, the EQEs of the silicon subcells differ from each other: the nanotexture diminishes the peaks and valleys caused by thin-film interference (green solid line), which occurs within the perovskite top cell. In addition, the integrated current densities ($J_{\text{ph}}$) of the silicon EQEs are similar. In contrast to the EQE measurements, nanotexturing reduces reflectance considerably from 3.30 to 2.82 mA cm$^{-2}$ current-density-equivalent. Results from a larger number of processed PSTSC devices confirm an average nanotexture-induced reduction of reflectance of around 0.5 mA cm$^{-2}$ current-density-equivalent. From larger number of processed PSTSC devices, a representative experimental EQE and $1 - R$ spectra for a typical pair of planar (black lines; dotted lines of $1-R$; dashed, perovskite EQE; solid, silicon EQE) and nanotextured (green lines; dotted, 1-$R$; dashed, perovskite EQE; solid, silicon EQE) PSTSCs with standard rear reflector (no RDBL) can be seen. Numbers are $J_{\text{ph}}$ in mA cm$^{-2}$.

Fig. 3 | Optical analysis of nanotextures in PSTSCs. a, Representative experimental EQE and $1 - R$ spectra for a typical pair of planar (black lines; dotted 1$ - R$; dashed, perovskite EQE; solid, silicon EQE) and nanotextured (green lines; dotted, 1-$R$; dashed, perovskite EQE; solid, silicon EQE) PSTSCs with standard rear reflector (no RDBL). Numbers are $J_{\text{ph}}$ in mA cm$^{-2}$. b, c, Box-plot diagrams of integrated current density from experimental reflection measurements ($J_{\text{ph}}$) (b) and the sum of integrated current densities in the perovskite and silicon subcell from EQE measurements ($J_{\text{ph,pero}} + J_{\text{ph,si}}$) (c). IQR, interquartile range. d, Cross section through a meshed unit cell of a PSTC with nanotextures between perovskite and silicon subcells, as used for FEM simulations. a-Si:H(i), intrinsic hydrogenated amorphous silicon. e, f, Sensitivity analysis for different layer thicknesses, based on optical simulations: minimum of photogenerated current densities $J_{\text{ph,pero}}$ and $J_{\text{ph,si}}$ as a function of perovskite (e) and nc-SiO$_2$:H(n) (f) layer thickness for planar and nanotextured PSTSCs. In e, the perovskite thickness for the nanotextured device is defined as the thickness of a planar layer with the same overall volume. For clarity, in f only data for samples with a perovskite thickness within $\pm$5 nm of the optimal perovskite thickness are shown.

In view of process robustness we performed a numerical sensitivity analysis to study the influence of layer thicknesses in the top cell on the photogenerated current density. We used data sets obtained during a Bayesian optimization based on the finite-element method (FEM), which rigorously solves Maxwell’s equations. Figure 3d shows a cross section through a meshed unit cell of a nanotextured PSTSC, as used in the simulations. A previous study showed no influence of the period of sinusoidal textures on the reflectivity of PSTSCs for periods ranging from 500 to 1,000 nm (ref. 12). This is in contrast to pyramidal textures, which reveal a strong dependence of antireflective effect on pyramid size$^{13,14}$. During the Bayesian optimization, the overall current density of the monolithic tandem device was optimized by maximizing the minimum of the two subcell photocurrent densities. We assume perfect Lambertian light trapping at the rear side of the silicon bottom cell without any parasitic absorption such that the following results mainly concern the perovskite top-cell layer design. The results of the sensitivity analysis are shown in Fig. 3e,f as functions of the perovskite and nc-SiO$_2$:H(n) layer thickness, respectively. According to these simulations, the optimized nanotextured PSTSC performs almost the same as the optimized planar device with matched photogenerated current densities of 20.24 and 20.22 mA cm$^{-2}$, respectively. As seen in Fig. 3e, the sensitivity to a changing perovskite thickness is also similar for the planar and nanotextured designs. However, texturing reduces the sensitivity to a changing nc-SiO$_2$:H(n) thickness, as seen in Fig. 3f: whereas for planar devices the photocurrent density peaks at around 100 nm nc-SiO$_2$:H(n) thickness, for the nanotextured design the photocurrent density reaches a plateau for nc-SiO$_2$:H(n) layers thicker than 100 nm. This means that nanotexturing of the PSTSC widens the process window for the optical nc-SiO$_2$:H(n) interlayer, which is an important aspect for industrialization of the tandem technology especially when processing on larger areas.
Optoelectronic analysis

Figure 4a shows the current-density/voltage characteristics of representative nanotextured and planar PSTSCs. Both devices show PCEs (η) above 29% with a $J_{SC}$ of 19.45 mA cm$^{-2}$. In addition, the fill factor (η) above 29% with a $V_{OC}$ of 1.88 V and a statistical improvement of the median $V_{OC}$ by around 15 mV for the nanotextured silicon compared with planar PSTSCs (Supplementary Fig. 12). We note that the pseudo-FF (pFF) of 84.4% and 84.1% for nanotextured and planar PSTSCs, respectively, clearly surpass the experimental values and thus point to a further optimization route for these solar cells (see also the results of intensity-dependent $J$–$V$ measurements in Supplementary Fig. 12). Further, the EL emission of perovskite in the nanotextured devices is slightly skewed (Fig. 4c, inset), which might suggest enhanced photon recycling. We note that the implied performance of both silicon subcells is identical and that the efficiency potential obtained with the nanostructured device is almost 32%. The exact mechanism that improves the EQEEL/$V_{OC}$ of the perovskite on the nanotextured silicon cell is not yet fully understood. The enhancement might stem from an improved optoelectronic quality of the absorber layer, possibly related to alterations in the crystal orientation (Fig. 2g). It is known that the impact of crystallite orientation on the carrier dynamics of layered perovskite is crucial; however, this relation has been the subject of controversy for three-dimensional perovskite. There are both research reports claiming that the improved crystallite orientation contributes to improved optoelectronic properties and those claiming that there is no clear correlation. In addition, slightly lower non-radiative recombination losses at the interfaces, or an alteration of the optical coupling between the perovskite and the silicon have also been discussed. In a study by Hou et al., a $V_{OC}$ enhancement of the perovskite on textured silicon was related to an alteration of the electric fields in the perovskite with extended drift regions. We observe no remarkable difference in the dark currents of nanotextured and planar solar cells, and almost no difference in the morphology (Fig. 2), hence the higher emission from EL (Fig. 4) indicates less non-radiative recombination in nanotextured devices.
High-performance tandem solar cells with optically advanced rear reflector

To reduce parasitic absorption losses in the rear reflector, we additionally implemented an RDBL27-48. State-of-the-art monofacial silicon heterojunction solar cells use a TCO/silver grid system as reflector. By designing a thin TCO layer and at the same time increasing the distance between metal and silicon by means of an RDBL, parasitic absorption losses can be reduced. Bush et al. integrated such a reflector into a PSTSC, thereby increasing the $J_{SC}$ and efficiency considerably48. Figure 5a,b illustrates PSTSC layer stacks with a standard reflector and with an RDBL. Three optically different regions can be distinguished: (1) standard reflector (120 nm TCO, no buffer layer), (2) RDBL (20 nm TCO, 180 nm SiO$_2$ buffer layer) and (3) grid fingers of the RDBL (20 nm TCO, no buffer layer). SEM images of the corresponding regions are presented in Supplementary Fig. 13. The SiO$_2$ layer thickness and the grid finger pitch were chosen according to optical and electrical simulations of the RDBL. To quantify the optical properties of these three regions, we performed simulations of the variation of the photogenerated current density in the silicon ($J_{ph,Si}$) with TCO and SiO$_2$ thickness (Fig. 5c). For the optimized layer thicknesses, the photocurrent density of the silicon bottom cell $J_{ph,Si}$ increases from 19.4 to 19.7 mA cm$^{-2}$ when replacing the standard reflector design (region 1) with the RDBL (region 2), mostly due to reduced parasitic absorption in the TCO–silver contact. A full-area design as in the grid finger (region 3) would lead to $J_{ph,Si}$ of 19.0 mA cm$^{-2}$.

Finally, the local contact area has to be optoelectronically optimized not only regarding the optical properties, but also in terms of lateral transport and contact resistive losses. This means that the TCO thickness and the grid finger geometry have to be balanced. Figure 5d shows the simulation results (see Methods for details). We find decreased power loss with TCO thickness reduction, since the benefit from reduced parasitic absorption dominates. For a silver-grid finger width of 40 µm we find an optoelectronic optimum for a grid pitch of roughly 1.1 mm (Fig. 5d). Hence, a finger pitch of 1 mm was implemented, resulting in local contacts covering roughly 4% of the active area. The optoelectronic analysis further shows that overall the RDBL potentially increases the power output by 0.3 mW cm$^{-2}$. The EQE spectra of experimentally realized PSTSCs with and without an RDBL are shown in Fig. 5e. In addition to the nanotexture-induced flattening of the silicon EQE, the RDBL features an increased absorption at the silicon band edge, as expected from the optical simulations. To account for the additional current density from the RDBL in the silicon subcell, we shifted the perovskite bandgap from 1.68 to 1.66 eV. As described by Peña-Camargo et al., the optoelectronic properties do not notably change when slightly adjusting the perovskite composition49; only the $V_{OC}$ changes according to the bandgap. One of the best PSTSCs, featuring both a nanotextured interface between the perovskite and silicon subcell and an RDBL was sent to Fraunhofer ISE Callab for independent certification (Fig. 5f; see Supplementary Fig. 14 for certificate). The cell reached a PCE of 29.75% when measured from $J_{SC}$ to $V_{OC}$ (designated area, 1.0163 cm$^2$), with a $V_{OC}$ of 1.92 V, an FF of 79.4% and a $J_{SC}$ of 19.56 mA cm$^{-2}$. The certified PCE, as determined from MPP tracking, is 29.80%.

Conclusions

In this study, we integrated gentle submicrometre-periodic nanostructures and an improved back-reflector design into monolithic PSTSCs. The nanotextures enable greatly enhanced process yield of the solution-processed perovskite top cell from around 50% in case of planar to about 95% for nanotextured silicon bottom cells. We further observed a reduction of reflection losses amounting to around 0.5 mA cm$^{-2}$-current-density-equivalent. As a result, combined short-circuit current densities in the perovskite and silicon subcells of up to 40.0 mA cm$^{-2}$ are observed, which is one of the highest values...
reported in the literature for two-terminal PSTSCs. A sensitivity analysis further indicates that the nanotextures substantially improve the performance robustness upon deviations from the optimal nanocrystalline silicon oxide layer thickness—an important aspect with regards to the industrialization of tandem technology and processing on larger areas. The main driver for improved PSTSC performance was an increase of the open-circuit voltage by around 15 mV that stems, according to our subcell-selective EL characterizations, solely from an improved perovskite-top-cell performance. In addition to the nanotextures at the silicon bottom-cell front side, we further implemented an RDBL at the rear side. This design further improves the current density in the silicon bottom cell by around 0.3 mA cm⁻² by reducing parasitic absorption losses. By combining nanotextures and RDBL in one PSTSC, we achieved a certified PCE of 29.80%. These results pave the way for the wide use of nano-optical designs in high-efficiency perovskite solar cells and other metal halide perovskite optoelectronic devices in the near future.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41565-022-01228-8.

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Methods

Device fabrication

**Nanotextured silicon bottom cells.** A three-step process, developed by Sutter et al.\(^4\), was used to create nanotextures in silicon wafers for PSTSCs. Sinusoidal master structures, manufactured by laser interference lithography\(^5\), were replicated by nanoimprint lithography with a polydimethylsiloxane stamp in an ultraviolet-curable resist (mNe II 210-300; Microresist Technologies) on a double side polished (100) silicon n-type float-zone wafer (Topsil) with a nominal resistivity of 1–5 Ω·cm and an approximate thickness of 280 μm. For the nanoimprint lithography process, the 4 inch wafers were cleaned according to the standard RCA procedure\(^5\). Ambient conditions, 700 μl of resist was spin-coated for 30 s at 3,200 r.p.m. on the wafer, prebaked for 3 min at 60 °C and cured by ultraviolet light with the polydimethylsiloxane stamp comprising the inverted nanotexture from the master on top for 2 min. In a second step, the reproduced nanotexture was anisotropically etched into the silicon substrate by reactive-ion etching with the etching gases sulfur hexafluoride and fluoroorufluorine (CHF₃) for 16 min at 90 W radiofrequency power and 20 mtorr pressure. In this step, the nanotextured resist served as a 3D etching mask and was able to replicate the texture into the underlying silicon. An additional reactive-ion etching oxygen plasma was applied to eliminate organic residues. Due to the diffusion of ions and silicon lattice distortion, a subsequent wet-chemical defect etch with nitric acid, phosphoric acid and fluoric acid (HNO₃ (65%) / H₃PO₄ (85%) / HF (50%)/ H₂O, 30:10:1:15) was performed. Prior to the texturing of the rear side with random pyramids with (111) facets (potassium hydroxide etched, 8 min at 83 °C, CellTex Ultra (ICB) as additive), the nanotextured front side was protected with a 320-nm-thick SiO₂ layer deposited by plasma-enhanced chemical vapour deposition (PECVD). After removal of the capping in HF, a final RCA clean and HF dip (1% dilution in water) were performed. Intrinsically and doped hydrogenated amorphous (a:Si:H(0)-a:Si:H(p)) and nc-SiO₃:H(n) layers were deposited by PECVD using an Applied Materials AKT1600 cluster tool. To account for the enhanced surface area, the process parameters for the front side of the nanotextured wafers were adjusted by a factor of 1.3 versus the planar case. A 20 nm layer of doped indium oxide as TCO was sputtered as the recombination mask with an area of 1.1 cm². After sputtering silver, the silicon wafers contacts of the silicon bottom cell were processed through a quadratic mask. The active area is defined by the metal frame and is slightly larger than 1 cm².

Characterization

Roll-off measurements were conducted using a contact angle goniometer (DSA 25, Krüss). In all measurements, 10 μl droplets of perovskite solution (see above) were used. For the roll-off angle measurements the samples were placed on fixed, inclined stages before dispensing the solution. The methodology used here enables qualitative analysis of the droplet retention behaviour of both planar and textured surfaces\(^6,7\).

Reflectance was measured under an angle of 8° in 5 nm steps from 300 to 1,200 nm with a PerkinElmer Lambda 1050+ ultraviolet/visible/near-infrared spectrophotometer, which was calibrated with a Spectralon. For statistics on the reflectance of PSTSCs, perovskite absorbers with bandgaps varying from 1.66 to 1.68 eV were used.

Atomic force microscopy (AFM) measurements were performed with a XE-70 (Park Systems). For accurate profile scans in the nanometre regime, high-aspect-ratio tips were used. SEM was performed with a Merlin field emission scanning electron microscope with a Gemini II optical column (Zeiss).

Ex situ GIWAXS measurements were conducted at the mySpot beamline at the electron storage ring BESSY II. The samples were measured at room temperature in reflection mode with incidence angles from 0.1° to 2° (39 scans, 0.05° interval), using a radiation energy of 9 keV (λ = 1.378 Å). The size of the beam was around 100 μm (ref. 8). The 2D images were plotted and integrated with Dpdak and GIXSGUI software\(^9,10\).

EL measurements. Absolute EL measurements were performed using a calibrated silicon photodetector and a Keithley 485 picoammeter. The detector (active area, ~1 cm²) was placed directly in front of the device, and the total photon flux was determined from the emission spectra of the perovskite and silicon subcell, and the EQE of the detector. To selectively pick up EL from the perovskite or crystalline silicon subcell, appropriate long-pass and short-pass filters were used. Under-estimation of the EQE was due to undetected photons that escaped to the side was compensated by additional measurements at different distances and with a larger detector (active area, ~2 cm²). During a typical EL measurement, a Keithley 2400 source meter was used to apply a forward bias to the cell, and the injected current was monitored. Measurements were conducted with a home-written LabVIEW routine. Relative EL spectra of the PSTSCs were measured with an Andor SR393i-B spectrometer equipped with an Andor iDus silicon charge-coupled device camera and an iDus InGaAs detector array. The spectral response of the system was calibrated by using a calibrated halogen lamp with specified spectral irradiance. The quasi-Fermi level splitting (QFEL) was calculated according to:

\[
\text{QFEL} = \frac{k_B T \times \ln \text{EQE}_{\text{EL}}}{\Delta \mu_{\text{EL}}},
\]

where \(k_B T\) is the thermal voltage.
is the thermal energy at room temperature, and \( J_{e,a} \) and \( J_{c} \) are the radiative recombination current in the dark and the 1-sun-equivalent (AM1.5G) generation current, respectively. The quantification of the \( J_{e,a} \) of both subcells is shown in Supplementary Fig. 15 and Supplementary Table 1.

Solar cell characteristics. The tandem solar cells were measured in air under AM1.5G (1 sun) equivalent illumination with a Wavelabs Sinus-70 light-emitting diode (LED) class AAA solar simulator as described in a previous publication by Al-Ashouri et al.7. The cells were not preconditioned. For calibration we used a slightly modified calibration routine as described in ref. 63. We adjusted the spectrum such that it led to the photogenerated current densities obtained by EQE measurements for both subcells. Thus, for a perovskite-limited cell, we first increased the intensity of the blue light to get a silicon-limited cell. Subsequently, the near-infrared region was adjusted until the \( J_{SC} \) of the silicon-limited tandem solar cell was equal to the \( J_{SC} \) (calculated from EQE and AM1.5G spectrum). Finally, the intensity of the blue light was decreased until the tandem solar cell was perovskite-limited again and the \( J_{SC} \) was equal to the \( J_{SC,prev} \). For a silicon-limited cell, the procedure is the opposite. The backside of the cell was contacted with a metal vacuum chuck at 25 °C, whereas the front side was contacted with two gold probes. A black laser-cut aperture mask covered the substrate outside of the active area. The \( J-V \) measurements and MPP tracks were recorded using a home-built LabVIEW software. The EQE spectra were recorded with a home-built set-up using chopped (79 Hz) monochromatic light from a xenon and helium lamp, each covering a specific part of the spectrum. To measure the EQE of the perovskite subcell, the silicon subcell was saturated using an LED with 850 nm peak emission. To maintain short-circuit conditions, a bias voltage of 0.6 V was applied. The silicon subcell was measured by saturating the perovskite subcell with blue light from an LED (455 nm) and applying a bias voltage of 1.0 V. For statistics on totalized integrated subcell current densities of PSTSCs, perovskite absorbers with bandgaps varying from 1.66 to 1.68 eV were used.

Simulations

3D optical simulations: JCMwave and Bayesian optimization. The data used for the sensitivity analysis presented in Fig. 3 were generated during a layer-thickness optimization with a Bayesian optimization algorithm65, similar to an optimization we presented in previous work66. During the optimization the thicknesses of the perovskite and nc-SiO2:H4(n) layers were varied to maximize the function \( \min(J_{ph,prev}) \), which is a well-suited optimization function for monolithic tandem solar cells, as it directly accounts for current matching67. The individual simulations were performed with the FEM solver JCMsuite68 for a 300–1,190 nm wavelength range and produced absorption spectra for all the layers of the solar cell stack. To be able to do the FEM simulations, we had to restrict the simulation domain to the perovskite top cell and to assume the silicon to be infinitely thick. To compensate, we corrected the absorption of the silicon layer by assuming perfect light trapping for the silicon wafer according to Tiedje and Yablonovitch59. From the absorption spectra, the photocurrent densities were calculated60. For example, for reflection the equation is given as

\[
J_{ph,R} = -\varepsilon \int_{300}^{1190} R(\lambda) \times \Phi_{AM1.5G}(\lambda) \, d\lambda
\]

where \( \varepsilon \) is the elementary charge, \( R \) is the spectral reflectance \( \Phi \) is the spectral photon flux under the AM1.5G condition65. Hence, the current-density equivalents can be regarded as solar-spectrum-weighted reflectance values and their relevance for the solar cell device can immediately be seen. Details on the layer thicknesses, optical material data and optimization results are given in Supplementary Table 2.

1D optical simulations: GenPro4. The optical simulations on the RDBL shown in Fig. 5c were performed with GenPro4 developed at TUDelft69, which uses the net-radiation method and ray tracing for scattering at the pyramidal back side of the solar cell. Details on the layer stack used for these simulations are given in Supplementary Table 3.

Optoelectrical simulations: Quokka. For the RDBL with local contacts, the sheet resistance of the TCO (\( R_{shear} \)), which depends on its electrical properties and thickness, and the resistance of the grid fingers, which depends on their pitch, geometry and resistivity, have to be optimized. To calculate this balance, electrical simulations were carried out with the Quokka3 software70 for which we assumed the wafer properties, finger geometry and resistivity, and Si/TCO/Ag contact resistivities to remain constant. Note also that \( R_{shear} \) does not increase linearly with thickness due to the sheet resistance of the top cell 

Data availability

All the data used to plot the figures are available via https://doi.org/10.5442/ND000009 (ref. 64). Source data are provided with this paper.

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Author contributions

P.T. and J.S. fabricated the perovskite top cell, finished the PSTSCs and characterized the PSTSC by sun simulator, EQE and reflection measurements. J.S. developed and prepared silicon substrates with nanotextures, and performed SEM and AFM measurements. J.S., P.W. and A.C. prepared the silicon bottom cells. A.C. developed the process for the RDBL and performed optical and electrical simulations concerning the RDBL. K.J. performed optical FEM simulations and Bayesian optimizations. D.Y. carried out and analysed contact-angle and roll-off experiments. F.L., M.G. and M.S. performed EL/PL measurements and analysed the data. B.L. carried out stability measurements. O.S. and J.L. performed and analysed the GIWAXS measurements. A.A.-A., B.L. and E.K. developed and supported the PSTSC fabrication process. E.U. supervised the GIWAXS activities. D.N. supervised the EL/PL activities. R.S. and B.S. supervised the silicon bottom-cell activities. B.R. supported all activities and gave advice on the structure of the manuscript. S.A. supervised the PSTSC activities. C.B. supervised the optics and nanotexture activities. P.T., J.S. and C.B. coordinated the project. P.T., J.S. and C.B. wrote the manuscript, and all authors commented on the paper.

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