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Efficient bifacial monolithic perovskite/silicon tandem solar cells via bandgap 1

engineering 2

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16 Abstract

15

Bifacial monolithic perovskite/silicon tandem solar cells exploit albedo - the diffuse 17 18 reflected light from the environment - to increase their performance above that of monofacial perovskite/silicon tandems. Here, we report bifacial tandems with certified power conversion 19 efficiencies (PCEs) > 25% under monofacial AM1.5G 1-sun illumination, that reach power-20 generation densities (PGDs) as high as ~26 mW/cm² under outdoor testing. We further 21 investigate the perovskite bandgap required to attain optimized current-matching under a 22 variety of realistic illumination and albedo conditions. We then compare the properties of these 23 bifacial tandems exposed to different albedos and provide energy yield calculations for two 24 25 locations with different environmental conditions. Finally, we present an outdoor test-field comparison of monofacial and bifacial perovskite/silicon tandems to demonstrate the added 26 value of tandem bifaciality for locations with albedos of practical relevance. 27

28 Main

29 Globally, an immense research effort is underway aimed at improving further the power conversion efficiencies (PCEs) of perovskite-based photovoltaics (PV).¹ Recent progress in 30 31 perovskite-based PV is founded on the remarkable optoelectronic properties of perovskites, as 32 well as on important advances made in materials and device engineering, such as the 33 formulation of stable compounds and bulk and surface defect passivation strategies.^{2,3} Thanks 34 to their high absorption coefficient, tunable bandgap and remarkable defect tolerance, 35 perovskites are also attractive for realizing efficient multi-junction, tandem devices.⁴ The combination of perovskites with market-dominant crystalline silicon (c-Si) solar cells as a 36 bottom cell technology is particularly attractive, since such tandems may increase the PCE of c-37 Si PV to values higher than its single-junction thermodynamic limit;⁴ PCE is a key driver of low 38 39 levelized cost of electricity (LCOE) at the PV-system level.⁵

Perovskite/c-Si tandem research was initially focused on stacked, four-terminal (4T) tandems owing to the simpler fabrication process.⁶⁻⁸ However, recent advances in device processing have enabled the two-terminal (2T) architecture, whose optical advantages have enabled the highest PCE for perovskite/c-Si tandems.⁹⁻¹¹ For the latest record performance, a front-flat c-Si cell was used; however, from both a cost and efficiency perspective, it is advantageous to use double-side textured c-Si cells.^{12,13} Despite this recent progress, further improvements in performance are necessary to push perovskite/c-Si tandems towards market readiness.

47 Bifaciality offers further increases in the energy yield of c-Si PV, and can be easily 48 implemented using silicon heterojunction (SHJ) technology by replacing the opaque rear metal 49 contact with grid metallization.⁵ As the rear-side of the cell is transparent, reflected and 50 scattered light from the surroundings (*i.e.* albedo) contributes to power generation.¹⁴ For 51 optimized single-junction devices, the generated device current increases linearly with the 52 albedo.

53 Calculations have shown that perovskite/c-Si tandems can also benefit from bifaciality.¹⁵⁻¹⁸ 54 Coletti et al. Indeed, recent works have recently explored this for 4T tandems, which offer a 55 relatively easily implementable testing platform.¹⁹ However, in the monolithic configuration, 56 bifacial tandems require judicious re-engineering of the perovskite bandgap for this purpose. As shown in the provious calculations by Onno et al., 16,20 as top and bottom cells feature larger 57 and smaller bandgaps respectively, the albedo will only increase the current generated in the 58 bottom cell. In tandems optimized for monofacial use, this may lead to tandem-current 59 60 mismatch and so a reduction in PCE, which can have a drastic impact on the system-level performance-as Dupré et al. predicted.¹⁷ Therefore, as for conventional current-matching 61 optimizations, the effect of the albedo should be accounted for by adjusting the thickness and 62 bandgap of the perovskite top cell.^{15-17,21,22} 63

Monofacial perovskite/c-Si tandems require a perovskite bandgap close to 1.7 eV,²³ which can be achieved by increasing the bromide-to-iodide ratio in the perovskite crystal.²⁴ However, this may result in phase segregation under prolonged light exposure, leading to device degradation.²⁵ Here we show that efficient bifacial tandems, in agreement with recent theoretical predictions, require a narrower perovskite bandgap to achieve current-matching, with a close to pure-iodide composition, thereby improving the operational stability of tandems and increasing their energy yield. **Commentato [TG1]:** We try to avoid naming individual researchers when citing their papers. Could you please revise the text accordingly?

Commentato [MDB2R1]: We revised the text and remove all naming references

71 Perovskite/silicon bifacial tandems

72 In the field, solar photons that strike the rear side of the device mainly originate from three sources: direct and diffuse sunlight reflected off the ground and surroundings, as well as diffuse 73 74 sunlight scattered in the atmosphere (Fig. 1a). Direct and diffuse light reflected by the ground is commonly referred to as albedo (adimensional); we use the term rear irradiance (in units of 75 76 mW/cm²) to refer to artificial rear-side illumination in the lab, which we use to study bifaciality. 77 To characterize the performance of tandems, we use PCE (%) for measurements at standard 78 test condition (i.e., under monofacial standard test conditions, AM1.5G spectrum, 1-Sun frontside illumination), and power generation density (PGD, in mW/cm²) for measurements under 79 STC with additional rear irradiance and test field measurements. When referring to the PGD at 80 a specific rear irradiance, we use the bificiality factor (BiFi), to indicate the intensity of the rear 81 irradiance (*i.e.* PGD_{BiFi 200} 26, means 26 mW/cm² with 200 W/m² of rear irradiance) 82

83 To understand the impact of albedo on the performance of bifacial perovskite/c-Si tandems, 84 we developed such devices with different perovskite bandgaps. Our tandem layout consists of a 85 both-sides textured silicon heterojunction (SHJ) bottom cell, onto which the perovskite top cell 86 is deposited by solution processing in the *p-i-n* configuration (implying electrons are collected 87 at the sunward side). Figures 1b and 1c sketch this tandem and show a cross-sectional scanning 88 electron micrograph (SEM), respectively. To increase the bifaciality, the SHJ rear contact was optimized to combine minimized series resistance and maximal albedo coupling into the c-Si 89 90 cell (Fig. 1d).

Commentato [TG3]: Please provide three or four section headings in the main text.

These should relate to the content of the article rather than being generic (ie, avoid Results, although it is fine to use Introduction and Conclusions/Discussion). Headings should be no longer than 60 characters (including spaces) and should not use punctuation.

Commentato [MDB4R3]: The main text is divided with three headings: Perovskite/silicon bifacial tandems Optical analysis Outdoor data and field-test performances Introduction and conclusions do not have headings

91 We experimentally fabricated bifacial perovskite/c-Si tandems with five different perovskite bandgaps (1.59, 1.62, 1.65, 1.68, and 1.7 eV; values determined from 92 photoluminescence spectroscopy, Fig. S1) by altering the iodide-to-bromide ratio. Fig. 1e and 93 Table S1 show the statistical distribution of the PV parameters for the tandem cells with 94 different perovskite bandgaps, measured under monofacial STC conditions. As expected, the 95 wider the perovskite bandgap, the larger the open circuit voltage (V_{oc}) of the tandems. $J_{sc tandem}$ 96 reaches a maximum at a perovskite bandgap of 1.68 eV, corresponding to optimal current 97 98 matching between the subcells of the tandems discussed here, and resulting in an independently-certified PCE of 25.2% under STC conditions (Fig. S2). Perovskite bandgaps 99 100 smaller than 1.68 eV result in a lower overall J_{sc tandem}, as the c-Si sub-cell becomes current-101 limiting. Similarly, perovskite bandgaps larger than 1.68 eV also result in a lower overall 102 J_{sc tandem}, as now the perovskite sub-cell becomes current-limiting. The fill factor is slightly higher under silicon-limited conditions than under perovskite-limited conditions, which is in 103 agreement with other reports.^{26,27} Overall, the PCE under STC conditions remains close to 25% 104 105 for tandems with perovskite bandgaps of 1.65, 1.68, and 1.7 eV.

To investigate experimentally the role of rear irradiance, we measured the bifacial tandems by placing them between two solar simulators. The front illumination (perovskite-side) was kept at 1-Sun (100 mW/cm²), whereas the device rear (silicon-side) was illuminated with intensities ranging from 0 to ~95 mW/cm² (*i.e.* 0.95 Suns equivalent); Fig. S3 shows the detailed characterization set-up. To facilitate contacting and prevent cell degradation during the experiment, the devices were vacuum-laminated between two sheets of glass, using butyl rubber as edge-sealant. Here, we note that we used single-lamp solar simulators for practical 113 convenience; these simulators are not ideal for the tandem configuration as their spectra vary

somewhat from the AM1.5G spectrum (see Fig. S3 for more details).²⁸

Fig. 1f compares the J-V curves of a bifacial tandem (perovskite bandgap of 1.62 eV), 115 116 before encapsulation measured with an LED-based solar simulator (yellow) and after encapsulation measured with the bifacial setup without rear irradiation (red). For the latter, the 117 118 reduction in the J_{sc} (1-1.5 mA/cm²) is caused both by the glass-encapsulation, (front glass 119 reflection and suboptimal refractive-index matching of the glass/vacuum/top-electrode stack 120 that increases the reflection losses), but also by the different frontside solar simulator used in the bifacial setup (Fig. S3). Fig. 1f also shows that the bifacial tandem (1.62 eV-yellow) 121 generates a slightly lower current (~0.5 mA/cm²) in monofacial operation mode, when 122 compared with an opaque metal rear-electrode; the latter aiding internal light trapping in the c-123 Si cell (1.68 eV-blue).²⁹ However, in the presence of 20 mW/cm² rear irradiance (orange), 124 J_{sc tandem} clearly surpasses its monofacial counterparts. Here we underline that such an albedo is 125 realistic for industrial solar parks optimized to operate with bifacial modules. In the near future 126 127 it is likely that albedos resulting in rear irradiances as high as 30 mW/cm² can be achieved, e.g. 128 with the implementation of a reflective coating covering the ground and proper site selection.³⁰

129 <u>To thoroughly explore the bifacial configuration, i</u>⁴n Fig. 1g₇ we explore the change in 130 device performance as a function of rear irradiance, ranging from 0 to ~95 mW/cm², of 131 encapsulated bifacial tandems with different perovskite bandgaps. In general, the tandem V_{oc} 132 slightly increases with rear irradiance by around 20 mV, as expected, given the higher density of 133 photo-generated charge carriers in the bottom cell. However, J_{sc_tandem} is the parameter that 134 benefits most from the presence of albedo. As the rear irradiance increases, J_{sc_tandem} rises **Commentato [TG5]:** This paragraph is quite long. We suggest you to break it into more paragraphs to improve readability.

Commentato [MDB6R5]: We split the paragraph, separating fig. 1f, 1g, and 1h. The paragraph of fig. 1g describe the key concepts of the bifacial experiments in a concise way, despite its length (35 lines)

rapidly, plateauing at ~20 mW/cm² of rear irradiance, for most bandgaps tested. The 135 enhancement in J_{sc tandem} with rear irradiance is most pronounced for the narrower bandgap 136 137 perovskites tested (1.59, 1.62 eV). The reason is that both sub cells simultaneously generate more current: the perovskite top cell due its smaller bandgap, and the c-Si bottom cell due to 138 the rear irradiance. Both experimental and calculated data show that with decreasing 139 bandgaps, the rear irradiance required to achieve current-matching slightly increases. The 140 141 effect of albedo on the FF is more complex. For all band gaps, the FF slightly drops as the rear 142 irradiance increases from 0 to 20 mW/cm² before partially recovering at irradiations higher than ~20 mW/cm². A similar correlation between FF and current-matching conditions is well 143 144 known for monofacial tandems when spectrally changing the incident solar radiation, as 145 recently show by Köhnen et al. and Boccard et al. 26,27 For a detailed explanation of this 146 phenomenon, we refer the reader to Section S4. As demonstrated in Fig. 1g and Table S1, J_{sc} strongly increases with stronger rear irradiance up to values around 10-20 mW/cm², which 147 empirically demonstrates the extent to which the tandems tested under monofacial, STC 148 149 condictions are current limited by the c-Si bottom cell. Along with improved current matching, the FF slightly decreases, as previously shown from basic two-diode considerations (see also 150 Fig. S4c). For a rear irradiance exceeding 20 mW/cm², the tandems enter the regime of current 151 limitation by the perovskite top cell as no further enhancement in J_{sc} is observed with 152 153 increasing rear irradiance. For this regime, the tandem again shows a slightly increased FF. As 154 stated earlier, in the presence of albedo, we use PGD (in mW/cm²) rather than PCE (%) to indicate the performance of bifacial tandems. Similar to the J_{sc} trend, the PGD of the bifacial 155 156 tandem strongly benefits from the addition of rear irradiance, achieving values as high as ~28

157 mW/cm² for perovskites with band gaps of 1.59 and 1.62 eV (with ~95 mW/cm² of rear irradiance: PGDBIFI950 27.85 mW/cm²). Notably, our measurements show that a rear irradiance 158 of 30 mW/cm² can improve the absolute PGD of a bifacial tandem (with 1.59 eV perovskite top 159 cell) by more than 25% with respect to its monofacial configuration (see Fig. S5 for more 160 details). Such an albedo is realistic in solar fields, where snow, sand or concrete may cover the 161 ground surface.^{22,31} When compared with monofacial perovskite/c-Si tandems, this 162 enhancement in power output favors bifacial technology over several monofacial 163 164 configurations, as shown in Table S3, underlining the potential of this technology.

To test our findings, we analyzed the enhancement in PGD for a batch of 29 bifacialtandem cells (with a perovskite bandgap of 1.59 eV), with and without 30 mW/cm² of rear irradiance (Fig. 1h; PGD_{BIFI0} blue, PGD_{BIFI300} yellow). Without albedo, the devices show a distribution of PGD_{BIFI0} centered at 21.5 mW/cm². Conversely, with an rear irradiance of 30 mW/cm² the overall PGD_{BIFI300} increases, and the average shifts to 25.5 mW/cm², an absolute increase of 19 % in power generation.

171 With rear irradiance, the operating temperature of the tandem is increased. In Fig. S6, 172 we investigate the temperature variation under different rear irradiance conditions alongside 173 their relative cooling relaxation times. Based on these cooling times, we established a minimum time-interval between the sequential measurements carried out in the lab for Fig. 1g, in order 174 to ensure a cell temperature close to STC conditions. However, the outdoor operational 175 temperature of a solar cell (especially in a sunny and hot climates) can reach 50 °C and more 176 177 (Fig. S7), even for perovskite/c-Si tandems where thermalization losses are significantly reduced 178 compared to single-junction devices.

Formattato: Rientro: Prima riga: 1,27 cm



179

180 Fig. 1. Perovskite/silicon bifacial tandems. (a) Sketch of light absorption in a bifacial perovskite/c-Si tandem, 181 featuring albedo. (b) Cross-sectional sketch of the perovskite/c-Si bifacial tandem. (c) Cross-section SEM 182 micrograph of the tandem realized on both-sides textured c-Si bottom cell, white scale bar: 1 µm. (d) Picture of the 183 front and rear contact of the device. (e) Photovoltaic performance for bifacial tandems with different perovskite 184 bandgaps, measured only with front light (1 Sun)(including: minimum, maximum ticks; median, lower quartile, and 185 upper quartile in the box plot). (f) Comparison of the J-V curves of a monofacial tandem (blue) and a bifacial 186 tandem (dark yellow) measured using an LED-based solar simulator. The same bifacial device encapsulated and 187 measured in the bifacial setup with front light only (red) and front light plus rear irradiance (orange). Solid line: 188 reverse voltage scan direction. Dashed line: forward voltage scan direction (g) Photovoltaic performance of bifacial

Commentato [TG7]: Could you please define all features – square, box lines, and ticks – of box-and-whisker plots?

Commentato [MDB8R7]: Included

Commentato [TG9]: Could you please specify what the dashed and solid curves refer to?

Commentato [MDB10R9]: done

tandems with different perovskite bandgap, as a function of the rear irradiance. (h) Statistical distribution of the power generation of 29 tandems measured with (yellow, BiFi300) and without (blue, BiFi0) rear irradiance. The fits
 are included as a guide for the eye.

192

193 Optical analysis

194 To further understand current-matching conditions for bifacial tandems, we collected external quantum efficiency spectra (EQEs) for the devices with different perovskite bandgaps 195 196 (Fig. 2a). By integrating the EQE-weighted solar spectrum, we can extract the current-matching 197 condition (for the monofacial tandem case), which is achieved for a perovskite bandgap 198 between 1.68 eV and 1.7 eV, in agreement with the trend for J_{sc} shown in Fig. 1e. To visualize the influence of the bandgap of the perovskite on J_{sc_tandem}, Fig. 2b plots the integrated currents 199 200 derived from the EQEs in Fig. 2a (closed circles) for both the perovskite (red) and silicon (blue) 201 sub-cells vs. the perovskite bandgap; we note that altering the perovskite bandgap does not notably alter its refractive index, and therefore the overall reflection of the tandem (Fig. S9). 202 We further compare these currents with those obtained from J-V measurements (Fig. 1g), 203 hollow circles for bifacial devices with ~95 mW/cm² rear irradiance (red) and without effective-204 205 albedo (blue). Fig. 2b again demonstrates that while a 1.7 eV perovskite bandgap is optimal for 206 monofacial tandems, in the bifacial configuration this offers little to no gain in current. For small 207 bandgaps (e.g. 1.59 eV, Fig. 3b), the 1-Sun integrated EQE shows a remarkable mismatch in current, due to a current limiting c-Si sub-cell. However, while this is disadvantageous in a 208 monofacial configuration, it enables the highest current gain in the bifacial configuration, 209 210 provided that sufficient rear irradiance is available.

Commentato [TG11]: Could you please describe the blue and yellow curves as fits or guides to the eye (details about the fit should be provided, if relevant)?

Commentato [MDB12R11]: We included the fit as a guide for the eye

Commentato [MDB13]: Second heading

ha formattato: Tipo di carattere: Grassetto

211 To further analyze possible loss mechanisms due to optical effects as a function of the layer stack, we performed optical simulations. Fig. 2c and Fig. 2d show the front and rear side 212 absorptance, respectively, considering the layers of the stack of Fig. 1b for a perovskite 213 bandgap of 1.68 eV (see Fig. S10 for details). The indium zinc oxide (IZO) and fullerene (C_{60}) top 214 layers cause significant parasitic absorption in the UV regime. Moreover, the IZO layer also 215 induces losses due to free carrier absorption between 800 and 1100 nm, a range where the c-Si 216 bottom features a high quantum efficiency, affecting thus significantly the current output. 217 218 Overall, under AM1.5G 1-Sun illumination, parasitic absorption and reflection losses translate into J_{sc} losses of 4.6 mA/cm² and 3.1 mA/cm², respectively; IR light transmission results in 219 220 another 0.9 mA/cm² in J_{sc} loss. Photons impinging on the bifacial tandem rear can only be 221 absorber by the c-Si bottom cell. Here, high energy photons could be parasitically absorbed in 222 the rear-contact stack of the SHJ cell. The 2-side textured c-Si wafer aids in geometric light trapping, reducing reflection losses in the 600 - 1000 nm wavelength range. For wavelengths 223 around 500 nm a significant reflection loss is apparent resulting in imperfect light incoupling in 224 225 these prototypes with the given rear-contact layer stack. Future work can address this loss by optimizing the refractive index combination in the rear stack and thereby enhancing the light 226 227 incoupling from the rear side. Finally, we extended our simulation to the encapsulated device (see Fig. S11), which, as experiments already showed, suffers from slightly increased reflection 228 229 losses.

230



231

Fig. 32. Optical analysis. (a) EQEs of the bifacial tandems with different bandgap. The orange lines correspond to the EQE of the perovskite top cell; the blue lines correspond to the EQE of the silicon bottom cells-. (b) Comparison between integrated EQE current_density and Jsc from J-V curves. Note that the EQE measurements were performed without encapsulation, but the J-V measurements were done with encapsulation, which lowers the J_{SC} compared to EQE values. Full dots: integrated EQE current_density for the perovskite (red) and c-Si (blue), as a function of the perovskite bandgap. Hollow circles: J_{sc} of the bifacial device without rear irradiance (black) and with rear irradiance (gray, ~95 mW/cm²), as a function of the perovskite bandgap, extrapolated from Fig. 2c. (c) Front and (d) rear side absorption of the layers composing the bifacial tandem with a perovskite bandgap of 1.68 eV.

240

241 Outdoor data and <u>f</u>Field-test performances

242 To further test the potential of the technology, we compared the outdoor performance

243 of monofacial and bifacial tandem devices under three different specific albedo conditions:

concrete, synthetic grass, and a white background. We installed the monofacial and the bifacial

devices in our outdoor test field on the KAUST campus and changed the ground material to

Commentato [TG14]: Could you please make clear in the caption or in the figure legend what the orange and blue curves refer to?

To improve the figure readability, it might be better if the legend is moved outside the plot and the boxes from each portion are removed.

Commentato [MDB15R14]: Fig 2a is improved and the caption modified accordingly

Commentato [TG16]: Could you please specify in the label axis and in the legend that Integrated EQE refers to a current density?

Commentato [MDB17R16]: Fig 2b is modified, we accepted the changes in the caption

Commentato [TG18]: Correct?

Commentato [MDB19]: Third heading

12

simulate these different albedo conditions (see Fig. S12 for more details, including the reflectance data from these surfaces). In this way, the performance relies on albedo rather than rear irradiance.

249 For each condition, we recorded the J-V characteristic with a time interval of 10 min during a measurement time of one hour. To achieve maximum consistency, we carried out the 250 experiments at peak sun hours, using a pyranometer and a calibrated c-Si solar cell to monitor 251 252 the light intensity. Fig. 3a shows the PGD and the J_{sc} of the bifacial (black) and monofacial (red) 253 tandem devices. Both devices consist of the same layer stack apart from the rear electrode (opaque vs. transparent electrode) and perovskite bandgaps. The bifacial tandem outperforms 254 its monofacial counterpart for every albedo condition. The gain in performance was particularly 255 striking when using concrete as the ground, where the bifacial tandem achieved a remarkable 256 257 PGD of 25.9 mW/cm². The increase in power output can be mainly attributed to the higher currents generated in the bifacial configuration. Overall, the average increase in bifacial power 258 output was 20% for concrete, 6% for a white background, and 4.3% for synthetic grass. We note 259 260 that certain materials such as snow have typically an even larger albedo than concrete.

We extended the comparison between monofacial and bifacial tandems to two testfield locations: Jeddah – Saudi Arabia, representing hot and sunny environments; and Karlsruhe – Germany, representing a typical moderate climate. Fig. 3b shows the PGD of the bifacial tandems, from dawn (06.00 am) to dusk (06.00 pm), measured at 10 minutes intervals, over five days of investigation. To highlight the different irradiation conditions, we did not normalize the output power density to Sun equivalents, but rather reported the Sun's intensity, obtained through a pyranometer and a calibrated c-Si reference cell. For the experiment, the cells were placed on a test-field structure, with similar orientation and distance from the ground, consisting of bright sand and concrete (Jeddah), and concrete (Karlsruhe). In both sites, the bifacial tandem performed significantly better than the monofacial one, particularly, during midday when the light intensity is close to 100 mW/cm² (Jeddah) or 80 mW/cm² (Karlsruhe). Furthermore, the Karlsruhe data reveals that the enhancement in PGD is more pronounced on sunny days (day 1, 2), which predominantly exhibit direct radiation compared to cloudy days with mostly diffuse irradiation (day 3, 4, 5).



275

Fig. 3. Outdoor testing of bifacial tandems. (a) Comparison of PGD and J_{sc} for bifacial (black) and monofacial (red)
 tandems with different albedo conditions: concrete, styrofoam, and grass. The perovskite bandgaps are 1.62 eV
 and 1.68 eV for the bifacial and monofacial tandems, respectively. For each point, ∓the performances current

Commentato [TG20]: Can you please describe the two different labels used for the X-axis (i.e. day time on top and the 0-50 range at the bottom)?

Commentato [MDB21R20]: We revised Fig 3a and use the same X-axis, top and bottom. In the previous version the bottom axis was in minutes.

279 density and therefore the PGD_are normalized with the respective Sun-solar irradianceintensity (blue line). The 280 gray areas in the plot represent the operational time to change the setup, from one background to another. (b) 281 Test-field power conversion comparison for bifacial (black) and monofacial (red) tandems, measure over five days 282 in two different locations: Jeddah, Saudia Arabia (22.302494, 39.110737); Karlsruhe, Germany (49.094577, 283 8.429605). For the Jeddah experiment, the devices were placed in the test-field in different times, therefore we 284 285 reported the intensity of the solar irradiance (red for monofacial, and black for bifacial) for each day. For the Karlsruhe experiment, the devices were placed in the test-field at the same time, therefore we reported only a 286 single solar irradiance (red). For each day and location we report the intensity of the solar irradiance. For each 287 device, the perovskite bandgap is 1.62 eV.

288

289 In Fig. 4a and 4b, we report the analysis of performances of the bifacial (4a) and 290 monofacial (4b) tandems with respect to the solar irradiance from the five days of field data 291 collected from the Jeddah location. Since the data are collected under different solar 292 irradiations, we normalized the J_{sc_tandem} and the PGD for direct comparison. For the J_{sc_tandem}, the trend of the monofacial tandem is linear. Conversely, for the bifacial tandem the current 293 294 shows some hysteresis with the solar irradiance, showing sub-linear behavior during the morning (from 06:00 am to 12:00 pm) that becomes linear in the afternoon (from 12:00 pm to 295 06:00 pm). The scattered data at low irradiance (10-25 mW/cm²) are an artifact induced by a 296 297 partial shading of the pyranometer during early mornings and late afternoons (Fig. S13). To understand the behavior of the current in the bifacial tandem, we measured the albedo of our 298 test-field over five consecutive days (Fig. S14). We found that, while the albedo is on an average 299 constant during the week (~0.25), it fluctuates during the day, with lower values in the morning 300 due to partial shading of the ground. This reflects the importance of controlling the albedo to 301 maximize the performance of the bifacial tandem. The trend of the V_{oc} is similar for both 302 devices, where the voltage reaches a constant value at solar irradiances of 15-20 mW/cm². The 303 FF shows a narrower distribution for the bifacial device, particularly at low solar irradiance, 304 305 without evident differences between the morning and the afternoon. Interestingly, for the

Commentato [TG22]: Could you please provide further details?

Commentato [MDB23R22]: The whole experiment lasted for three hours outdoor, during this time the intensity of the Sun changed (blue line). To be able to precisely compare the different conditions of albedo, we normalized the current (and then the PGD) to the Sun intensity.

Commentato [TG24]: Why the Jeddah location has both a bifacial (black) and monofacial (red) related irradiance while the Karlsruhe one does not?

Commentato [MDB25R24]: For Karlsruhe the two devices were installed simultaneously, therefore they were exposed to the same solar irradiance. We clarified this in our response letter after the first revision.

To better clarify this point, we modified the caption accordingly.

bifacial tandem, the normalized PGD reflects the effect of the lower current during the
morning, to improve in the afternoon. Overall, the normalized PGD distribution is similar for the
two devices over the day.





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Commentato [TG26]: Is this correct? Commentato [MDB27R26]: correct

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318 Finally, we performed energy yield simulations to assess the performance of bifacial tandems

319 under realistic outdoor conditions (see results in Fig. 4c). Two locations, Phoenix and Seattle,

320 have been chosen to represent two very different climatic conditions. Moreover, the annual

energy yield was computed for different perovskite bandgaps and albedo conditions (see 321 322 Section S15 for more details). The highest monofacial energy yield is achieved with a perovskite 323 bandgap of 1.68 eV and 1.65 eV for both locations, using the identical layer stack of the bifacial architecture and an optimized perovskite thickness, respectively. Despite not featuring an ideal 324 bandgap for monofacial tandems (between 1.70 eV and 1.80 eV), this bandgap results in the 325 most optimal current matching throughout the whole year for a 1000 nm thick perovskite layer 326 327 and thus achieves the highest energy yield. The lowest monofacial performance occurs for the 328 smallest bandgap (1.59 eV), due to significant current-mismatch losses. The scenario changes significantly for the bifacial configuration. Even in the presence of a ground with a reflectivity as 329 330 low as dark concrete (average albedo reflectivity equal to 28%), the optimum bandgap shifts to 331 lower values: in Seattle, which represents a temperate climate, as well as in Phoenix, which 332 represents a sunny, desert climate, it is 1.59 eV, due to the larger share of direct sunlight, resulting in a stronger rear irradiance. Notably, bifacial energy yield improvements of around 333 32% in Seattle and 37% in Phoenix (relative to the best monofacial tandems with optimized 334 335 layers thicknesses and a bandgap of 1.65 eV) are computed with a bandgap of 1.59 eV for the perovskite and the most reflective ground. Materials with high reflectivity could be used to 336 enhance the albedo in locations with a high share of direct irradiation, to fully exploit the 337 potential of bifacial perovskite/c-Si tandems with narrow perovskite bandgaps. It should be 338 339 noted that in order to maintain generality, the energy yield calculations provided in Figure 4c 340 do not consider installation specific aspects such as self-shading of the module or shading due to adjacent modules. However as shown in Fig. S16 and Fig. S17, the key trends presented are 341 342 valid for representative installation scenarios that would consider such shading.

Commentato [TG28]: Folowing the concerns raised by Reviewer #1, could you please be more explicit that this might means the numbers are overestimated?

343 Conclusions

344 We have experimentally shown how bifaciality can be used to enhance the performance of 345 monolithic perovskite/c-Si tandems. The device configuration with a transparent back electrode 346 relies on the albedo to enhance the current generation in the bottom cell, while simultaneously enhancing the current generation in the perovskite top cell, thanks to a narrower perovskite 347 348 bandgap. This matching is achieved for a 1.59-1.62 eV bandgap perovskite, where the bromide 349 content is minimized, thereby strongly reducing the stability issues related to halide 350 segregation. We evaluated the bifacial tandem performance in test-field experiments and we predicted the energy yield for bifacial and monofacial tandem configurations in different 351 climates. In both cases, the bifacial tandem outperformed the monofacial configuration, 352 validating the promise of this technology. This work demonstrates the potential for a new class 353 354 of efficient solar cells, which can close the gap with the 30 mW/cm² power generation density barrier, for a highly performant and affordable technology. From here, further improvements in 355 device performance and scaling-up of the technology are logical next steps, bringing this 356 357 technology closer to the PV market.

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361 Methods

362 Device Fabrication: Silicon heterojunction bottom cells are fabricated on float-zone double side-textured four 363 inches wafers (TOPSIL, n-doped, resistivity 1-5 ohm/cm, and thickness 250-280 mum). The wafers are processed 364 with alkaline solution to obtained random pyramids texturing. After the wafers are cleaned in RCA1 and RCA2 365 solutions and dipped in hydrofluoric acid to remove the silicon oxide layer. The intrinsic I (5 nm) and doped 366 amorphous and nanocrystalline layers (p, n 12 nm and 40 nm respectively) are deposited via plasma enhanced 367 chemical vapour deposition (PECVD) in an Octopus2 cluster (Indeotec). The ITO rear contact (100 nm) and the 368 recombination junction (15nm) are sputtered in the physical vapour deposition (PVD) part of the Octopus2 cluster 369 with base pressure 1 x 10⁻⁵ Torr, 13.56 Mhz RF source, 0.9 W/cm² power density, in an Ar/O₂ atmosphere (0.8% O₂ 370 content) and a process pressure is 1 x 10⁻³ Torr (ITO target from Vital Thin Film materials – 97% In₂O₃ 3% SnO₂).

371 After the PVD deposition the bottom cells are annealed 5 min at 200 °C. For the top cell, NiOx (17 nm, 372 Plasmaterials) is sputtered (Angstrom EvoVac) at base pressure of $< 1 \times 10^{-6}$ Torr in pure Ar atmosphere with no 373 intentional heating or cooling of the substrate, at a power density 1.97 W/cm² and 13.56 MHz RF source.³² Prior to 374 the perovskite deposition the NiOx layer is passivated with 4-bromobenzoic acid (Sigma Aldrich). The process is 375 done by spin casting of 2 mg/ml 4-bromobenzoic acid in ethanol. After spin casting the films were annealed at 90 376 °C and after cooling down washed with ethanol for several times.. The triple cation perovskite solution (1.68 M) is 377 prepared in a 4:1 Dimethylformamide Dimethyl sulfoxide (DMF:DMSO, Sigma Aldrich) using 36.4 mg of caesium 378 iodide (CsI, Alfa Aesar), 44,8 mg of methyalmmonium bromide (MABr, Greatcell), 389 mg of fomamidinium iodide 379 (FAI, Greatcell), lead bromide (PbBr₂, Sigma Aldrich) and lead iodide (PbI₂, Alfa Aesar). The solution was stirred 380 until complete dissolution of the precursors. The Pbl2 and PbBr2 amount varied according to the desired bandgap. 381 For the perovskite film formation, the perovskite precursors is spin coated on the bottom cell substrate with a 382 three-steps process: initially 600 rpm, then 2000 rpm, finally 7000 rpm. During the acceleration between the 383 second and third step, anisole is dripped as solvent quencher. Finally, the devices are annealed in nitrogen at 100 384 °C for 15 min. On top of the perovskite, lithium fluoride LiF (1 nm Alfa Aesar) and C $_{60}$ (20 nm, NanoC) are thermally 385 evaporated as electron transport layer (Angstrom EvoVac). A layer of 20 nm of tin oxide (SnOx 386 Tetrakis(dimethylamino)tin and H₂O as precursors, with N₂ as the gas carrier) deposited via atomic layer deposition 387 (ALD, Picosun) is used as protective buffer layer. As top electrode, 110 nm of indium zinc oxide are sputtered in an 388 Angstrom EvoVac sputtering system (base pressure $< 1 \times 10-6$ Torr) with RF power of 42 W (90% In₂O₃/ 10% ZnO, 389 99.9% Plasmaterials). To functionally contact the top and bottom transparent electrodes, we thermally evaporated 390 (Angstrom EvoVac) 350 nm of silver contacts (base pressure 1 x 10⁻⁶ Torr), on the front and afterwards on the rear 391 of the tandem using an aperture mask. Lastly, 95 nm of MgF2 as antireflection film are thermally evaporated 392 (Angstrom EvoVac) on top of the bifacial device.

393 Device Characterization: To evaluate the performances of the tandems without rear irradiance, we used a 394 calibrated Wavelabs Sinus 220 LED based solar simulator with AM 1.5G irradiance spectrum as our light source and 395 we coupled it with a Kiethley 2400 series SourceMeter to take the J-V measurements. The data is recorded via a 396 homemade MATLAB based software. The solar cells are measured from -0.1 V to + 1.9 V at 200 mV/s in both 397 forward and reverse scan directions and the illuminated area, defined by a laser cut shadow mask, is 1.03 cm². EQE 398 measurements are performed using PV-Tools LOANA equipment. For the rear irradiance setup we used a Abet 399 Technologies Sun 3000 Class AAA and a Newport Oriel Sol3A Class AAA, both Xenon (Xe) arc lamp based. For the 400 rear irradiance measurements, the stability test, and the field-test investigation, we encapsulated the bifacial 401 tandem with a vacuum laminator (Ecolam 5 Ecoprogetti) using glass and butyl rubber 10 mm wide and 1 mm thick 402 butyl rubber Solargain[®] edge sealant with desiccant (Quanex, SET LP03).-

403 Test field experiment: For the field-test, we used an I-V tracer from EKO (model MP-160). The I-V characteristics of 404 multiple samples are probed successively using the multiplexers MI-520 again from EKO. Current-voltage curves 405 are acquired with a scan rate of 200 mV/s, and we measured all physical parameters with a time interval of 10 min. 406 The global horizontal irradiance on the plane of the devices is measured using the pyranometer MS-802 (EKO), 407 mounted on the same structure as the devices. The solar cells are mounted on a structure with a tilt angle of 25 408 degrees and South orientation, located in KAUST's outdoor testing field on the KAUST campus, near the village of 409 Thuwal (Saudi Arabia; 22.302494, 39.110737). Furthermore, solar spectra is acquired using the spectrometers 410 QE65PRO (visible spectral region) and NIRQuest512 (NIR spectral region) from Ocean Optics. The spectrometers 411 are built into a temperature-controlled housing, and possess a wavelength resolution of < 2 nm across the entire 412 VIS/NIR. For the field-test in Karlsruhe, we used a Keithley 2600 series SourceMeter to record I-V curves with a 413 time interval of 3 minutes. A homemade LabVIEW program is used to select successively the two solar cells using 414 multiplexers and save the data. Then through a MATLAB code the MPP of each curve is extracted. The solar cells 415 are mounted on a homemade metallic frame with a tilt angle of 45 degrees and South orientation. A calibrated c-Si 416 solar cells mounted next to the bifacial cells is used to extract the Suns, by computing the ratio between the short-417 circuit current in the test-field for each data point and the short-cicruit current under a solar simulator with 418 AM1.5G irradiance spectrum.

Simulations and Enery Yield Modelling: The optical simulations and energy yield modelling platform are accurately
 described by Schmager et alelsewhere. ³³ Here we provide a short description of its main features. The modelling
 platform combines four modelus together: (i) optics module, (ii) irradiance module, (ii) electrics module and (iv)

Commentato [MDB29]: Included

422 enrgy yield core module. For the simulations in Fig.2c, Fig.2d and Fig.S6, the optics module alone was used. This 423 module employs a combination of transfer matrix method (TMM) for thin, optically coherent layers and series 424 expansions of Lambert-Beer law for optically thick layers, taking into account multiple reflections at contiguous 425 interfaces. Textured interfaces were handled using geometrical ray tracing, as suggested by Baker-Finch and McIntosh.³⁴ To model as closely as possible the fabricated devices, complex refrative indeces of most of the layers 426 427 were measured in-house at KAUST. The output of the module is stored in multi-dimensional matrices, namely the 428 reflectance matrix, the transmittance matrix and the absorptance matrix for each layer in the stack. Each matrix is 429 spectrally and angularly resolved for a discrete number of photon wavelength and incoming angle. Data for normal 430 incidence were used for the optical simulations in Fig.3c, Fig.3d and Fig.S6. For the energy yield simulations, the 431 remaining three modules work together with the optics module. The irradiance module uses typical 432 meteorological year (TMY3) data sets from the National Renewable Energy Laboratory (NREL) to compute 433 angularly and spectrally resolved clear sky irradiance data of hundreds of locations in the USA with a time 434 resolution of one hour, using the 'simple model of atmospheric radiative transfer of sunshine' (SMARTS).^{35 36} Then 435 a simple model is used to account for cloud coverage, in order to obtain realistic direct and diffuse irradiance 436 data.³⁷ The energy yield core module combines the otput of the irradiance and optics module to compute the light-437 collected current J_{nh} in the perovskite and silicon sub-cells. In the bifacial configuration, the albedo contribution 438 was computed using reflection data from the ECOSTRESS library ³⁸. Shading due to the module itself and the other 439 rows of modules was not taken into consideration. Then the electrics module computes the maximum power point 440 calling the circuit simulator LTspice. An equivalent circuit identical to the one in Fig.S2a was used for the simulations of the tandem perovskite/silicon cells. Finally, the energy yield module sums the contributions for each 441 442 hour of the typical meteorological year and extracts the annual energy yield.

443 Acknowledgments

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462 Contributions

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M.D.B conceived the idea; M.D.B. and A.J.M. fabricated the devices; Y.H., B.C., and A.S.S. developed the perovskite
bandgaps; E.A. developed the tandem top contact and layout; E.A. and F.H.I. developed the tandem hole transport
layer; T.G.A. performed the tandem simulations; M.D.B, T.G.A., E.V.K. developed the silicon bottom cell; F.G.,
U.W.P. and L.X. performed the optical modeling; J.L. performed the electrical modeling; M.F.S., F.G., J.T. and J.L.
developed the field-test setup; F.G. and U.W.P. performed the energy yield calculations; M.F.S supervised the fieldtest experiment; M.D.B., M.F.S., A.S.S., F.G., and U.W.P wrote the manuscript; D.B., B.F., E.H.S, and S.D.W.
supervised the project.

471 Data availability



Commentato [MDB31]: Revised

Commentato [TG32]: In the present case, I suggest that you introduce a DAS worded as follows: "All data generated or analysed during this study are included in the published article and its Supplementary Information."

Commentato [MDB33R32]: Included

472	All da	ta generated or analysed during this study are included in the published article and its Supplementary		ha formattato: Tipo di carattere: 10 pt
473 474	Inform	ation		ha formattato: Tipo di carattere: 9 pt
475	Comp	eting Interests		Commentato [TG34]: Please include a statement that
476 477	<u>The au</u>	thors declare no competing interests,		declares any financial and non-financial competing interests you or your co-authors may have. For more details, please see our Competing Interests policy page: <u>http://www.nature.com/authors/policies/competing.html</u>
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