Scalable two-terminal all-perovskite tandem solar modules with a 19.1% efficiency

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Monolithic all-perovskite tandem photovoltaics promise to combine low-cost and high-efficiency solar energy harvesting with the advantages of all-thin-film technologies. To date, laboratory-scale all-perovskite tandem solar cells have only been fabricated using non-scalable fabrication techniques. In response, this work reports on laser-scribed all-perovskite tandem modules processed exclusively with scalable fabrication methods (blade coating and vacuum deposition), demonstrating power conversion efficiencies up to 19.1% (aperture area, 12.25 cm²; geometric fill factor, 94.7%) and stable power output. Compared to the performance of our spin-coated reference tandem solar cells (efficiency, 23.5%; area, 0.1 cm²), our prototypes demonstrate substantial advances in the technological readiness of all-perovskite tandem photovoltaics. By means of electroluminescence imaging and laser-beam-induced current mapping, we demonstrate the homogeneous current collection in both subcells over the entire module area, which explains low losses (<5% rel) in open-circuit voltage and fill factor for our scalable modules.

T rigged by the advances of single-junction organic–inorganic metal halide perovskite solar cells (PSC) with a wide range of bandgaps1–3, perovskite-based tandem photovoltaics (PVs) have come into research focus in recent years4–6. The promise of any tandem PV technology lies in high power conversion efficiencies (PCEs), beyond the Shockley–Queisser limit of single-junction solar cells7–11. Along with perovskite/silicon12–14 and perovskite/Cu(In,Ga)Se215–17 tandem photovoltaics, two-terminal all-perovskite tandem solar cells (2TPT-SCs) have raised great interest in recent years, combining a wide bandgap (WBG) perovskite top subcell (Eg ≈ 1.6–1.8 eV) and a narrow-bandgap (NBG) perovskite bottom subcell (Eg < 1.3 eV)18–23. Several studies reported efficiencies of laboratory-scale 2TPT-SCs exhibiting a PCE of >23% (refs. 7,18,19–23), with the current certified record being 26.4% (ref. 26). Considering that these values exceed the PCEs of commercial single-junction multicrystalline silicon or copper indium gallium selenide (CIGS) cells22,23, it is time to take the next step and develop scalable fabrication processes and interconnection schemes for 2TPT-solar modules (2TPT-SMs).

While laser-scribed interconnection schemes of single-junction perovskite solar modules were successfully implemented in the past at very high geometrical fill factors of >98% (ref. 29), there is yet no report addressing the very specific challenges of the monolithic all-perovskite tandem module. Similar to single-junction perovskite solar modules, scaling 2TPT-SCs will adapt the established thin-film module interconnection concept of elongated PSC stripes that are separated by equidistant interconnections formed by three scribing lines21,22. However, realizing laser-scribed interconnection lines in 2TPT-SMs necessitates processing in an inert atmosphere and a scribing process that is compatible with a tandem solar cell layer stack containing two types of perovskite thin films. Next to the module interconnection, 2TPT-SM device architectures processed with industrially scalable deposition methods need to be developed. So far, all 2TPT-SCs have required several spin-coating steps, which limited the device area to a laboratory scale of up to around 12 cm² (ref. 25). For large areas, scalable methods such as thermal evaporation17, blade coating37,38,39, spray coating40 and slot-die coating41,42 have been investigated for perovskite photovoltaics. Among them, blade coating (PCE: 17.8%)19 and slot-die coating (PCE: 20.8%)42 have demonstrated remarkable potential in processing single-junction perovskite solar modules. The key challenge in processing 2TPT-SMs with scalable solution-based methods is the sequential layer deposition considering the similar solubility of the WBG and NBG perovskite layer in the top and bottom cell, respectively. The latter poses a major hurdle to the sequential deposition of the 2TPT device architecture as it induces facile degradation of the underlying layer (for example, the WBG perovskite) when processing the solution of the NBG bottom solar cell.

In response to the above-mentioned challenges in upscaling 2TPT PV, this study reports on laser-scribed 2TPT-SMs processed exclusively with scalable fabrication techniques. The study introduces the scientific and technological advances that led to these prototypes. First, we introduce our reference device architecture, which benefits from reduced optical losses and shows close performance in small-scale 2TPT-SCs (aperture area of 0.1 cm²) compared to the record devices reported in the literature26. Second, we report on a high-throughput laser-scribing process in an inert atmosphere and demonstrate high geometric fill factors (GFF) in small-scale spin-coated 2TPT-SMs (aperture area of 2.25 cm², referred to as ‘spin-coated 2TPT-SMs’). Using time-of-flight secondary ion mass spectrometry (ToF-SIMS), we reveal the highly selective laser patterning of the scribing lines. Finally, we demonstrate

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The disadvantage of the percolated Au layers is a reduced transmission compared to ITO recombination layers (Supplementary Fig. 10). According to our observations, even the conductivity of thin ITO layers of thickness ~15 nm (~450–500 Ωsq⁻¹ sheet resistance) is sufficient to induce notable shunt resistance losses. For this reason, a percolated Au recombination layer is employed here despite the high parasitic absorption losses. These absorption losses are attributed to plasmonic losses of percolated Au layers in the recombination junction (SnO₂/Au/PEDOT:PSS), even at the lower nominal thicknesses of Au down to 0.3 nm (Supplementary Note 3 and Supplementary Figs. 6–12). However, neither a percolated Au recombination layer nor a thin ITO recombination layer (~15 nm) are optimal and substantial future improvement potential lies in the development of improved recombination or tunnel junctions for all-perovskite tandem PVs.

Figure 1e shows an all-laser-scribed spin-coated 2TPT-SM comprising four serially interconnected cell stripes with a total aperture area of 2.56 cm² and a 94.7% GFF (Fig. 1d,e and Supplementary Fig. 21a,b). It should be noted that the low stability of mixed Sn/Pb-based perovskite narrow-bandgap bottom solar cells poses a general challenge to the industrial upscaling of all-perovskite-based tandem photovoltaics. Exposure of the devices to the ambient atmosphere needs to be avoided at any stage. For this purpose, a custom-built laser-scribing set-up is employed for the fabrication of the monolithic interconnections of 2TPT-SMs; the set-up allows patterning in an inert atmosphere to prevent O₂ exposure and degradation of the NBG bottom subcell during the scribing process.

Compared to other thin-film module technologies (GFF up to 96% (ref. 4)), as well as single-junction perovskite modules (GFF ~95% (refs. 3, 25–26)), the achieved GFF of 94.7% is very respectable. For details on the laser-scribing process, we refer to the experimental section and Supplementary Fig. 21. As shown in Fig. 1e, the Vₘₚ of each cell stripe of the spin-coated tandem module corresponds to around 2.01 V, while the total Vₘₚ of the module equates to the expected 8.0 V (Fig. 1e). This implies a minimal loss in Vₘₚ of only 10 mV per cell stripe (0.5% relative). Moreover, our modules retain a high FF of 75%, demonstrating a negligible detrimental effect of introduced module interconnections (compared with single-junction solar cells showing FF of ~75% (Fig. 1b)). Our in-house measurement of the champion module (without encapsulation and masking, measured in ambient atmosphere) with an aperture area of 2.56 cm² (defined by laser scribing) exhibits a high PCE of 22.2% (23.7% active area PCE) with Vₘₚ = 8.0 V, short-circuit current Iₛₚ = 9.57 mA and FF = 75% (Fig. 1c). The MPP tracking of the modules under continuous illumination (1 Sun) shows a SPCE of 21.4% (and stable power output of 54.7 mW) for 15 hours (Fig. 1f). Moreover, the module shows stable power output at the maximum power point for 3.5 hours and 85°C in an inert atmosphere (Fig. 1g).

The good performance of the spin-coated 2TPT-SM was certified by the accredited CalLab PV Cells of Fraunhofer ISE. Given the sensitivity of the NBG perovskites towards ambient atmosphere and the requirements of the certification standards, the module was encapsulated and covered with an additional external mask. The encapsulated and masked champion module (aperture area of 2.43 cm²) exhibited an in-house PCE of 19.8% (I–V measurement, backward scan) and an externally measured PCE of up to 18.3% (Fraunhofer ISE CalLab PV Cells). The certified PCE determined by steady-state MPP is 17.99% ± 0.63% (Fraunhofer ISE CalLab PV Cells, see Supplementary Fig. 22). The apparent difference between the certified and in-house PCE is attributed to degradation losses that originate from the encapsulation process (which is conducted in ambient atmosphere) and the extended time of transfer (see Supplementary Fig. 22, Supplementary Table 2 and Supplementary Note 6 for a detailed discussion). Therefore, to reach higher stability for the 2TPT-SMs, proper encapsulation under an inert atmosphere...
is required. Further advances in the stability of all-perovskite tandem photovoltaics will be inspired by the research and development of additives and passivation layers, such as the latest work of Lin et al., which demonstrated high stability by grain-boundary passivation\textsuperscript{26}.

Using ToF-SIMS, we provide a three-dimensional (3D) depth profile of the material composition at the 2TPT-SMs interconnection, highlighting the selective laser-ablated material removal at the P1–P3 scribing investigated here by ToF-SIMS exhibits a very similar width (dead area) compared to the previous analysis via scanning electron microscopy (SEM), of 220\,µm (Fig. 2b). Considering the dead area width of 210\,µm, which can be estimated by ToF-SIMS, we determine a GFF of 94.7\% (± 0.1\%).

Having investigated the selective laser-ablated material removal by means of 3D profiles and SEM surface scans, we assess the homogeneity of the tandem module with regard to: (1) photocurrent generation in the subcells over the entire module area using LBIC mapping; and (2) individual defects and shunts in the top and bottom subcells of the module by electroluminescence imaging.

To study the homogeneity of photocurrent generation in each subcell of our tandem module, we employed an adaptable configuration of tandem solar cells and modules.

### Design and performance of all-perovskite tandem solar cells and modules.

**a.** Schematic and layer stack of all-perovskite tandem solar cells applied in this work. The champion tandem solar cells and modules employ sputtered indium tin oxide layers (thickness ~15\,nm) and percolated Au thin films (nominal thickness ~1–2\,nm) as a recombination layer, respectively. NBG and WBG are the abbreviations of the narrow and wide bandgap. **b.** Current density–voltage (J–V) curve and power conversion efficiency tracked at the maximum power point of the champion tandem devices for 5 min (in the inset). **c.** EQE of top and bottom subcell as well as the sum of both (grey symbolled line), and total absorbance calculated by 1 − reflectance (black solid line) for monolithic all-perovskite tandem solar cells. The light and dark blue regions denote the parasitic absorption and reflection losses, respectively. The corresponding losses in current density are provided. **d.** Schematic illustration of the two-terminal all-perovskite tandem solar module (not to scale) interconnection denoting the active area and scribing lines (for more details see Supplementary Fig. 21). The colour used for the module layers is the same as for tandem solar cells. **e.** J–V characteristics of individual tandem cell stripes of the module and the respective fill factors after a stepwise inclusion of cell stripes included in the measurement. **f.** Power, voltage and current at the maximum power point of the champion tandem solar module under continuous AM 1.5G illumination. **g.** Normalized power (Norm.), voltage and current at the maximum power point under temperature stress at 85 \,°C in nitrogen atmosphere. Panels **a–c** refer to tandem solar cells while panels **d–g** to tandem modules.
of LBIC mapping (Fig. 2d–f). The layer stack of modules for the LBIC study is identical to the tandem modules as shown in Fig. 1a (employing Au as a recombination layer). Conventional LBIC is an established method to investigate the homogeneity of perovskite layers in single-junction solar cells and modules46–50.

To discriminate between the photogenerated current in the top and bottom subcells, the LBIC set-up is expanded by a second excitation laser, and now comprises laser sources of wavelength 530 nm and 850 nm, respectively. First, exciting the module with the 530 nm laser, we track the LBIC signal in the top subcell, as photons of this wavelength are fully absorbed in the WBG perovskite absorber layer (Fig. 2d,e). The corresponding LBIC map shows uniform photocurrent generation in the entire active area of the module. Second, using the 850 nm laser, we selectively excited the bottom subcell, as photons of this energy are not absorbed by the WBG perovskite top subcell, but only by the NBG perovskite bottom subcell (Fig. 2d,f). Similar to the LBIC map of the top subcell, the LBIC signal of the bottom subcell also exhibits a homogeneous signal over the entire module area. Interestingly, some defects are apparent as dark spots in the LBIC signal of the top subcell but appear bright in the LBIC signal of the bottom subcell. These defects are attributed to pinholes in the top subcell and inhomogeneity in the perovskite thin-film thickness. Overall, the LBIC data prove an even photocurrent generation and extraction over the entire active area for both the top and bottom subcells of our module.

To study the distribution of local defects and confined local charge barriers in our tandem modules, we further employ electroluminescence imaging, which exhibits only indirect information on current collection but at much higher resolution (for our set-up a pixel coarsely represents about 13 µm × 13 µm) compared to LBIC (our LBIC resolution is limited by laser spot diameter;~2 μm and 500–2000 nm in our measurements). To distinguish the electroluminescence signal of the two subcells in our tandem module (1.78 eV WBG perovskite top subcell (Supplementary Fig. 24) and 1.26 eV NBG perovskite bottom subcell52), the electroluminescence signal is imaged with: (1) a neutral density filter to track the full electroluminescence signal; (2) a longpass filter (~760 nm) for imaging the bottom subcell; and (3) a shortpass filter (~775 nm) for imaging the top subcell (Fig. 2g, Supplementary Figs. 25–27 and Supplementary Video 1). Given the two-terminal series connection of the top and bottom subcells, the electroluminescence signal rises only for a voltage add-up across each cell stripe close to the sum of the Voc of both subcells (that is, close to 8 V for our module of four cell stripes; see Supplementary Fig. 28). As shown in Fig. 2h,i, the recorded electroluminescence images reveal overall good homogeneity, confirming the LBIC data. However, several local defects are present in the active area (indicated by black arrows for the top and bottom subcells).
Fig. 3 | Upscaling the tandem modules and their characteristics. a, Schematic of the fabrication sequence for scalable processing of the tandem modules using a combination of blade-coating and vacuum-deposition techniques. b, The front side of the fabricated tandem module with seven cell strips and an aperture area of 12.25 cm². c, Current density–voltage (J–V) characteristic of stepwise accumulated tandem cell stripes of the module and the respective fill factors. d, Power at the maximum power point of the champion tandem solar mini-module with 12.25 cm² aperture area under continuous AM 1.5G illumination and nitrogen atmosphere. e, Photoluminescence of the tandem perovskite layers with excitation lasers of 513 nm. The insets are the photoluminescence imaging of the top and bottom subcells by use of 775 nm shortpass and 760 nm longpass filters, respectively. f, Photograph and corresponding J–V characteristics for the electroluminescence image. g, Electroluminescence of the tandem perovskite layers. The insets are the electroluminescence imaging of the top and bottom subcells by use of 775 nm shortpass and 760 nm longpass filters, respectively.
We attribute these defects to either local variations in charge-carrier injection (due to different series resistance) or local variation in out-coupling of luminescence from the device (for example, due to variations in surface roughness and thickness).

At the same time, most features are visible in both electroluminescence images, indicating that the fabrication defects in the top subcell affect the quality of the bottom subcell too. However, some features are only present in images that display the particular defects or inhomogeneity in each subcell (Fig. 2h,i). Although electroluminescence and LBIC analyses agree in terms of homogeneity in the active area as well as laser-scribed interconnection lines, electroluminescence imaging allows us to reveal individual fabrication defects of the modules in both deposition and laser-scribing steps, facilitating process optimization towards high-efficiency 2TPT-SMs (Fig. 2e,f,h,i). Overall, we demonstrate in this section the ability to scale up the spin-coated 2TPT-SCs to modules with only a 6% relative drop in PCE, which is in very good agreement with the GFF loss associated with the module interconnection.

**Upscaling all-perovskite tandem solar modules**

Having developed an interconnection scheme with high GFF, next we present a scalable 2TPT-SM architecture that enabled a prototype module (aperture area of 12.25 cm²) fabricated with scalable thin-film deposition methods (Fig. 3a,b). The scalable modules employ the same architecture as the spin-coated modules (see also Supplementary Note 7). The fabrication of the modules is conducted
with a combination of blade-coating (2PACz, WBG perovskite, PEDOT:PSS, NBG perovskite and PCBM) and vacuum-deposition (LiF, Ca, SnO, Au, BCP and MgF) methods (Fig. 3a).

While several of these deposition steps are adapted from the literature (Methods), the processing of the NBG perovskite bottom solar cell required a major innovation to address the challenge of processing the NBG perovskite layer on top of the WBG perovskite top cell without dissolving the underlying layer (see the next section). Prototype modules reached a PCE of 19.1% (aperture area of 12.25 cm²; seven cell strips and 94.7% GFF; Fig. 3c) and 18.3% MAP tracking (corresponds to 224 mW) for 20 hours with less than 7% drop (Fig. 3d). As shown in Fig. 3c and Table 1, in the current blade-coated module, the single 2T tandem cell strips with 1.75 cm² active area on the module show almost the same FF, Voc and Jsc (with an average PCE of 19.4%), which results in a 19.1% PCE for modules. Compared to our spin-coated modules with 2.56 cm² aperture area of solvents away from the sample surface53. As a result, the resting time of the NBG drops to <5 s, which prevents degradation of the underlying WBG top solar cell (Fig. 4a).

Important, this nitrogen gas flow-assisted crystallization of the NBG perovskite on the top subcell produces a microscopic, defect-free morphology (PV2; Fig. 4c,f), while in the absence of nitrogen gas flow (PV0; Fig. 4c,d) or insufficient nitrogen gas flow (PV1; Fig. 4c,e) a large number of defects appear that lead to degradation of the WBG perovskite top solar cell by the solvents of the NBG perovskite solution.

The importance of nitrogen gas flow in the VAGC of NBG perovskite thin films is further highlighted by comparing the performance of isolated small-area 2TPT-SCs (aperture area: 0.1 cm²) processed from a large-area substrate (>30 cm²) in the same architecture as the scalable 2TPT-SMs (Supplementary Fig. 32). As shown in Supplementary Fig. 32, 2TPT-SM with NBG perovskite thin films processed only by VAGC show considerably lower overall performance compared to VAGC with gas flow. Furthermore, the different PCE of the devices at different locations on the large-area substrate is attributed to the defects and variation in the thickness of the blade-coated layers at different locations. Therefore, we show that scaling up the 2TPT-SM architecture by scalable deposition methods is feasible as long as very fast extraction of solvents of the bottom subcell is guaranteed. In this work, we employ a combination of VAGC with nitrogen gas flow to realize fast drying of the solution-processed NBG perovskite thin film in a mild vacuum range of 10⁻⁴–10⁻⁵ Pa. Similar to gas quenching, the technique realizes a fast mass transport of evaporated solvents away from the surface of the wet film64. In view of future in-line large-area production, gas quenching, as well as combinations of vacuum quenching with gas flow, would need to be realized shortly after the slot- or blade-coating step to ensure sufficiently fast drying.

Overall, this work highlights the facile upscaling route of the 2TPT-SCs from 0.1 cm² up to 12.25 cm² in 2TPT-SMs. Having employed only scalable deposition techniques (blade coating and vacuum deposition) for all 12 layers of the fabricated 2TPT-SMs, the feasibility of upscaling this technology is proven. The demonstrated GFF of our tandem module is comparable to other thin-film PV technologies, such as tandem thin-film silicon photovoltaics (~98% GFF)29 and CIGS photovoltaics (~93% GFF)56. It is noteworthy that the laser set-up applied in this work uses a rather conventional and inexpensive nanosecond laser that is less complex than the widely used picosecond or femtosecond lasers for thin-film solar module patterning in laboratory-scale modules. The technological bottleneck of the current development remains the limited stability of the 2TPT-SMs14,24. Fortunately, given recent advances in engineering the composition14,20,24 as well as defect passivation of NBG solar cells21,25, there is encouraging progress in improving the stability of the NBG bottom subcell.

Conclusions
This work advances two-terminal all-perovskite tandem photovoltaics by introducing a scalable high-efficiency, two-terminal, all-perovskite tandem module. The upscaling of laboratory-scale, widely used picosecond or femtosecond lasers for thin-film solar module patterning in laboratory-scale modules.

### Table 1 | The photovoltaic characteristics of the module and a single reference cell strip of the champion module in forward (FW) and backward (BW) scan directions

| Device         | Area (cm²) | Voc (V) | FF (%) | Jsc (mA) | PCE (%) |
|----------------|-----------|---------|--------|----------|---------|
| Cell strip (BW)| 1.75      | 1.93    | 71     | 24.9     | 19.4    |
| Cell strip (FW)| 1.91      | 70      | 24.5   | 18.8     |
| Module (BW)    | 12.25     | 13.3    | 71     | 24.8     | 19.1    |
| Cell strip (FW)| 13.1      | 71      | 24.8   | 18.8     |
spinning, two-terminal all-perovskite tandem solar cells (0.1 cm², PCE of 23.5%) to laboratory-scale modules (12.25 cm² aperture area) is realized by a combination of vacuum-based deposition processes and blade coating, as well as a scalable interconnection scheme. For successful upsampling and degradation-free growth of NBG on the WBG at the top subcell, a vacuum-assisted growth control is employed. The module interconnection is realized with an all-laser-scribed patterning process in an inert atmosphere, enabling a GFA of up to 94.7%. The best spin-coated and scalable modules presented here demonstrates a PCE of 22.2% (aperture area of 2.56 cm²) and 19.1% (aperture area of 12.25 cm²), respectively. The prototype module presents 95% of initial SPCE for 15 h and 89% for 125 h. The prototype module presents 95% of initial SPCE for 15 h and 89% for 125 h.

### Methods

#### Preparation of the precursor solution for the NBG Cs₆F(Ag⁺,MA⁺)ₓ⁻x−SnO₂-Pb₅f, absorber.

The NBG precursor solution was prepared following the recipe described in our previous work. A 2 M concentration NBG solution was used. In brief, to prepare 1 ml NBG perovskite solution, we dissolved 275 mg ammonium iodide (FAI, GreatCell), 63.6 mg methylammonium iodide (MAI, GreatCell), 372 mg tin iodide (SnI₂, Sigma-Aldrich, 99.999%), 469 mg lead iodide (PbI₂, Alfa Aesar, 99.99%), 10.43 mg lead thiocyanate (PbS₂CN, Sigma-Aldrich, 99.95%) and 3.73 mg tin fluoride (SnF₂, Sigma-Aldrich, 99%) in a 9:1 (v:v) mixture of dimethylformamide (DMF, Sigma-Aldrich, anhydrous, 99.8%) and dimethyl sulfoxide (DMSO, Sigma-Aldrich, anhydrous, 99.9%). We added 32 µl of a previously prepared cesium iodide (CsI, Alfa Aesar) stock solution (1.5 M CsI in DMSO) into 1 ml of the NBG precursor solution. The solution was kept overnight in a nitrogen glovebox (<0.2 ppm and H₂O <0.4 ppm) and, just before deposition, it was filtered using a 0.2 µm PFTE filter.

#### Preparation of the precursor solution for the WBG FA₀.₈Cs₀.₂(I₀.₆Br₀.₄)₃ thin film.

To deposit the WBG perovskite, we used our previously reported V AGC method. In brief, to prepare 1 ml WBG perovskite solution, we dissolved 275 mg SnI₂ (99.99%-Sn, Strem Chemicals) and 370 mg PbI₂ (99.99% PbI₂, Alfa Aesar) in 1,2-dichlorobenzene (Sigma-Aldrich, anhydrous, 99.9%) at 100 °C. The precursor solution was allowed to cool down to room temperature and then deposited on the PEDOT:PSS substrate at 6,000 r.p.m. for 12 s dynamically to avoid uniform coverage. Furthermore, the modules use a percolated Au 15 nm thick LiF thin film deposited by thermal evaporation as the rear electrode. The active area of the tandem perovskite/tandem cell was 0.1 cm².

#### Fabrication of the control two-terminal all-perovskite tandem solar cell with modified architecture (Arch. 1).

Similar to the control sample with Arch. 2, the samples with modified architectures of two-terminal all-perovskite tandem solar cells (Arch. 1) were fabricated in the p-i-n architecture. Glass substrates coated with a 220 nm thick TiO₂ front electrode were used. Cleaning the substrates was conducted consequently in deionized water, acetone and isopropanol for 10 min each in an ultrasonic bath. Immediately before deposition of the HTL, substrates were treated using oxygen plasma for 3 min. As HTL, poly(bis(4-phenyl)(2,4,6-trimethylphenyl)amine (PTAA, 2 mg mL⁻¹ in toluene) was used. For deposition of the PTAA, 60 µl of the PTAA solution in toluene was deposited onto the subcell (16 mm × 16 mm) by spin coating at 6,000 r.p.m. for 30 s, followed by annealing at 100 °C for 10 min. Subsequently, the samples were transferred to a glovebox for deposition of the perovskite thin films. A 15 nm thick Cs₁ₓSnO₂ thin film was deposited by thermal evaporation (OPTIvap, CreaPhys) as an ETL. Subsequently, a 35 nm thick SnO₂ thin film was deposited by ALD. As a recrystallization layer, different thicknesses of Au, of 0.3, 0.6, 1.2 and 1.8 nm, were deposited on the SnO₂ layer by thermal evaporation with a low rate of 0.1 Å s⁻¹ to reach the maximum possible uniformity of Au on the substrates. After filtering the aqueous PEDOT:PSS dispersion (Ossila, Al 4083) with a 0.45 µm polyvinylidene fluoride (PVDF) filter, the solution was deposited on the substrates directly by spin coating at 6,000 r.p.m. for 30 s to deposit Au. Next, the substrates were annealed on a hotplate at 120 °C for 20 min. Afterward, fullerene (C₆₀, 25 nm) and BCP (~7 nm) were thermally evaporated as the electron transport material, followed by the deposition of a Cu (~150 nm) or Ag (~60 nm) thin film as the rear electrode. The active area of the tandem perovskite/tandem cell was 0.1 cm².

#### Fabrication of the spin-coated two-terminal all-perovskite tandem solar cell modules with 2.56 cm² aperture area.

For fabrication of the primitive modules with the spin-coating method and an aperture area of 2.56 cm², 30 mm by 30 mm ITO:H substrates were used (to have extra space for glass–glass encapsulation). We followed all procedures described for the fabrication of small-area (0.105 cm²) 2T tandem modules with our modified architecture, except for using a higher amount of solution to reach uniform coverage. Furthermore, the modules are used as a percolation junction (nominal thickness 1.2 nm). While such percolated Au layers introduce some optical losses, they exhibit a very low lateral conductivity. The architecture of the spin-coated 2TFT-SMs was the same as shown in Fig. 1a, with a SnO₂/percolated Au recombination junction (see also Supplementary Fig. 1, 2 and Supplementary Note 7). The laser-scribing procedures are summarized in the next section was used in the fabrication of our modules. The final modules were encapsulated with butyl rubber at ambient atmosphere for further LBC and electroluminescence characterizations in the atmosphere.

#### Laser scribing of module interconnection lines.

Fabrication of monolithically interconnected two-terminal modules on substrates of 30 mm by 30 mm was facilitated by integrating three scribing lines (P1, P2, P3) in our device stack. On the basis of the modified architecture, P1 was employed to ablate the ITO:H, P2 for ablating the multilayers of 2PACz/WBG perovskite/LiF/C₆₀/Al/AI 4083) with a 0.45 µm polyvinylidene fluoride (PVDF) film, the PEDOT:PSS solution to reach uniform coverage. Furthermore, the modules use a percolated Au as a recombination junction (nominal thickness 1.2 nm). While such percolated Au layers introduce some optical losses, they exhibit a very low lateral conductivity. The architecture of the spin-coated 2TFT-SMs was the same as shown in Fig. 1a, with a SnO₂/percolated Au recombination junction (see also Supplementary Fig. 1, 2 and Supplementary Note 7). The laser-scribing procedures are summarized in the next section was used in the fabrication of our modules. The final modules were encapsulated with butyl rubber at ambient atmosphere for further LBC and electroluminescence characterizations in the atmosphere.
with a 2.56 cm² aperture area was used. A custom-built laser-scribing set-up developed in conjunction with Bergfeld Lasertech was employed, integrating a 1 ns Nd:YVO₄ laser, a Picso AOT-10-MOPA, InnoLas Laser) with available 1064 nm and 532 nm wavelengths, which are typically used in photovoltaics for different purposes. The laser pulse energy was set to 0.6 V s⁻¹ using a source meter (Keithley 2400 A). For NBG single-junction and all tandem solar cells and modules, the solar simulator was calibrated with a certified silicon solar cell (Newport, #2446 and #1758). For J–V measurements of the WBG single-junction perovskite solar cells, a silicon solar cell (Newport) equipped with a KG5 bandpass filter was used for calibration. The stable power output efficiency of the PSCs was determined by tracking the MPP under continuous AM 1.5G illumination. The temperature of the solar cells was actively controlled by a Pellet element connected to a microcontroller and set to 25 °C (85 °C for MPP tracking at thermal stress) while conducting the J–V analyses and MPP tracking. As the J–V characteristics are measured with a single laser scribing set-up, the J_sc recommended for the laser scribing (< 100 μm²) was employed to avoid degradation of the perovskite material. For the certification, external manual masking of the module was required, which decreased the aperture area to 2.43 cm².

Up-scaling the two-terminal all-perovskite tandem solar modules with 12.25 cm² aperture area. For the fabrication of two-terminal all-perovskite tandem solar modules with 12.25 cm² aperture area, fully scalable methods were used in the deposition of the entire layer. For this purpose, a combination of vacuum deposition and blade coating (blade gap of 100 μm) was employed. Substrates of 6.4 x 5 cm² ITO:H were used such that the inhomogeneous region in the blade-coating procedure (7 mm from the beginning and 7 mm from the rate of 10 kHz) could be cut away. The final substrate size for our modules was 5 x 5 cm², in which a 0.5 mm distance from each edge was reserved for gas-scraping and a 1.25 cm² aperture area was used. As shown in Fig. 4, the modules have an aperture area of 12.25 cm² (3.5 cm x 3.5 cm), which contains seven cell strips (with an active area of 1.75 cm² on each). The fabrication of two-terminal tandems was conducted as follows. (1) After P1 deposition, the 2P2A was coated with blade coating with a rate of 10 mm s⁻¹ followed by drying in a nitrogen atmosphere and annealing at 100 °C for 10 minutes. (2) The WBG perovskite layer was then laser scribed from a dilute NBG solution (600 ppm) with a solvent mixt (4:1) of DMF:DMSO. Optimal device performance and perovskite thin-film quality (thickness: 400 – 430 nm, root mean square (r.m.s. roughness: < 20 nm) were obtained for a blade speed of 15 mm s⁻¹. (3) After deposition of the WBG perovskite layer, 1 nm LiF and subsequently 15 nm C60 were deposited by thermal evaporation. (4) Later, a 35 nm thin layer of the WBG causes a low yield of tandem devices due to the degradation of the perovskite thin films and devices were measured with a Bentham PVE300 photovoltaic characterization system by illuminating the solar cell with a high-contrast monochromatic light. For the EQE measurements of the tandem solar cells, we followed the steps recommended in the literature. A filtered bias light with a bandpass region 335–610 nm (FGB37M) was used to measure the bottom subcell, and a longpass filter with cut-off wavelength at 850 nm (FGFL850M) was used for measuring the EQE of the top subcell. The bias voltage was varied as recommended. The EQE measurements of the subcells were used for correcting the J_sc, obtained from J–V measurements. The EQE of the top subcell was measured using an Ocean Optics HDX spectrometer. All measurements were performed in ambient air on encapsulated samples.

Optical characterization and EQE measurements. The absorption spectra of the perovskite thick films and devices were measured and a 2.1 megapixel scientific CMOS camera (Quantalux sCMOS camera, Thorlabs). The electroluminescence was filtered with four different optical filters mounted on a rotating wheel: a 775 nm shortpass (Edmund Optics), a 760 nm longpass (Edmund Optics). The modules were excited using a bright blue LED with a 470 nm emission peak wavelength. A 665 nm longpass filter was used to cut out the blue LED interference. The set-up is shown in Supplementary Fig. 25. The electroluminescence spectra of the cells were acquired using an Ocean Optics HDS spectrometer. All measurements were performed in ambient air on encapsulated samples.

Photoluminescence measurement and imaging. The photoluminescence images were acquired with a 2.1 megapixel scientific CMOS camera (Quantalux sCMOS camera, Thorlabs) and the electroluminescence was filtered with two different optical filters mounted on a stationary wheel: a 775 nm shortpass (Edmund Optics) and a 760 nm longpass (Edmund Optics). The modules were excited using a bright blue LED with a 470 nm emission peak wavelength. A 665 nm longpass filter was used to cut out the blue LED interference. The set-up is shown in Supplementary Fig. 25. The photoluminescence signal was collected with a ×50 lens and filtered with a 665 nm longpass. The excitation wavelength was 515 nm at a repetition rate of 20 kHz with a pulse width of approximately 260 μs. The beam area was 0.787 mm² and the power was 7.9 mW, leading to a fluence of roughly 50 mW cm⁻². The sample (without contact layer and coated with ethyl vinyl acetate) was excited from both sides to obtain the full accumulative photoluminescence spectrum from WBG and NBG perovskites. The EQE measurements of the tandem solar cells were performed using a 2.1 megapixel scientific CMOS camera (Quantalux sCMOS camera, Thorlabs) and the electroluminescence was filtered with four different optical filters mounted on a rotating wheel: a 775 nm shortpass (Edmund Optics) and a 760 nm longpass (Thorlabs). The modules were biased using a Keithley 2450 SMU. The set-up is shown in Supplementary Fig. 25. The electroluminescence spectra of the cells were acquired using an Ocean Optics HDS spectrometer. All measurements were performed in ambient air on encapsulated samples.
cesium ions at 2 keV, scanned over a field of 500 × 500 μm² (target current 120 nA) and was applied to erode the sample, whereas the primary ion source was scanned over a concentric field of 350 × 350 μm² (250 × 250 data points). Spectra were calibrated on the omnipresent C⁺, C₂⁺, C³⁻, O⁺ and Br⁻ peaks. On the basis of these datasets the chemicals assignments for characteristic fragments were determined. For data visualization, secondary ion intensities were rendered to 3D depth profile images.

**Reporting summary.** Further information on research design is available in the Nature Research Reporting Summary linked to this article.

**Data availability**

All data generated or analysed during this study are included in the published article and/or its Supplementary Information. Additional original and unprocessed data used in the article, as well as in the Supplementary Information, are available from the corresponding author upon reasonable request, along with reference/calibration measurements and background information.

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Author contributions

B.A.N. and U.W.P. developed the idea and designed the experiments of this study. D.B.R. conducted the scribing for modules using pulsed laser ablation for the cell area definition and two-terminal serial interconnection. B.A.N. designed and developed the modified two-terminal all-perovskite tandem architecture. B.A.N. designed and fabricated the scalable all-perovskite tandem solar modules. E.A. provided the IO:H substrates. R. Schmager simulated the tandem solar cells. B.A.N., S.M. and H.H. developed the NBG perovskite solar cells. T.F. and H.H. developed and optimized the ALD deposition of the SnO_2. T.F. contributed to the development of MgF_2 thin films. F.S. and F.L. conducted the electroluminescence measurements. B.A.N. and F.S. modified the blade coating of the WBG perovskite. B.A.N. modified the blade coating of the NBG perovskite and charge transport layers. R. Singh encapsulated the modules. R.A. conducted the ToF-SIMS measurement. T.A. coordinated the laser scribing and ALD buffer development and gave technical support during the deposition of vacuum-processed charge transport layers. M.K. conducted the photoluminescence measurements. S.G. helped in developing the HTL layer in the top subcell. D.B.R. and H.H. had contributed equally. B.A.N., U.L., B.S.R. and U.W.P. concluded the discussion of the results. U.W.P. supervised the project. All authors contributed to writing the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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### Experimental design

Please check: are the following details reported in the manuscript?

1. **Dimensions**
   - Area of the tested solar cells
     - Yes: 12.25 cm² and 2.56 cm² for monolithic all-perovskite tandem solar mini-modules and 0.1 cm² for monolithic all-perovskite tandem solar cells.
     - No: Metal masks. Area of masks was determined by optical microscopy measurements. The aperture area of the certified sample was determined by ISE Fraunhofer (see Fig. S22 and attachment).
   - Method used to determine the device area
     - Yes: Metal masks. Area of masks was determined by optical microscopy measurements. The aperture area of the certified sample was determined by ISE Fraunhofer (see Fig. S22 and attachment).
     - No: Metal masks. Area of masks was determined by optical microscopy measurements. The aperture area of the certified sample was determined by ISE Fraunhofer (see Fig. S22 and attachment).

2. **Current-voltage characterization**
   - Current density-voltage (J-V) plots in both forward and backward direction
     - Yes: The data is provided in Fig. 3 as well as the certified device (see Fig. S22.)
     - No: The data is provided in Fig. 3 as well as the certified device (see Fig. S22.)
   - Voltage scan conditions
     - Yes: The voltage scan conditions are described in detail in section "Current-density-voltage (J-V) measurements".
     - No: The voltage scan conditions are described in detail in section "Current-density-voltage (J-V) measurements".
   - Test environment
     - Yes: The test conditions was described in "Current-density-voltage (J-V) measurements" section.
     - No: The test conditions was described in "Current-density-voltage (J-V) measurements" section.
   - Protocol for preconditioning of the device before its characterization
     - Yes: The protocol of measurement was described in "Current-density-voltage (J-V) measurements" section.
     - No: The protocol of measurement was described in "Current-density-voltage (J-V) measurements" section.
   - Stability of the J-V characteristic
     - Yes: The stability of the power output was shown by tracking the maximum power point over time. See "Current-density-voltage (J-V) measurements" section.
     - No: The stability of the power output was shown by tracking the maximum power point over time. See "Current-density-voltage (J-V) measurements" section.

3. **Hysteresis or any other unusual behaviour**
   - Description of the unusual behaviour observed during the characterization
     - Yes: Some devices presented in this work exhibit hysteresis as common for perovskite solar cells. Where apparent and relevant, the effect of the hysteresis is discussed.
     - No: Since this study focuses on the fabrication of monolithic all-perovskite tandem solar mini-modules, some plots only exhibit the I-V data of the backward scan. For champion forward and backward scans as well as MPP tracking is provided (e.g. Fig. 3)
   - Related experimental data
     - Yes: Since this study focuses on the fabrication of monolithic all-perovskite tandem solar mini-modules, some plots only exhibit the I-V data of the backward scan. For champion forward and backward scans as well as MPP tracking is provided (e.g. Fig. 3)
     - No: Since this study focuses on the fabrication of monolithic all-perovskite tandem solar mini-modules, some plots only exhibit the I-V data of the backward scan. For champion forward and backward scans as well as MPP tracking is provided (e.g. Fig. 3)

4. **Efficiency**
   - External quantum efficiency (EQE) or incident photons to current efficiency (IPCE)
     - Yes: See Fig. S19
     - No: The integrated Jsc values are consistent with those from J-V measurements.
   - A comparison between the integrated response under the standard reference spectrum and the response measure under the simulator
     - Yes: Given in method.
     - No: Given in method.
   - For tandem solar cells, the bias illumination and bias voltage used for each subcell
     - Yes: Given in “Current-density-voltage (J-V) measurements” section
     - No: Given in “Current-density-voltage (J-V) measurements” section

5. **Calibration**
   - Light source and reference cell or sensor used for the characterization
     - Yes: Given in “Current-density-voltage (J-V) measurements” section
     - No: Given in “Current-density-voltage (J-V) measurements” section
## Confirmation that the reference cell was calibrated and certified

- **Yes**
- **No**

 Provided in "Current-density-voltage (I-V) measurements" section

## Calculation of spectral mismatch between the reference cell and the devices under test

- **Yes**
- **No**

 Spectral mismatch was conducted on the the certified modules. As we measured the cells with one lamp solar simulator, in the J-V measurements we took the Jsc from the EQE.

## 6. Mask/aperture

### Size of the mask/aperture used during testing

- **Yes**
- **No**

 The active area of the WBG and NBG single-junction perovskite solar cells is defined by a metal mask to 0.1 cm². The aperture area of the laser-scribed modules is defined by the laser scribing (2.56 cm²). For the certification, additional external manual masking of the module was required which decreased the aperture area down to 2.43 cm². For the measurements of the 12.25 cm² modules, a metal mask with a 12.25 cm² aperture area was used. For the latter device, no difference with and without a mask is apparent, highlighting the accuracy of the laser scribing.

### Variation of the measured short-circuit current density with the mask/aperture area

- **Yes**
- **No**

 EQE measurement was used to approve the measured current density by J-V measurement. See Fig 1b and Fig. S19

## 7. Performance certification

### Identity of the independent certification laboratory that confirmed the photovoltaic performance

- **Yes**
- **No**

 Certified J–V characteristics (CalLab at Fraunhofer, ISE) of one of the best working solar modules (with an aperture area of 2.43 cm²) is provided.

### A copy of any certificate(s)

- **Yes**
- **No**

 See Fig. S22

## 8. Statistics

### Number of solar cells tested

- **Yes**
- **No**

 At least around 32 samples for the tandem solar cell and 16 for the solar modules.

### Statistical analysis of the device performance

- **Yes**
- **No**

 See Supplementary Information for statistic of any variation. The tandem solar cell results were collected from at least 32 sam.

## 9. Long-term stability analysis

### Type of analysis, bias conditions and environmental conditions

- **Yes**
- **No**

 Provided in "Current-density-voltage (I-V) measurements" section. See Fig. 1e and Fig. S29. The encapsulation described in the “Fabrication of the modules” section in the main text.