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Metal halide perovskite tandem and multiple-junction photovoltaics

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Abstract | Metal halide perovskite-based solar cells have attracted considerable attention in recent years owing to their inexpensive and easy fabrication and rapidly increasing efficiencies, which already match those of the industrially dominant multi-crystalline silicon. The incorporation of perovskite absorber materials into multiple (multi-)junction cells could potentially allow us to go well beyond silicon-based technology and reach even higher power conversion efficiencies.

Layering multiple solar-absorber junctions on top of each other enables the absorption of different regions of the solar spectrum, so that more energy can be extracted from sunlight. The possibility of tuning the bandgap of perovskite materials over a wide range, along with the ability to generate high open-circuit voltages from wide-bandgap absorbers, make perovskites ideal candidates.

Perovskites can be used in combination with or as a substitute for silicon in photovoltaic technologies already in use and can be assembled in hybrid tandem architectures or layered in all-perovskite multi-junction cells. In this Review, we discuss opportunities for perovskite

multi-junction cells, explore the progress made so far, describe the theoretical possibilities and discuss perspectives and challenges for the future of this emergent technology.

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Silicon-based solar panels, the prevalent photovoltaic (PV) technology, continue to become more affordable owing to improvements in manufacturing processes, abatement of raw material and installation costs, and a progressive increase in the panel (module) efficiency. The cost decreases seem likely to continue for decades to come, although such panels are not yet inexpensive enough to compete globally with conventional fossil-fuel-based power sources without government subsidies. However, it seems inevitable that Si-based solar panels will provide the most affordable means of energy conversion in the near future, as they already do in the sunniest locations in the world1,2. The near future foresees PVs as a major component of the multi-trillion-dollar global energy mar- ket, which represents an extremely promising prospect for a low-carbon energy economy and the consequent mitigation of the impending effects of climate change. The increasingly critical repercussions of climate change call for rapid replacement of carbon-emitting power sources with renewable technologies, and many coun- tries have committed to targets that will require large-scale investment in clean technologies3.

Recent years have witnessed an increase in research and commercial activity in PVs that are based on non-silicon materials, which was originally motivated by the opportunity to deliver PV technologies funda- mentally less expensive than silicon. Any technology

potentially able to undercut the costs of silicon-based solar cells would rapidly prevail globally. However, as silicon-based solar cells decrease in cost, the challenge to outperform their power output-to-cost ratio becomes more difficult2. For perovskite-based solar cells, which remain to be implemented on a large scale and tested under outdoor conditions over extended periods of time, the path from a test cell to a large-area panel appears incredibly challenging. There are many unique chal- lenges associated with large-scale production, including how to scale-up deposition methods, connect sub- modules, design effective quality control protocols and test lifetime stability. For such a new technology, with- out the years of streamlining manufacturing that Si has had, beating the cost of Si modules will be extremely difficult. However, although the costs of silicon cells con- tinue to decrease, their power efficiency is not expected to increase much further, as it is already close to the practical maximum value4,5. Therefore, the opportu- nity is ripe for a new PV technology that delivers much higher efficiencies at a similar cost to Si-based technol- ogy. A key aspect of PV deployment is that the cost of the whole PV module is less than half the cost of the total installation, and the cost of constituent Si-based PV cells is less than half the cost of the module (including the laminated glass, metallic interconnects and electronic junction box). Less than a quarter of the fixed costs in a

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Power conversion efficiency (PCE). The most important metric of a solar cell. It is the fraction of incident solar power that is converted into electrical power at the ideal operating voltage and is defined as

PCE = *V* × *J* × FF/P , where

PV device are related to the cell, and the remaining costs primarily scale with the area rather than the power otuput of the panel. This means that increasing the efficiency of the cell will most directly result in less expensive electricity production in terms of the total installed levelized cost of electricity (LCOE) per kilowatt hour produced over a period of 25 years.

PV cells composed of tandem (or two-junction) and multiple III−V semiconductor junctions achieve efficiencies up to 46% under concentrated sunlight

— much higher values than those reported for silicon cells6. The higher efficiencies of these alternative PV cells are due to the leveraging of multiple semi- conducting materials that absorb different sections of the solar irradiance spectrum. The electrical connection of stacked semiconductors results in lower fundamental losses in the device and higher efficiencies (see the sec- tion ‘Tandem and multi-junction solar cells’). However, these cells are extremely expensive owing to the very slow growth processes needed to fabricate the required high-quality, defect-free semiconductor materials. Developing inexpensive materials that can be stacked in a similar fashion to attain equivalently high efficien- cies is thus an exciting goal of PV research and of key technological interest.

In this Review, we attempt to provide a concise but complete overview of perovskite-based multi-junction PVs. We detail the main advances that have been made and give a general picture of the field today and its pos- sible outlook. We discuss the various designs of multi- junction cells, introduce halide perovskites and their promise for incorporation in multi-junction cells and review the current state of the art. We consider both the theoretical and realistic limits for these devices, guided by modelling, and finish by considering the challenges that lie ahead for this technology.

different bandgaps. By stacking different light absorbers, the highest-energy photons are captured by the material with the largest bandgap, and lower-energy photons are allowed to pass through the layer to reach a smaller- bandgap semiconductor, enabling a more efficient use of photonenergy. This is the fundamental concept of a multi- junction solar cell, as illustrated in FIG. 1a,b. Connecting two semiconductor junctions (‘sub-cells’) with ideal bandgaps would increase the fundamental efficiency limit to 46% under the standard solar spectrum and intensity; three junctions would lead to 52% efficiency and, theoretically, infinite junctions would lead to 68% efficiency7,8. No scheme has been conceived to make an infinite-layered device, but the cells with the highest effi- ciency produced so far (~39% under normal solar light irradiance and 46% under concentrated sunlight) contain up to five stacked junctions4.

There are two main designs for tandem solar cells: two-terminal (2T) and four-terminal (4T) architectures (FIG. 1c). The 2T design involves the monolithic fabri- cation of a second sub-cell on top of the first sub-cell, whereas in the 4T design the two sub-cells are fabricated on separate substrates, operate independently and are physically stacked on top of each other. In a 2T tandem cell, the sub-cells are connected monolithically by a tun- nel junction or recombination layer (BOX 1), meaning that only two external electrical contacts are required, and only one of them must be semi-transparent to let light into the device. The 4T design involves con- necting each sub-cell separately to an external circuit, requiring four contacts for one tandem cell, of which three must be semi-transparent to allow light into both sub-cells. Because some photons will be absorbed in the semi-transparent contacts, the lower number of semi-transparent electrical contacts gives a slight overall efficiency advantage to the 2T design; however, process- ing the 2T cells is more challenging. This is especially

oc sc in

Pin is the incident power density and FF is the fill factor. It is normally defined at the standard test illumination of 100 mW cm–2 with the AM1.5 spectrum.

Short-circuit current

(*J*sc). Effectively, the maximum current that the cell can provide under standard illumination conditions due to the collection of photogenerated carriers when held at short circuit (that is, zero volts across the junction).

Open-circuit voltage

(*V*oc). The voltage built up in a solar cell under standard illumination conditions when no current is allowed to flow out of the cell. Its value depends on factors including the bandgap of the absorber, the electron–hole recombination rate, the carrier diffusion length and the defect density in the cell.

**Tandem and multi-junction solar cells** Semiconductor materials are the key components of solar cells. Upon light absorption, electrons in the valence band of a semiconductor material absorb the photon energy and move into the conduction band. A semiconductor with a particular bandgap can absorb only those photons with energies higher than its bandgap. However, the excess energy from photons with a higher energy than the bandgap is lost through a thermalization process, in which the excited elec- trons relax to the conduction band edge. The trade-off between having a low bandgap semiconductor (so that a larger section of the solar spectrum can be absorbed) and minimal thermalization loss fundamentally limits solar cells efficiency. The Shockley–Queisser limit, based on thermodynamic principles, dictates the maximum power conversion efficiency (PCE) that can be attained for a single-junction device with a particular band- gap. The highest efficiencies of ~34% can be reached by using semiconductors with bandgaps between 1.1 and 1.4 eV — Si has a bandgap of 1.1 eV. To circumvent these traditional limits of PV performance, tandem (or two-junction) and multi-junction (two or more junc- tions) solar cells make use of several absorber layers with

true for solution-processed 2T tandem devices, where the solvents used to deposit the second cell can often damage the underlying layers. We note, however, that all examples of commercially manufactured tandem solar cells, which include high-efficiency concentrator and thin-film amorphous silicon ‘micro-morph’ cells, are based on a 2T monolithic interconnected designs8.

FIGURE 1d,e shows the maximum theoretical effi- ciency as a function of the bandgap of the two sub-cells in both 2T and 4T tandem cells, assuming no losses from the contacts. The method of operation for tan- dem solar cells is similar to that for a single-junction solar cell. Each sub-cell generates a short-circuit current (*J*sc) under illumination and short-circuit conditions.

When disconnected from an external circuit, the photo-

current is zero and the sub-cells build up an open-circuit voltage (*V*oc), which arises from the electrical potential generated between the two terminals when the device is illuminated. Under operation, the cell is held by the external electronics at a voltage between *V*oc and zero, at which a current is generated. Ideally, the cell will be operated at the maximum power point condition, at which the generated power (current × voltage) is maximized; the current–voltage relationship describes

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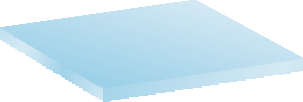
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Solar ﬂux (W m–2 nm–1)

Lower-energy photons

Higher-energy photons

1.8

1.6

1.4

1.2

1.0

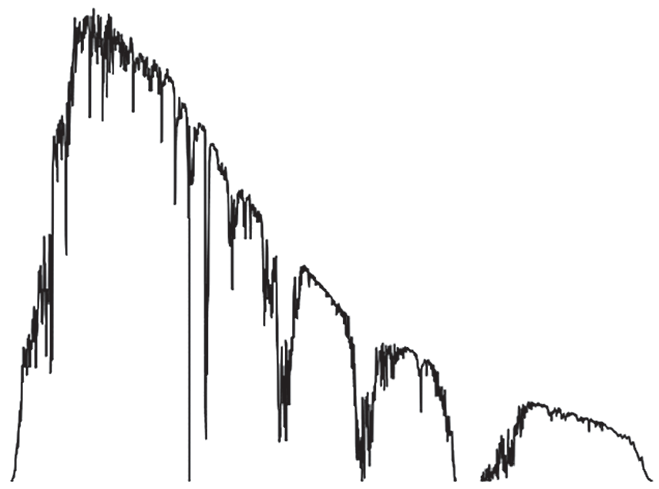
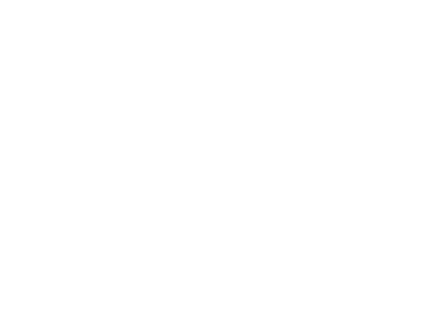
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400 600 800 1,000 1,200 1,400 1,600 1,800

Wavelength (nm)

### c

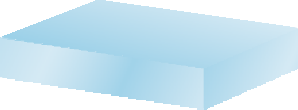
Top electrode



Selective contacts

**2T tandem 4T tandem**

Rear sub-cell



Recombination layer

Front sub-cell

ITO glass

Rear sub-cell

ITO glass Air gap

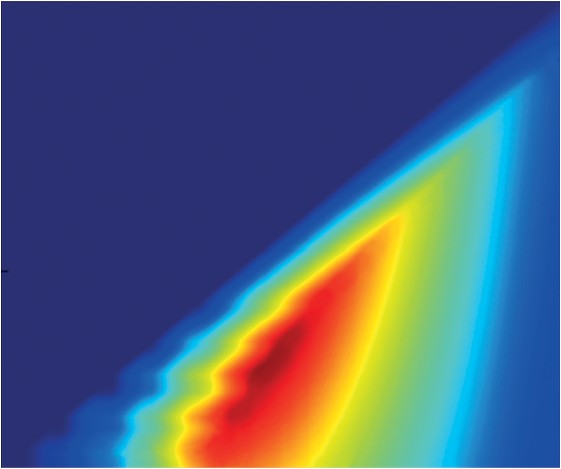
Front sub-cell

ITO glass

Wide-bandgap material Narrow-bandgap material

**d 2T tandem**

3.0



2.5

Bottom cell bandgap (eV)

2.0

1.5

1.0

0.5

0.5 1.0 1.5 2.0 2.5 3.0

Top cell bandgap (eV)

PCE (%)

46.0



40.9

35.8

30.7

25.6

20.4

15.3

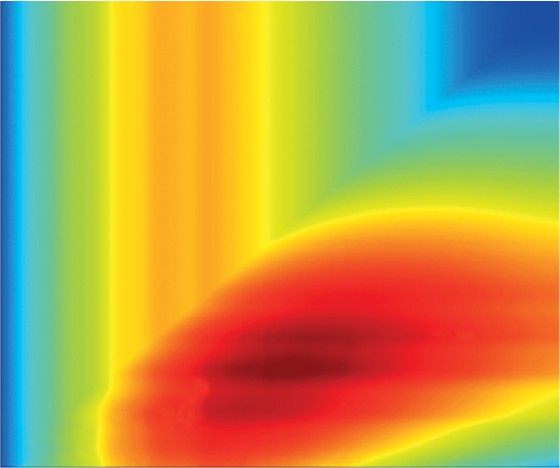
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**e 4T tandem**

3.0



2.5

Bottom cell bandgap (eV)

2.0

1.5

1.0

0.5

0.5 1.0 1.5 2.0 2.5 3.0

Top cell bandgap (eV)

PCE (%)

46.1



41.0

35.8

30.7

25.6

20.5

15.4

10.24

5.12

0

Figure 1 | **Operating principle and efficiency limits for tandem solar cells. a** | Tandem solar cell concept, where higher-energy photons are absorbed from the wider-bandgap semiconductor (in blue) and lower-energy photons can be

absorbed by the narrower-bandgap semiconductor (in red). **b** | Solar irradiance spectrum showing the spectral regions over which the two semiconductors could absorb (the colour scheme is the same as in part **a**). **c** | Diagram of the architecture of two-terminal (2T) and four-terminal (4T) tandem solar cells. Theoretical maximum power conversion efficiency (PCE) as a function of front (top) and rear sub-cell bandgap for 2T (part **d**) and 4T (part **e**) architectures, assuming no absorption losses. The dashed line traces the peak efficiency for the whole range of front-cell bandgaps, and the white circles indicate the maximum power conversion efficiencies. ITO, indium tin oxide. Part **c** is adapted with permission from REF. 23, AAAS.

the efficiency of the cell. In a 2T configuration, the sub-cells are electronically in series, so the voltages generated by each sub-cell are added. Kirchhoff ’s law dictates that the currents flowing through each sub-cell must match, meaning that the photocurrent is limited by the lower sub-cell current. Because the sub-cells must harvest different regions of the solar spectrum, achieving

photocurrent matching at the maximum power point condition of each sub-cell requires careful bandgap and efficiency engineering (FIG. 1d). In the 4T configuration, the sub-cells are effectively operated separately and can be connected in whichever way desired; therefore, in this case, current-matching limitations are avoided and less stringent bandgap optimizations are required (FIG. 1e).

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Box 1 | **Tunnel junctions, recombination layers and selective contacts**

Tunnel junctions and recombination layers are terms for layers with a number of designs used to electrically connect the sub-cells of monolithic multi-junction solar cells. They must provide a low-resistance and low-optical-loss connection between the p-type side of one cell and the n-type side of the other, allowing the hole current from one sub-cell to recombine with the electron current from the next. A tunnel junction is effectively a highly doped p-n diode, where the high doping provides a small depletion zone, allowing the easy tunnelling of carriers through the junction.

A recombination layer can also be made using a layer, such as metals or transparent conductive oxides, in which both electrons and holes have high conductivity and will recombine efficiently. This design works well in between a sandwich of effective hole-blocking and electron-blocking layers in order to prevent recombination of the wrong species in the layer.

Solar cells and the sub-cells of tandem solar cells require electrons to be extracted from one side of the junction and holes from the other side. In order to achieve this, it is necessary to ensure that only the correct species can be extracted from each side. This is normally achieved in one of two ways: by heavily doping each side of the photoactive semiconductor, as in conventional silicon photovoltaics, to form a p–(i)–n homojunction or by depositing additional layers in contact with each side of an intrinsic or lightly doped semiconductor, such as a perovskite. One of these materials will only transport holes, and the other only electrons, which forms a p–i–n heterojunction. The doped regions or selective layers are termed the selective contacts.

### Bandgap tunability of perovskites

Halide perovskites have attracted much attention due to the rapid increase in the power conversion efficiencies of single-junction solar cells utilizing these materials. A distinct advantage that makes perovskites highly relevant for multi-junction solar cells is the ease with which the bandgap can be tuned by compositional engineering of the constituent elements. The halide perovskites relevant to PVs exhibits a three-dimen- sional ABX3 structure, consisting of corner-sharing metal halide octahedra, as we illustrate in the unit cell shown in FIG. 2a. In particular, A is a small organic or inorganic cation with a +1 charge, normally Cs, formamidinium (FA) or methylammonium (MA), B a metal cation with a +2 charge (Sn, Pb or Ge) and X is a halide anion with a –1 charge (Cl, Br or I)9. Only ions of a particular size range can inhabit each site, depending on the other components of the lattice. The ability of a particular set of ions to form a stable perovskite struc- ture is predicted by the Goldschmidt tolerance factor, *t* (REF. 10). Outside the desired range of the tolerance factor, non-perovskite or lower-symmetry structures are formed, which generally have less desirable optical and electronic properties11.

The first perovskite to achieve notable PV efficien- cies was methylammonium lead triiodide (MAPbI3), which has a bandgap of ~1.6 eV (REFS 12–14). After the initial reports of this material, various groups showed that it was possible to dramatically change the bandgap of the material by changing the halide component15 and the A cation16. Moreover, it was realized that interme- diate compositions (with a mixture of different halides or cations) formed crystalline alloys with intermedi- ate bandgaps, which allowed for the fine tuning of the bandgap in the range 1.5−3 eV (REF. 17). Computational studies elucidated that the origins of the bandgap tuna- bility lie in changes in the density of states resulting from the effects of the inclusion of different cations and anions

on the X–Pb–X bond angle and length, and the differing chemical composition18.

The large bandgap range brought the perovskite mate- rials close to the optimum semiconductor bandgap for a single junction and, following these findings, device effi- ciencies rapidly increased19. Spurred on by the prospect of creating Pb-free perovskite solar cells, functioning solar cells based on Sn started to appear soon after the reports of the first perovskite solar cells20,21. Despite a promis- ing lower bandgap (~1.3 eV), Sn-based perovskites are extremely unstable to oxidation, and since their initial reports, there has been little progress, as their instability makes their processing very challenging.

Alloying Sn-based and Pb-based perovskites was shown to produce materials with lower bandgaps than neat Sn or Pb materials, reaching down to ~1.2 eV (REF. 22). This was the first hint that perovskites could achieve the low bandgap needed for the narrow-gap sub- cell in a tandem device, as we will discuss later. These alloyed materials appear to have unprecedented high stability, which is more characteristic of Pb-based than Sn-based perovskite compounds23. The bandgap does not scale linearly with changes in the lattice parameter, as is observed for semiconductors that obey Vegard’s law, and as seen when alloying the A and X sites only. The rationale behind the mechanism for tuning the band- gap is less obvious for these materials, and conflicting theories have been reported. It was initially thought that the competition between spin–orbit coupling and lattice distortion might play a role24, but more recently, it has been shown that strong bandgap bowing could arise as a result of certain arrangements or motifs of Sn and Pb in the lattice23. The origin of the enhanced stability of perovskites emerging from Pb–Sn alloying is currently unknown but is clearly a very important property that merits further investigation.

We summarize in FIG. 2b the range of bandgaps that are now currently attainable by substituting ions occu- pying various lattice sites in Sn or Pb halide perovskite compounds.

The low-bandgap cells that are attainable via Pb–Sn alloying in perovskites are well suited for use as the low-bandgap junction in a tandem cell. However, obtaining wide-bandgap solar-absorber materials that also deliver comparatively high *V*oc in solar cells is histor-

ically a greater challenge25. The high *V*oc values reported

for perovskite solar cells in 2012 (REF. 12), and the grow- ing awareness that halide substitution in perovskites would produce bandgaps higher than 1.6 eV, resulted in a growing interest in employing mixed-halide mate- rials as front cells in hybrid tandem cells, as described in detail later26. The ideal perovskite material for use in combination with Si would have a bandgap around 1.7 eV (REF. 7). However, it was also realized that mixed-halide perovskites reversibly undergo phase segregation under illumination, separating into Br-rich and I-rich regions27. These regions have different bandgaps than homogene- ous mixed-halide perovskites, meaning that the desired bandgap is effectively unstable, as shown by the photolu- minescence measured over time for a mixed-halide film under illumination. FIGURE 2c illustrates this change;

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3.2

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X-site substn

A-site substn

B-site substn

2.3



Cl

Br

I

Cs

MA Pb FA

Pb Sn

*y* 1–*y*

*y* = 0.87

*y* = 0.63 *y* = 0.38

Sn

*y* = 0.5

*y* = 0.25

1.8

1.6



A B X

Bandgap (eV)

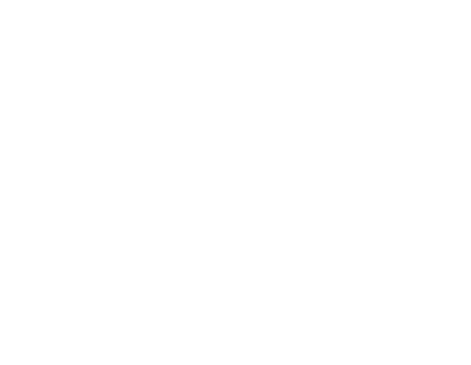
1.4

1.2

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### c d

700



CsPb(Br*x*I1–*x*)3

*x* = 0

*x* = 0.2

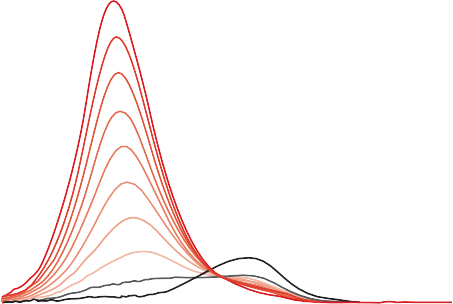
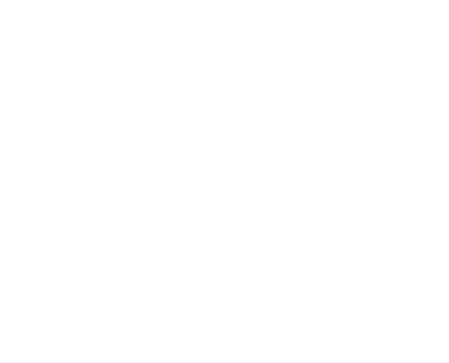
*x* = 0.33

*x* = 0.4

*x* = 0.8

*x* = 1

*x* = 0.6



Iodine-rich photoluminescence peak (grows under illumination)

Initial (unsegregated) photoluminescence peak

680

CsPbX3

APbI3 FABI3

Composition

6

PL (arb. units)

4

2

0

1.6 1.7 1.8 1.9 2.0

Energy (eV)

660

640

Wavelength (nm)

620

600

580

560

540

520

0 1 2 3 4 5 6 7 8 9

Time (min)

Figure 2 | **Perovskite structure, bandgaps and bandgap stability. a** | Cubic perovskite unit cell showing the ABX3 lattice structure, where A is a small organic or inorganic cation (+1 charge, such as Cs, formamidinium or methylammonium), B a metal cation (+2 charge, usually Sn, Pb or Ge) and X is a halide anion (–1 charge, such as Cl, Br or I). **b** | Plot showing all bandgaps currently attainable for the tin and lead perovskite halide system via substitution of the different lattice components. Bandgaps intermediate to those plotted can also be attained via intermediate compositions. Data were extracted with permission from REFS 15,16,23. **c** | Photoluminescence (PL) of methylammonium-based mixed-halide MAPb(Br0.4I0.6)3 perovskite over time measured every 5s under 457 nm, 15 mW cm−2 light at 300 K. The PL peak is observed to rapidly shift (within 1 minute) from ~1.85 eV to ~1.68 eV, representing a shift from an unsegregated to a segregated material, with iodide-rich domains and a lower bandgap27. **d** | PL peak position as a function of time for CsPb(Br*x*I1−*x*)3 materials under

~1 Sun illumination (100 mW cm−2), showing the variation in PL stability as a function of halide content29. Part **c** is adapted

with permission from REF. 27, CC-BY 3.0. Part **d** is adapted with permission from REF. 29, American Chemical Society.

the initial photoluminescence peak of a homogeneous MAPb(Br0.4I0.6)3 film at 1.85 eV decreases over time under illumination, and a peak at 1.68 eV appears, which rep- resents the photoluminescence of I-rich regions. The bandgap is effectively defined by the lowest-energy photoluminescence, so it shifts from 1.85 eV to 1.68 eV. Owing to phase segregation, carriers rapidly accumulate in the lowest-bandgap regions, losing energy and limiting the attainable *V*oc. If this voltage is limited, the maximum power conversion efficiencies established in FIG. 1d,e are no longer achievable, and the use of mixed-halide perovskites becomes non-ideal in tandem solar cells. Halide segregation can be minimized by replacing MA with alloys of Cs and FA, which facilitate the attain- ment of somewhat higher voltages28. Fully Cs-based perovskite films also exhibit a wider range of stable

bandgaps with respect to MA-based perovskite films29 (FIG. 2d). It has also been shown that the morphology can play an important role in the bandgap stability of mixed-halide films, with larger grains being more stable to phase segregation30. The exact role of these contribu- tions or others, however, have yet to be fully determined. Indeed, different compositions have been reported to be stable under illumination and dark31. Furthermore, even with the apparently more phase-stable materials, halide segregation still occurs in perovskites with cer- tain compositions and under certain processing condi- tions. Recent work has shed light on the origin of this effect, finding that halide segregation occurs via halide defects. This suggests that halide segregation could be minimized by reducing the defect density32. However, although large-bandgap (>1.65 eV) mixed halides

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have been used to realize comparatively high-voltage devices, they have thus far exhibited greater losses than their lower-bandgap analogues. Until equally low-loss, large-bandgap mixed-halide perovskite solar cells are experimentally realized, we will not know whether the problem of halide segregation has been truly solved.

### Hybrid tandem cells

Before the advent of small-bandgap perovskite mate- rials, perovskites were initially used in hybrid tandem devices (referring to the hybridization of two or more types of PV technologies) in which they constitute the higher-bandgap sub-cell, whereas a more established technology, such as Si or copper indium gallium (di) selenide (CIGS), constitutes the rear, lower-bandgap cell. Si and CIGS have similar bandgaps of around

* 1. eV. However, in addition to being grown under high- vacuum conditions, CIGS layerscanbeformedbysolution- processing methods, similar to those used for perovskites, substantially cutting production costs33. The attraction of the hybrid approach is that a perovskite layer could be easily and inexpensively incorporated in a tandem solar cell to increase the efficiency of scaled-up, commercially available solar panels. Such an approach provides a way for perovskite tandem solar cells to secure a foothold in the extremely competitive PV market and capitalize upon the already mature manufacturing processes of Si and CIGS. Initial efforts employed the more easily fabricated 4T architecture and the prototypical MAPbI3 perovskite

with a 1.6 eV bandgap, which is not an ideal bandgap

to use in combination with 1.1 eV absorbers. The first challenge to overcome was the development of semi- transparent electrodes for these 4T devices. Deposition of a transparent electrode on top of a perovskite cell is a non-trivial problem — sputter coating, the established way to deposit transparent conductive oxides (TCOs), damages the top-most organic charge-extraction layer34. Here, we do not assess in detail the different methods and opportunities for the development of novel semi-transparent electrodes for perovskite tandem solar cells but rather provide an overview. This aspect is covered in detail in a recent review by Huang and co-workers35. The challenge was first addressed by McGehee and co-workers, who developed a pressure-transfer approach (effectively laminating a pre-deposited nanowire film under pressure) to deposit a silver-nanowire-based elec- trode onto the perovskite cell. By combining the nano- wire-contacted MAPbI3 cell with either a Si or CIGS rear cell, efficiencies of 17% and 18.6%, respectively, were attained36. Importantly, the inclusion of the perovskite layer increased the efficiency of the single-junction rear cells to above their initial levels of 11.4% and 17%, respec- tively. However, the corrosion of Ag by hydrohalic acid effused from the perovskite and the diffusion of Ag from the nanowires into the perovskite limits the stability of these electrodes37. Other approaches to develop benign semi-transparent electrodes followed in quick succession. Kranz *et al.* and Werner *et al.* simultaneously developed methods for sputter coating ZnO:Al or indium zinc oxide TCOs on top of the perovskite cell. Sputter-coating damage was prevented by depositing a thin MoO*x* buffer

layer, resulting in 4T tandem efficiencies of 19.5% on CIGS and 18.2% on Si38–40. Concern over the long-term stability41 of this buffer layer motivated the development of a highly stable layer of solution-processed aluminium zinc oxide nanoparticles, which enabled the sputtering of indium tin oxide (ITO), and an 18.0% efficiency was achieved42. Taking a slightly different approach, Yang and co-workers developed a dielectric–metal–dielectric elec- trode composed of an ultrathin Au-seeded Ag layer sand- wiched between two thin MoO*x* layers, reaching tandem efficiencies of 15.5% with solution-processed CIGS43.

Rather than a material with a 1.6 eV bandgap, an absorber with a bandgap of ~1.75 eV is ideal to use as the front cell in combination with a 1.1 eV Si or CIGS rear cell in both the 2T and 4T configurations, as shown in REF. 5 and FIG. 1d,e. Halide segregation prevented MA-based mixed-halide cells from attaining com- petitive efficiencies, with voltages remaining limited to ~1.1 V, dictated by the I-rich phases. However, our group reported that the introduction of FA–Cs alloys as a replacement for MA could provide a potential effective solution to this issue. We were able to fabricate phase- stable FA0.83Cs0.17Pb(I0.6Br0.4)3 devices (bandgap of 1.7 eV) with a 1.2 V *V*oc. A sputter-coated layer of ITO comple- mented by a buffer layer of ITO nanoparticles was used as the semi-transparent electrode for the perovskite cell. By coupling these devices with a Si rear cell, efficiencies of 19.8% were demonstrated and a practical road map to surpassing 25% efficiency was laid out28.

Most approaches detailed thus far used a standard architecture for Si or CIGS rear cells with no special modifications for the tandem application. However, standard Si cells are not optimized for infrared absorp- tion. Aiming to address this, Chen *et al.* and Werner *et al.* made further advances by modifying the design of the rear silicon heterojunction (SHJ) cell to boost its near- infrared absorption. This was performed by making use of customized anti-reflection coatings and surface textur- ing, such as intentionally roughening the surface of the cell to enhance light absorption44,45. Combining this with advances in perovskite-cell processing, an efficiency of 25.2% was attained for small-area (0.25 cm2) hybrid 4T tandem cells and 23.0% for 1 cm2 cells45. Most recently, the inclusion of Rb cations in the perovskite was shown to produce cells with a stable bandgap of 1.73 eV, with no obvious halide segregation. Combining these cells with interdigitated back-contact silicon cells gave an impressive efficiency of 26.4% for a small-area (0.16 cm2) 4T tandem cell46. FIGURE 3a,b shows the external quan- tum efficiency and the current–voltage characteristics resulting in these efficiencies.

The more ideal 2T hybrid tandem cells were initially much less effective than the 4T devices owing to the increased difficulty of fabrication. The first reported instance of perovskites being monolithically grown on another junction was with kesterite (a general term for copper zinc tin sulfides, selenides and alloys) rear cells; sub-optimal contacts (aluminium, with only 50% trans- mission) and the kesterite bandgap limited the efficiency to 4.7%47. This soon improved to 10.9% by using CIGS, a more transparent Ca contact and a wider-bandgap

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# REVIEWS

**a** 100

80

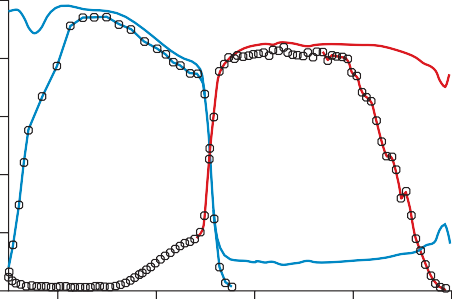
60

EQE (%)

40

20

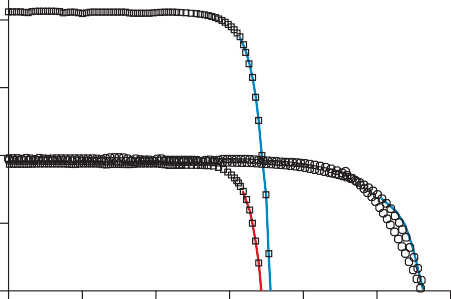
0



400 600 800 1,000 1,200

Wavelength (nm)

### b

40

Current density (mA cm –2)

30

20

10

0

0.0 0.2 0.4 0.6 0.8 1.0 1.2

Voltage (V)

EQE silicon — under perovskite Perovskite transmittance

EQE perovskite Perovskite absorption

Silicon — original

Silicon — under perovskite Perovskite — reverse Perovskite — forward Perovskite — steady state

**c** 100

90

80

70

60

EQE (%)

50

40

30

20

10

0

Blue parasitic loss

* 1. mA cm–2

Perovskite

18.9 mA cm–2

Reﬂection loss

4.8 mA cm–2

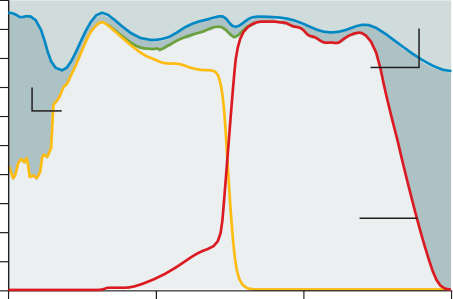
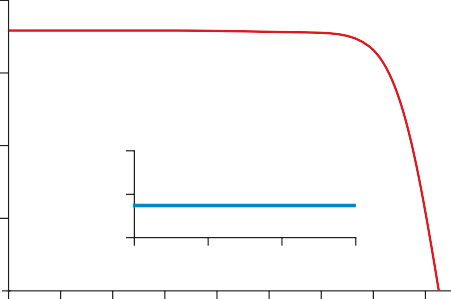
NIR parasitic loss

3.3 mA cm–2

Silicon

18.5 mA cm–2

**d** 20

15

Current density (mA cm –2)

10

5

0

30

25

Eﬃciency (%)

20

0 10 20 30

Time (min)

300 600 900 1,200

Wavelength (nm)

0.0 0.2 0.4 0.6 0.8 1.0 1.2 1.4 1.6

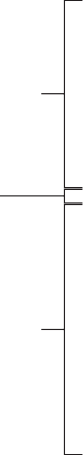
Voltage (V)

## 

Perovskite absorption Silicon cell

Perovskite cell

Perovskite and silicon EQE

**e** Ag

ITO PC60BM

Front sub-cell

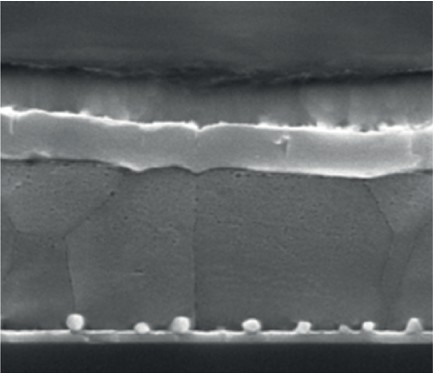
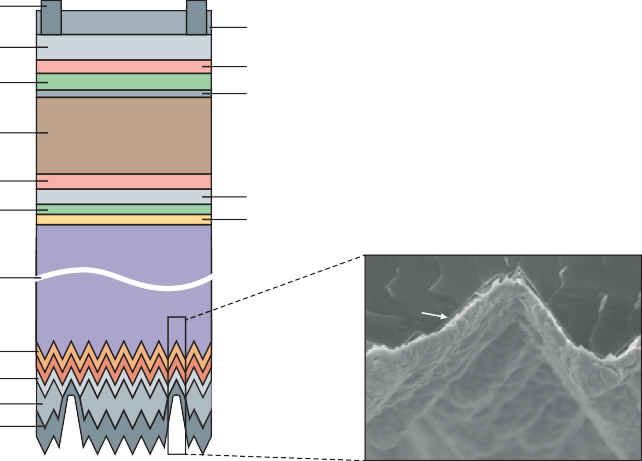
(wide bandgap)

Perovskite

Recombination layer

**f**

LiF SnO2/ZTO



500 nm

LiF ITO

Perovskite NiO

LiF

NiO

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s r e h s

i l b

u P

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l l i

m

c a M

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+ ITO

a-Si:H (n )

a-Si:H (i)

### g

(narrow bandage)

c-Si (n)

Si

ITO

500 nm

Rear sub-cell

a-Si:H (i) a-Si:H (P+)

Ag

ITO

SiNP

Ag

Figure 3 | **Hybrid tandems.** External quantum efficiency (EQE) and absorption (part **a**) and current–voltage characteristics (part **b**) measured under AM 1.5 and 100 mW cm−2 illumination for the best four-terminal hybrid tandem attained so far46. As shown in part **b**, ‘forward’ and ‘reverse’ indicate the direction of the voltage scan (forward=0–1.2 V, and vice versa for reverse). EQE and absorbance (part **c**) and current–voltage characteristics (part **d**) measured under AM 1.5 100 mW cm−2 illumination for the best two-terminal hybrid tandem attained so far53. Inset: efficiency as a function of time, showing the steady-state power output when held at the maximum power point. Light that is absorbed and then successfully converted to current is represented by the area under the EQE plot; the shaded area shows the light that is absorbed but not converted to current (part **c**). Schematic of the architecture for the best two-terminal hybrid tandem cell is shown in part **e**. Parts **f** and **g** show scanning electron microscopy images of different sections of this device. Thin layers are not obvious in the scanning electron microscopy images and are thus not labelled. ITO, indium tin oxide; NIR, near infrared; ZTO, zinc tin oxide. Parts **a** and **b** are adapted with permission from REF. 46, Wiley. Parts **c**, **d**, **e, f** and **g** are adapted from REF. 53, Macmillan Publishers Limited.



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perovskite cell47,48. Building 2T tandem cells on more stable Si rear cells allowed a wider thermal processing window. Thus, by using high-temperature-processed protective oxide-layers — as was done in the most effi- cient perovskite cells at the time — and by employing the same Ag-nanowire electrode used in 4T tandem cells, Mailoa *et al.* reported a 13.7% efficiency for a monolithic perovskite-on-silicon tandem cell49. The same advances reported for 4T tandem cells that allowed the sputtering of TCOs were used to good effect in advancing the 2T tandem system, along with the advent of efficient planar perovskite solar cells, which do not contain any compo- nents requiring high-temperature thermal processing50. An 18.1% efficiency was attained with a MoO*x* buffer layer and the low-temperature atomic-layer deposition of SnO*x*, on top of the Si cell, as an electron-selective contact (BOX 1), minimizing thermal damage to the Si cell51. A similar low-temperature route, which uses the same buffer layer but a [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) electron-selective contact, resulted in 21.2% efficiency52. Most recently, McGehee and co-workers employed a more stable and better bandgap-matched FA–Cs perovskite. In this case, it was possible to deposit a SnO*x* buffer layer on top of the perovskite cell by atomic layer deposition, which ena- bled the sputter deposition of ITO as a top electrode53 (FIG. 3e,f,g). An impressive certified efficiency of 23.6% was observed by depositing this perovskite cell on top of an infrared-tuned SHJ cell. The external quantum effi- ciency (EQE) and current–voltage characteristics defin- ing this PCE are shown in FIG. 3c,d, respectively. Notably, this 2T device has a larger area (1 cm2) than most lab- oratory-scale devices (typically ~0.1 cm2), which bodes well for the scale-up of these cells, and is it more efficient than the best 1 cm2 4T devices produced so far?

In addition to the combination of perovskites with established technologies, a hybrid tandem cell based on another emergent technology has seen some research effort, driven by the possibility for low-cost, low-temper- ature, fully solution-processed tandem cells. By combin- ing an infrared-absorbing organic bulk heterojunction solar cell, a technology that has been posited as having the potential to become an inexpensive solution-pro- cessable technology, with a perovskite cell in a mono- lithic structure, Yang and co-workers were able to obtain an efficiency of 10.2%. This was then pushed further by Russel and co-workers, demonstrating a 16.0% efficiency by employing a graded recombination layer54,55.

### All-perovskite tandem cells

The idea of all-perovskite tandem cells is attractive, as these devices would retain all the cost and processing advantages of low-temperature-processed PV thin films,

which are lost to a certain extent in hybrid tandem archi-

* materials with bandgaps far from the ideal range (1.6–2.2 eV) — and reported a 10.8% efficiency56. Processing two perovskite cells directly on top of each other (referred to as monolithically) remains a challenge. The strongly coordinating polar solvents used in the monolithic construction easily dissolve the underlying perovskite absorber layer during the deposition of the subsequent cell. Zhou and co-workers first addressed this challenge by developing a robust recombination layer composed of several organic layers. They processed two

1.6 eV sub-cells on top of each other to demonstrate good voltage addition. However, only a 7.0% overall power conversion efficiency was reached as a result of the low conductivity of the recombination layer and unsuitable bandgaps for current matching57.

A key breakthrough in all-perovskite tandem cells came from the effective use of low-bandgap perovskites. Low-bandgap perovskites, resulting from the strong bandgap bowing in Sn–Pb alloys, suitable for use in low- gap sub-cells (<1.5 eV) were already known. However, poor efficiencies were reported (7%), and low stability was expected owing to the presence of air-sensitive Sn2+ (REF. 22). The formation of smooth, high-quality films, which are necessary for devices, is challenging owing to the rapid crystallization of Sn-based compounds. However, several groups have recently reported advanced processing methods based on the use of precursor phases containing dimethyl sulfoxide (DMSO) and anti-solvent treatments that have enabled the fabrication of high- quality Sn–Pb perovskite layers23,58,59. These perovskites are characterized by 1.2–1.3 eV bandgaps and have shown much higher efficiencies of (~15%). Moreover, the stability of these Sn–Pb alloys was unprecedentedly high — more similar to Pb-based than Sn-based materials

* making them potentially useful in real devices23. This finding opened the door to the possibility of using low- gap perovskites in all-perovskite tandem cells with opti- mum bandgap matching as a replacement for Si or CIGS, and combining them with a wider-gap perovskite sub-cell. 4T all-perovskite tandem cells became relatively easy to fabricate once efficient low-gap perovskites were obtained. By combining a 1.3 eV MA0.5FA0.5Pb0.75Sn0.25I3

perovskite with a 1.6 eV MAPbI3 and applying a sput-

tered ITO top contact, Jen and co-workers achieved 19.1% efficiency58. As observed in FIG. 1e, this is not an ideal bandgap combination; our group, in collabo- ration with the McGehee group, and co-workers, went one step further, developing a FA0.75Cs0.25Pb0.5Sn0.5I3

perovskite with a more ideal 1.2 eV bandgap to fabricate

a 4T cell with 20.3% efficiency23. Very recently, Yan and co-workers pushed the efficiency of Sn–Pb alloys to 17% and attained a 4T cell with 21.0% efficiency, showing the potential of all-perovskite tandem cells to compete with established technologies60.

External quantum efficiency (EQE.) Number of carriers collected relative to the number of photons incident on the cell. The EQE of a cell defines the *J*sc by integrating the product of the EQE and the AM 1.5G solar spectrum.

tectures. However, the lack of low-bandgap perovskites and the practical difficulties of solution processing mul- tiple layers on top of each other have kept all-perovskite tandem cells from being anything other than a novelty until recently. Heo *et al.* used a post-deposition lamina- tion process to fabricate a prototype 2T all-perovskite tandem cell based on MA lead tribromide and triiodide

2T all-perovskite tandem cells represent a tougher challenge because of the more stringent bandgap match- ing and processing complications, as already discussed. Breakthroughs in low-gap perovskites have enabled ideal bandgap combinations to be reached; a 1.2 eV low- gap sub-cell is best combined with a 1.8 eV wide-gap sub-cell, which are now feasible to achieve with the

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perovskite system. Developing robust interlayers, allowing perovskite-on-perovskite deposition with low- loss electrical properties, was the final piece of the puzzle. This interlayer was realized by our group, in collaboration with the McGehee group, and co-workers, by combining tin oxides and sputter-coated ITO using the atomic layer deposition technique23. The secondcell was thenprocessed directly on top of the sputter-coated ITO, which protected the cell underneath and had excellent electrical and opti- cal properties. By processing a 1.2eV FA0.75Cs0.25Pb0.5Sn0.5I3 perovskite sub-cell ontopof a 1.8eV FA0.83Cs0.17Pb(I0.5Br0.5)3 sub-cell, it was possible to assemble 2T tandem cells with efficiencies of 17.0% and a high voltage of 1.65 V. FIGURE 4a shows a cross-sectional scanning electron microscope image of the resultant device, and FIG. 4b,c reports its EQE and current–voltage characteristics23.

A way to circumvent the requirement for a complex interlayer that is compatible with solution processing is to thermally deposit all the layers that comprise the entirety of the second processed cell onto the completed underlying cell, including the selective contacts and the perovskite itself. This has recently been enabled by developments in vacuum-processable doped organic semiconductors, which can be used as selective con- tacts and recombination layers. Bolink and co-workers demonstrated that efficiencies of 18% could be attained if a 1.6 eV MAPbI3 perovskite sub-cell is evaporated on top of a solution-processed 2 eV Cs0.15FA0.85Pb(I0.3Br0.7)3 cell and the two cells are connected through doped organic semiconductors, as illustrated in REF. 61 and FIG. 4d. The EQE and current–voltage characteristics of representa- tive 2T tandem cells fabricated in this way are shown in FIG. 4e,f. The 2 and 1.6 eV bandgap combination is not ideal owing to present limitations in the perovskite com- positions that can be thermally evaporated, leading to an overall tandem efficiency lower than that of the equiv-

alent 1.6 eV cell on its own. However, as developments

Calculation of the Shockley–Queisser limit5 for com- binations of absorber layers with different bandgaps provides the maximum limiting efficiencies of the PV device. However, we know that experimentally not every incident photon with energy larger than the bandgap will be converted into charge. The most significant losses in efficiency arise from optical inefficiencies, including reflective losses, interference effects, lack of absorption by the perovskite layer due to thickness limitations and ‘para- sitic’ absorption in the non-semiconducting layers, which do not contribute to photocurrent. These can be evalu- ated by determining the optical constants of perovskites and the other involved materials62–64. Therefore, optical models were developed to calculate the absorbed electric field within the active layers. Slightly more sophisticated calculations were carried out by making use of Lambertian models that showed how simple light-trapping mecha- nisms, such as Bragg mirrors or scattering back reflectors, could enhance the perovskite-on-silicon tandem device efficiencies beyond 35%65,66.

In order to achieve a more precise calculation of absorbed, reflected and transmitted light within the solar cell layers, the electric-field distribution within the materials must be calculated. This can be done by numeri- cally solving Maxwell’s equations along all the dimensions of the system by making use of finite-element or finite- difference time-domain analyses67,68. Such simulations are generally computationally expensive; therefore, more efficient methods can be applied by reducing the space dimensionality from 3D to 2D or 1D so that the com- monly known transfer matrix method (TMM)69,70 can be used. This method enables the prediction of electro- magnetic wave propagation (and therefore the power of light) through the layers by introducing interface matri- ces, which include Fresnel coefficients, and layer dissipation matrices, which include Beer–Lambert absorption.

Early predictions of the material composition and

Lambertian models

Describe light trapping within a solar cell resulting from light that is reflected through a Lambertian reflector, which has an isotropic radiance; the luminous intensity is proportional to the cosine of the angle between incident light and normal.

Fresnel coefficients Coefficients resulting from Fresnel’s equations, which define the transmission and reflectance of an electric field at an interface between two homogeneous media as a function of angle of incidence.

Fill factor

Ratio of the power produced at the maximum power point voltage to the product of *J*sc and *V*oc. The maximum power point voltage is that at which voltage × current, that is, power, is maximum and the PCE is defined.

are made to enable the thermal deposition of multi-com- ponent alloyed compositions, this approach will become very promising.

A chronological summary of the major developments in the evolution of perovskite tandem cells is shown in FIG. 5; a plot of the progression of the reported maximum efficiencies for the various technologies and a summary of the current record efficiencies are reported in FIG. 6 and TABLE 1, respectively.

**Theoretical approaches to tandem solar cells** Early on during the development of perovskite tandem solar cells, computational studies based on optical models started to emerge. These efforts are still motivated by the desire to predict the achievable power conversion efficien- cies before putting effort into the complex fabrication of multi-junction devices. Furthermore, 2T tandem devices require current-matching conditions to convert the max- imum amount of energy and therefore predictions of the optimized thicknesses of the layers can lead to substantial improvements. Tandem stacks involve a number of thin layers that introduce complex interference effects, which can also diminish photon absorption in the active layers, unless the thicknesses and materials are carefully selected.

optimum layer thickness for perovskite-on-CIGS devices were carried out by Chen *et al.* using calculations based on TMM71. Similarly, Filipic *et al.*72 performed extensive optical simulations of 2T and 4T perovskite-on-silicon heterojunction tandem devices. Following optimization of the layer thicknesses, they calculated the resulting short- circuit currents and combined these two values with the state-of-the-art *V*oc values and fill factors. By doing so, they determined more accurate values of the optical losses and were therefore able to propose more realistically achieva- ble efficiencies above 30%. The order of different layers in the solar cell stack together with the selection of optically ideal materials as hole-transporting layers (HTLs) and electron-transporting layers (ETLs) — that is, materials with minimal parasitic absorption — also play a role in determining the device efficiency. Recent studies73 suggest that if layers are arranged so that light passes through the ETL first, parasitic and reflective absorption losses can be reduced. This is due to better refractive indices, which match those of TCOs. The improved photocurrent sup- ports the matching condition and leads to a substantially higher overall power conversion efficiency (PCE) of 31%. Similarly, Asadpour *et al.*74 analysed the beneficial effect of a bifacial 2T perovskite-on-SHJ tandem cell through an

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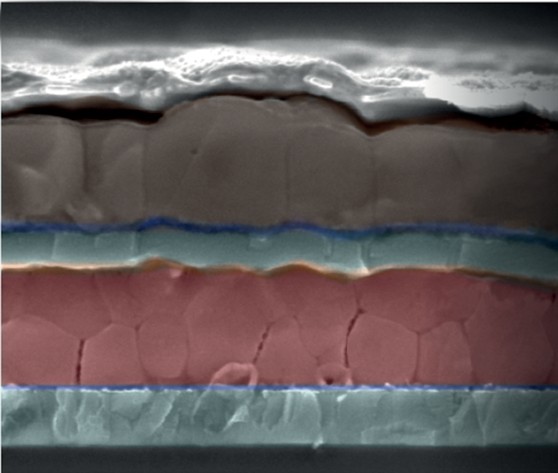
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# REVIEWS

### a

Ag C60/BCP



500 nm

**d**

Rear sub-cell (narrow gap)

Au

TaTm:F6-TCNNQ

TaTm

MAPbI3

1.2 eV perovskite

PEDOT:PSS ITO SnO2/PCBM

C60

Recombination C :Phlm

layer T 60 -TCNNQ

aTm:F6

TaTm

1.8 eV perovskite NiO

ITO

Front sub-cell (wide gap)

Cs0.15FA0.85Pb(I0.3Br0.7)3

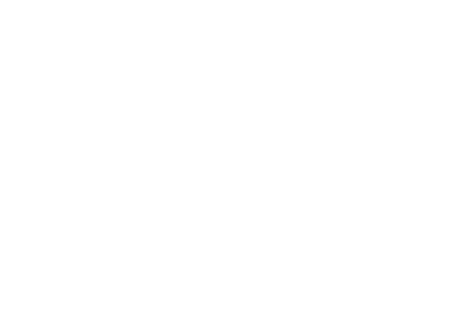
IPH TiO2 ITO

### b

100

**e**

100



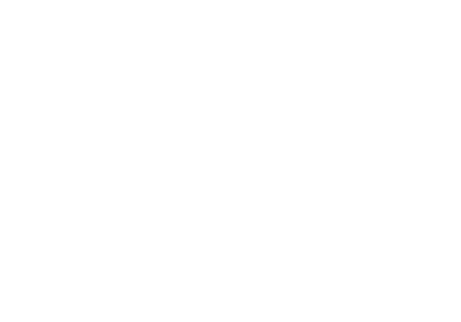
Top and bottom EQE

Top cell

Bottom cell

9.13 mA cm–2

9.29 mA cm–2



1.8 eV sub-cell

1.2 eV sub-cell

14.1 mA cm–2

15.8 mA cm–2

80 80

60 60

EQE (%)

EQE (%)

40 40

20 20

0

400

500 600 700 800 900 1,000

Wavelength (nm)

0

400

500 600 700 800

Wavelength (nm)

### c

5

0

Current density (mA cm–2)

–5

–10

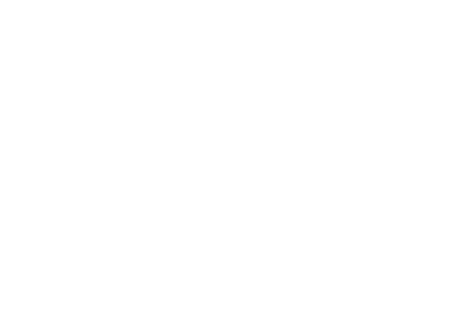
–15

–20

–25

–30

0.0 0.5 1.0 1.5



SC

FF

*V*OC (V)

PCE (%)

FWD REV

*J* (mA cm–2) 9.6 9.7

72.2 75.6

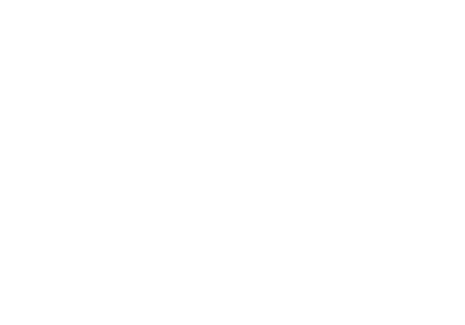
2.132 2.137

14.8 15.6

FWD REV

**f**

10



SC

FF

*V*OC (V)

PCE (%)

*J* (mA cm–2) 14.5

0.70

1.66

16.9

1.2 eV cell

1.8 eV cell

2T tandem

5

Current density (mA cm–2)

0

–5

–10

0.0

0.4

0.8

1.2 1.6 2.0 2.4

Voltage (V)

Voltage (V)

Figure 4 | **All-perovskite tandem cells with solution-processed and evaporated recombination layers. a** | Scanning electron microscopy image of a solution-processed 1.2 eV and 1.8 eV all-perovskite tandem cell.

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**b** | External quantum efficiency (EQE) of the two sub-cells of the solution-processed all-perovskite tandem cell with currents extracted by integrating the product of EQE and the AM 1.5 spectrum detailed in the plot. **c** | Current–voltage characteristics under AM 1.5G illumination for the sub-cells and full cell of the solution-processed all-perovskite tandem cell. The inset details the photovoltaic parameters for the two-terminal (2T) tandem (green curve). **d** | Device stack for part-solution, part-evaporated 1.6 eV and 2.0 eV all-perovskite tandem cell containing doped organic interlayers. The Cs0.15FA0.85Pb(I0.3Br0.7)3 cell (2.0 eV) is solution processed, and the methylammonium lead triiodide (MAPbI3,1.6 eV) is evaporated, as are all other layers. **e** | EQE for the two sub-cells of the part-solution, part-evaporated all-perovskite tandem cell. **f** | Current–voltage characteristics under AM 1.5G illumination for the part-solution, part-evaporated

all-perovskite tandem cell. FWD and REV refer to the forward (voltage from 0 to 2.4 V) and reverse (voltage from 2.4 V to 0 V) scans of the applied voltage. BCP, bathocuproine; FF, fill factor; IPH, indene-C60-propionic acid hexyl ester; ITO, indium tin oxide; *J*sc, short-circuit current; PCE, power conversion efficiency; PEDOT:PSS, poly(3,4-ethylenedioxythiophene): polystyrene sulfonate; Phlm, N1,N4-bis(tri-*p*-tolylphosphoranylidene) benzene-1,4-diamine; TaTm, N4,N4,N4ʹʹ,N4ʹʹ-tetra([1,1ʹ- biphenyl]-4-yl)-[1,1ʹ:4ʹ,1ʹʹ-terphenyl]-4,4ʹʹ-diamine; F6-TCNNQ, 2,2ʹ-(perﬂuoronaphthalene-2,6-diylidene) dimalononitrile; *V*oc, open-circuit voltage. Parts **a**, **b** and **c** are adapted with permission from REF. 23, AAAS. Parts **d**, **e** and **f** are adapted with permission from REF. 61, Wiley.

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# REVIEWS

**2009**

**2012**

**2013**

**2014**

**2015**

(Aug 2016) Advances in processing enable low-bandgap (1.2–1.3 eV) perovskite cells with ~15% eﬃciency. First four-terminal all-perovskite tandems demonstrated using advances in low-gap perovskites23

**2016**

**2017**

Perovskite research

Electrode/interlayer research

All-perovskite tandem cells

Hybrid tandem cells

(Apr 2009) First use of halide perovskite as a sensitizer in a solar cell; very unstable, 3.8% PCE (REF. 14)

(Oct 2012) More stable perovskite devices with

~10% eﬃciency achieved using solid-state

hole-transport materials. Unexpectedly low loss

in potential from bandgap to *V*oc makes them

attractive for tandem cells12,13

(Mar 2013) Bandgap tuning by halide substitution opens the range to 1.6 – 2.3 V (REF. 16)

(Feb 2017) Extremely stable two-terminal perovskite–Si tandem cell with 23.6% eﬃciency demonstrated via ﬁne-tuning of Si and perovskite cells53

(Jan 2014) Bandgap tuning by A-site cation substitution to narrow the bandgap to 1.48 eV (REF. 16)

(May 2014) Tin–lead alloying reported to show anomalously low-bandgap perovskites, extending the accessible range down to

1.2 eV, but the cells were ineﬃcient22

(Oct 2014) First two-terminal perovskite–kesterite

hybrid tandem cell fabricated (4.7% PCE)47

(Jun 2015) Methods to allow

sputter coating of transparent electrodes developed and used eﬀectively in

four-terminal tandems cells38,39

(Mar 2015) First two-terminal perovskite–Si

hybrid tandem cell fabricated using

silver-nanowire electrode49

(May 2014) Functioning (6%) tin-based

perovskite solar cells reported20,21

(Dec 2015) Formamidinium-

cesium hybridization aﬀords a stable,

ideal-bandgap perovskite top cell for 19.8% eﬃciency in four-terminal tandem cells28

(Dec 2015) First monolithic two-terminal all-perovskite tandem cell fabricated using organic interlayers (7% PCE)57

(Oct 2016) Monolithic two-terminal all-perovskite tandem cells with ideal bandgaps (1.2 and 1.8 eV) fabricated, leveraging protective ITO interlayer and eﬃcient low-gap perovskites23

(Dec 2014) Silver-nanowire electrode enables the ﬁrst four-terminal perovskite–Si hybrid tandem cell (18.6% PCE)36

Figure 5 | **Timeline showing the major developments in the evolution of perovskite tandem solar cells.** ITO, indium tin oxide; PCE, power conversion efficiency; *V*oc, open-circuit voltage.

optical TMM and a drift-diffusion-based device simula- tion. The resulting enhancement in the absorption in the perovskite film improves the tandem cell efficiency from 25% to 33%, as light reflected from Earth’s surface also boosts light absorption through the transparent rear side. The light-management behaviour of typically struc- tured silicon cells conformally coated with a perovskite front cell was simulated by a ray-tracing technique in combination with TMM by Schneider *et al.*75 and by a finite-difference time-domain analysis by Foster and John76. Both studies report that a conformal coating of thin silicon layers presenting an inverted-pyramid structure in combination with an anti-reflective coating can lead to near perfect absorption and PCEs well above 30%. Various light-trapping mechanisms from flat lay- ers to complex geometrical structures were compared by Santbergen *et al.*77, who came to a similar conclusion that light management has an important role in photocurrent enhancement. Entire module-performance simulations that take into account the losses from 4T cell intercon- nections were carried out through a combination of

optical and electronic modelling by Zhang *et al.*78 Their results indicate the crucial effect of the combination of a carefully selected TCO and an optimized layer thickness, which can lead to mini-modules (which have a larger area than a standard test cell but smaller than a full module) with a realistic efficiency close to 20%.

From FIG. 1d, we find that the optimal bandgap of a front cell to combine with silicon should be slightly wider than the 1.6 eV bandgap of the commonly used MAPbI3. By theoretically shifting the optical con-

stants for MAPbI3 horizontally along the energy axis,

Albrecht *et al.*79 showed that the ideal bandgap for the perovskite absorber on top of a SHJ cell is 1.73 eV. This leads to a matched current condition at ~17.5 mA cm−2 and hence a monolithic tandem cell efficiency of 28.4%. This model assumes fixed fill factors and *V*oc scaling

with higher bandgaps. Notably, in all these optical mod-

els, boundary conditions have to be implemented, such as the minimum and maximum thickness for the layers involved. The experimentally measured and certified cells of McGehee and co-workers delivered a short-circuit

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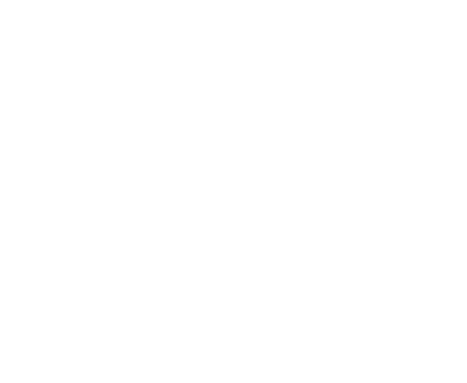
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# REVIEWS

Detailed balance theory Originally proposed by Shockley and Queisser, it allows the calculation of the thermodynamic efficiency limit of solar cells by taking into account the balance between absorbed and emitted photon flux.

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20



Two-terminal hybrid

Four-terminal hybrid

Two-terminal all-perovskite

Four-terminal all-perovskite

15

PCE (%)

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Year

Figure 6 | **Perovskite tandem cell efficiencies.** Plot of maximum reported power conversion efficiency (PCE) of each perovskite tandem technology over time.

current of over 18 mA cm−2, largely due to the excellent red response of the silicon cell and the low parasitic absorbance of the employed thin fullerene ETL53. This experimental result outperforms the theoretical predic- tions and indicates that once the most suitable perovskite junctions are integrated into hybrid tandem devices, power conversion efficiencies around 30% could be realistically achieved.

Anaya *et al.*80 studied the influence of the bandgap variation of Sn–Pb-based perovskite rear-cell absorbers in all-perovskite tandem cells. They obtained the opti- cal constants for perovskite films with different band- gaps by measuring the reflectance and transmission properties of the films and fitting them to the Forouhi– Bloomer model. The authors then used a TMM model for stacked layers and claimed that a matched photocur- rent of 17.2 mA cm−2 and combined PCE of 34% could potentially be achieved, assuming the state-of-the-art Voc

values and a slightly ambitious fill factor of 0.88.

An important aspect to consider when designing tandem solar cells is the energy output under real con- ditions. Conventional experimental tests make use of a light source filtered to result in the standardized solar illumination having passed through the equivalent of

1.5 Earth atmospheres (air mass (AM) 1.5) with a total equivalent solar irradiance of 100 mW cm−2 and per- pendicular incidence. The experimental and theoretical results shown above demonstrate that perovskite-based tandem solar cells can lead to remarkable efficiency improvements under standardized conditions over that of single-junction devices. However, because the real- world spectrum varies from AM 1.5 and the angle of solar incidence changes, the annual energy yield could be significantly affected owing to the discrete spectral responses and optical properties of the absorbing lay- ers. Multi-junction cells are expected to be more prone to losing efficiency owing to variations in spectral irradiance. Specifically, to maximize the PCE, mono- lithic tandem architectures require equally generated photocurrent within each absorber layer. Although the cells can be optimized to meet this condition under

standardized testing conditions, actual weather condi- tions and locations alter the spectral attenuation of sun- light as it passes through the atmosphere, leading to a reduced energy yield.

Models for the calculation of energy yields have been implemented with various levels of complexity for numerous PV materials and tandem devices81–83. A typical approach for analysing the impact of vary- ing solar spectra in different locations is by calculating their average photon energy (APE)84,85. By combining the APE of the changing spectra in the Netherlands and Colorado with a combined detailed balance theory (akin to the Shockley–Queisser theory), Futscher and Ehrler86 determined that the performance of 4T and even 2T perovskite-on-silicon tandem architectures are not sig- nificantly more affected by spectral and temperature variations than single-junction silicon cells. We came to a similar conclusion after implementing a model with rigorous optical and electronic considerations for the calculation of energy yields of precise perovskite-on- silicon tandem solar cell stacks. The model takes into account the measured irradiance spectra of three dif- ferent US locations with their anisotropic diffuse radi- ance spectra in combination with a TMM for the optical model and a one-diode equivalent circuit for the device model87. This approach enabled them to compare the realistic energy yield of single-junction silicon hetero- junction cells with 2T and 4T perovskite-on-silicon tandem cells mounted on different sun-tracking systems (see FIG. 7a). To evaluate whether an enhanced PCE under AM 1.5 conditions would also lead to an equal improvement in energy yield, they introduced an efficiency ‘derating’ factor. This factor compares the ratio of annual energy yield to the AM 1.5 PCE of the tandem device against a single-junction SHJ device (a value of 1 indicates that the difference in actual yield compared with the AM 1.5 yield is the same as for a silicon single junction). The authors show that this factor is >1 for 4T devices and ranges from 0.99 to 1.02 for 2T devices. This indicates that neither tandem device configuration significantly suffers under outdoor conditions and that PCE improvements could directly lead to improvements in the energy yield (see FIG. 7b). The undesirable cur- rent mismatch of tandem junctions can, however, result in an enhanced fill factor, which can almost entirely compensate for the current loss.

A slightly worse outcome for monolithic perovskite- on-silicon tandem devices was found by Duck *et al.*88, who implemented a similar model and found that com- pared with 4T devices, a relative energy yield loss of 4.5% is to be expected when 2T devices were implemented in Newcastle, Australia. Their model focused heavily on correctly accounting for temperature variation, which was found to be responsible for <1% of the total variation in the energy yield.

Accordingly, Futscher and Ehrler89 recently extended their initial simplified energy-yield model with an addi- tional one-diode equivalent device model. Their cal- culations were carried out by keeping the perovskite bandgap at the non-ideal level of 1.49 eV and by choosing conservative loss parameters for the perovskite sub-cell,

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# REVIEWS

which led them to the conclusion that the efficiency of sucha tandemcell configuration wouldonly be marginally improved over that of a single-junction silicon cell.

### Outlook and challenges

With the successful realization of tandem solar cells, the question emerges whether we should go one step further and add an additional absorber to form a triple-junction device. For other PV technologies, the efficiency gain of such an architecture has been proved to outweigh the disadvantages of additional fabrication efforts, although, notably, these already commercial technologies are not solution processed8. In FIG. 8a,b, we show the theo- retical efficiency potential of triple-junction solar cells, evaluated by plotting the detailed balance limit in two cases, using a 1.22 eV perovskite (FAPb0.5Sn0.5I3)-based absorber and a 1.1 eV silicon rear absorber, and vary- ing the middle-cell and front-cell bandgaps. The all- perovskite triple-junction can lead to efficiencies of 46.9% when absorber layers with bandgaps of ~2.1 eV,

~1.6 eV and 1.22 eV are combined, which is only slightly higher than the Shockley–Queisser limit of all- perovskite double-junction 2T tandem cells (46.0%). The lower bandgap of silicon accounts for an additional PCE gain to reach 49.4% when combined with bandgaps of ~1.5 eV and ~2.0 eV, which is a considerable boost. As discussed above, the real PCE is generally lower than the Shockley–Queisser limit. Therefore, Hörantner *et al.*90 used the transfer matrix and device modelling approach to simulate a more realistic expectation for the PCE. The bandgaps of the perovskite layers were shifted, and the thicknesses were optimized to reach the highest achiev- able efficiency. We show the results for double-junction and triple-junction perovskite solar cells in FIG. 8c,d,e. The extra processing steps necessary for the fabrication of all-perovskite triple-junction cells results in a rela- tively small efficiency gain compared with all-perovskite double-junction cells (from 32.2 to 33.0%), making the idea of all-perovskite triple-junction devices unlikely to be worthwhile. However, a solar cell composed of a perovskite double junction on top of an SHJ could achieve an efficiency of 35.3% (compared with 31.8% for the perovskite–Si tandem cell), which is more likely to be attractive for implementation. It is notable, therefore,

that in order to make triple-junction all-perovskite cells feasible, we need to discover how to further lower the bandgap of the perovskite absorber towards 1.1 eV. We show the calculated EQEs and current–voltage curves of the sub-cells in an optimized perovskite-on- perovskite-on-silicon device in FIG. 8f,g. These esti- mates lead to a promising outlook for triple-junction perovskite-on-perovskite-on-silicon devices.

There are several outlying challenges to be solved before the full potential of perovskite-based tandem solar cells can be exploited. Although impressive for early-stage development, the efficiencies are still far from the theoretical maximum (42%) in both hybrid and all-perovskite tandem cells.

Losses in perovskite tandem cells fall into two cate- gories: optical losses and electronic losses. Optical losses include reflective losses from the top surface, parasitic absorption at the semi-transparent contacts and inter- layers (any layer that is not one of the two semiconduct- ing absorber materials, where absorbed photons are not converted into current) and incomplete absorption in the sub-cells. Optical interference represents an issue that is more important in tandem devices than in single-junc- tion devices because of the greater number of smooth layers with thicknesses often close to the relevant photon wavelengths. Coherent interference results from reflec- tions off the multiple smooth internal surfaces, and hence, the optical power density and ensuing light absorption within each material can be strongly attenu- ated or enhanced by a small change in the layer thickness. Electronic losses include excited carriers that are not extracted from the device and/or excited carriers that lose energy in the extraction process, such as those arising from non-radiative recombination pathways and imper- fect contact materials. These are generally the same issues that need to be avoided in order to create highly efficient single-junction devices, with the added consideration of the interlayer in 2T devices. Moreover, design con- straints in tandem cells require somewhat different cri- teria from those in single-junction cells, which can affect the electronic properties of the constituent junctions. For example, in the most effective tandem device, the front cell should be thick enough to absorb all photons above the bandgap in a single pass of light, rather than

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Table 1 | **Current record reported efficiencies and configurations for each of the perovskite tandem technologies**

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Architecture** | **Wide-gap material** | **Wide bandgap (eV)** | **Narrow-gap material** | **Narrow bandgap (eV)** | **Transparent electrode/ interlayer** | **Efficiency (%)** | **Refs** |
| Four-terminal hybrid | Rb-FA0.75MA0.15Cs0.1PbI2Br | 1.73 | Interdigitated  back-contact silicon | 1.1 | MoO*x*/ITO | 26.4 | 46 |
| Two-terminal hybrid | Cs0.17FA0.83Pb(Br0.17I0.83)3 | 1.63 | Silicon heterojunction (IR-enhanced) | 1.1 | ITO/SnO2/ZTO  and ITO | 23.6 | 53 |
| Four-terminal all-perovskite | FA0.3MA0.7PbI3 | 1.58 | FA0.6MA0.4Sn0.6Pb0.4I3 | 1.25 | MoO*x*/Au/MoO*x* | 21.0 | 60 |
| Two-terminal all-perovskite (solution processed) | FA0.83Cs0.17Pb(I0.5Br0.5)3 | 1.80 | FA0.75Cs0.25Pb0.5Sn0.5I3 | 1.24 | ITO/SnO2/ZTO | 17.0 | 23 |
| Two-terminal all-perovskite (vacuum-deposited front cell) | Cs0.15FA0.85Pb(I0.3Br0.7)3 | 2.0 | MAPbI3 | 1.6 | Doped C60/TaTm | 18.0 | 61 |

ITO, indium tin oxide; TaTm, N4,N4,N4ʹʹ,N4ʹʹ-tetra([1,1ʹ-biphenyl]-4-yl)-[1,1ʹ:4ʹ,1ʹʹ-terphenyl]-4,4”-diamine; ZTO, zinc tin oxide.

NATURE REVIEWS | **CHEMISTRY** VOLUME 1 | ARTICLE NUMBER 0095 | **13**

# REVIEWS

**a** 1,250

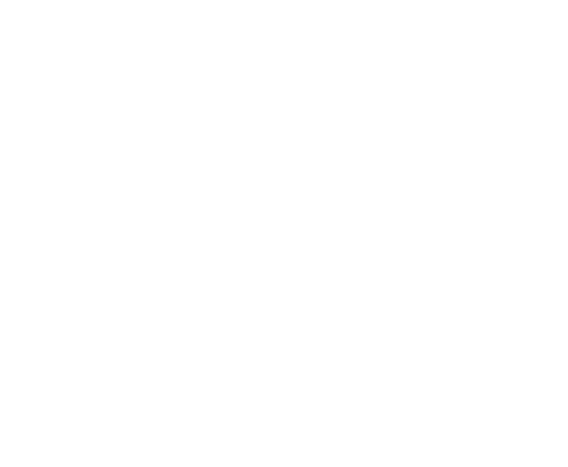
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Maximum annual energy yield (kWh m–2)

750

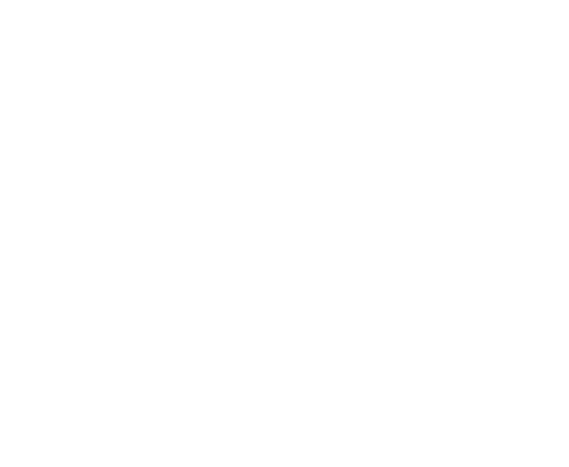
**b** 1.04

1.03



2T

Pk Si



2T

4T

Pk

1.02

1.01

Eﬃciency derating factor

500

1.00

0.99

0.98

250

0.97

0 0.96

0ax 1ax 2ax 0ax 1ax 2ax 0ax 1ax

2ax

0ax 1ax 2ax 0ax 1ax 2ax 0ax 1ax

2ax

Golden, CO Mohave desert Seattle, WA Golden, CO Mohave desert Seattle, WA

Figure 7 | **Realistic energy yield of perovskite–Si tandems compared with single-junction silicon and perovskite cells. a** | Calculated maximum annual energy yield of optimized perovskite-on-silicon tandem solar cells at different

locations with fixed (0ax), one-axis (1ax) or two-axis (2ax) sun-tracking systems. The tracking systems rotate the photovoltaic modules to face the optimum direction for power production in either the east–west (1ax) or east–west and north–south axes (2ax). **b** | Efficiency derating factor (describing how well a cell performs under real conditions in comparison with a

single-junction silicon cell) for optimized perovskite-on-silicon tandem solar cells and single-junction perovskite cells at different locations with fixed (0ax), one-axis (1ax) or two-axis (2ax) sun-tracking systems. 2T, two-terminal perovskite-Si tandem cell; 4T, four-terminal perovskite–Si tandem cell; Pk, single-junction perovskite cell; Si, single-junction silicon cell. Parts **a** and **b** are adapted with permission from REF. 87, Royal Society of Chemistry.

benefiting from two passes of light due to reflection from a rear electrode, as is the case for single-junction cells employing metallic rear electrodes. The carrier diffusion length limits the thickness at which a perovskite cell can operate efficiently: if the cell thickness exceeds the carrier diffusion length, voltage and current will be lost.

In the most efficient 4T hybrid tandem cells, reflection from the top of the perovskite cell (10–20% of incident light) and parasitic absorption in the transparent contacts (15–30% of incident light) dominate optical losses44. This results in EQEs of only 50–65% in the silicon sub-cell in the region beyond the bandgap of the perovskite cell (at which the perovskite cell does not absorb, but pho- tons have to pass through the cell before reaching the Si layer), which greatly limits the performance. The use of a perovskite with a non-ideal bandgap also limits the over- all efficiency. The same kind of losses dominate the most efficient all-perovskite 4T tandem cells, where a maxi- mum EQE of 40% attained in the rear low-gap perovskite cell results in only a 2.9% efficiency boost60. As a sim- ple thought experiment, if this 40% EQE in the infrared region could be increased to 90%, then an all-perovskite tandem cell efficiency of nearly 25% could be achieved. Better rear-cell performance in an all-perovskite tandem cell was achieved by Snaith and co-workers, reaching a 60% EQE and an overall 4.5% efficiency boost, likely due to the use of more transparent contacts. However, the total tandem performance was slightly lower owing to higher voltage losses in the front 1.6 eV cell23. In all of these 4T devices, gains were made most readily by improvements in the transmission of the transparent

contacts and optimization of the perovskite front sub- cell; however, we note that the challenge of making trans- parent enough contacts is a fundamental problem for the 4T architecture.

In the most efficient 2T hybrid tandems, optical losses again dominate, with front-surface reflection and parasitic absorption accounting for an effective 20–30% reduction in short-circuit current compared with the expected values53. The efficiency could be increased by minimizing front-surface reflection and by employing a slightly wider- gap perovskite, which, owing to the current-matching criteria, would allow both increased current and voltage in the tandem cell. The most efficient all-perovskite 2T tandem cell (the architecture in which one sub-cell is thermally deposited and the other solution processed) generates a lower efficiency than that of the impressively efficient 1.6 eV sub-cell alone. This is due to the use of a far-from-ideal bandgap combination, which is a result of the limited choice of perovskites that can be vacuum deposited61. Solution-processed all-perovskite tandem cells are limited mainly by electronic losses related to the wider-gap perovskite sub-cells; they suffer a large voltage deficit of 0.8 V between the bandgap and *V*oc, whereas

the best perovskite devices have voltage deficits of only

~0.4 V (REF. 23). In fact, the narrow-gap Pb–Sn mixed- metal perovskite has few electronic losses, with an impressively low deficit of 0.41 V. The loss in the wide- gap sub-cell is likely due to a combination of non-ideal contact materials and voltage losses due to halide segre- gation. The wide-gap cell also limits the photocurrent. Remarkable improvements could thus be achieved by

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# REVIEWS

2.6

Bottom cell bandgap (eV)

2.4

2.2

2.0

1.8

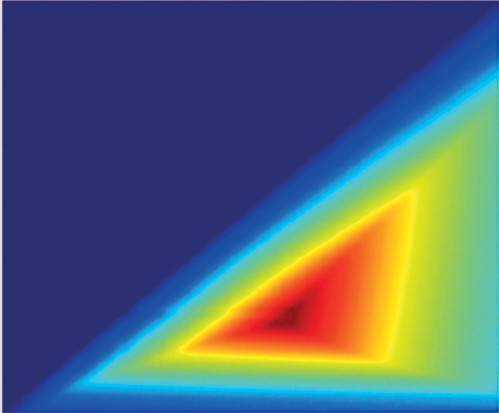
1.6

1.4

**a All-perovskite triple junction**

PCE (%)

46.9



Shockley–Queisser

1.22 eV base absorber



40.2

33.4

26.8

20.1

13.4

6.70

2.6

2.4

Bottom cell bandgap (eV)

2.2

2.0

1.8

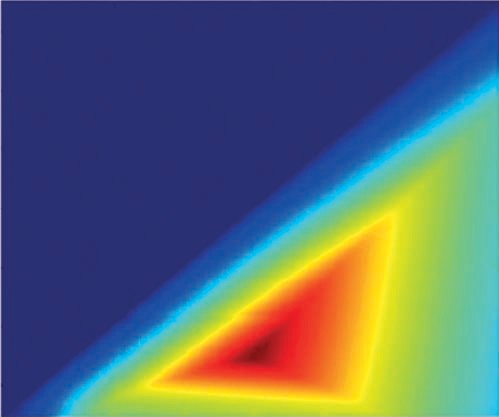
1.6

1.4

**b Si-perovskite triple junction**

PCE (%)

49.4



Shockley–Queisser

1.1 eV base absorber



42.3

35.3

28.2

21.2

14.1

7.10

1.4 1.6 1.8 2.0

2.2 2.4 2.6

0.00

1.4 1.6 1.8 2.0

2.2 2.4 2.6

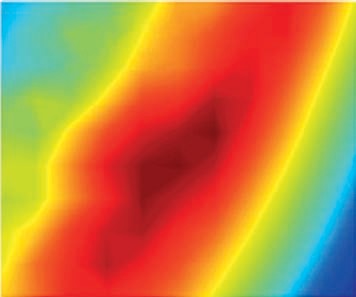
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Top cell bandgap (eV) Top cell bandgap (eV)

### c

Bottom cell bandgap (eV)

1.3



TMM optimization Perovskite–perovskite

1.2

1.1

1.0

0.9

PCE (%)

32.2

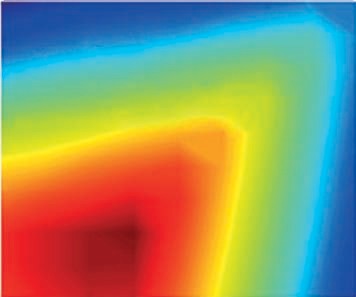
29.8

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**d**

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TMM optimization Perovskite–perovskite– perovskite (1.22 eV)

Bottom cell bandgap (eV)

1.7

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PCE (%)

33.0

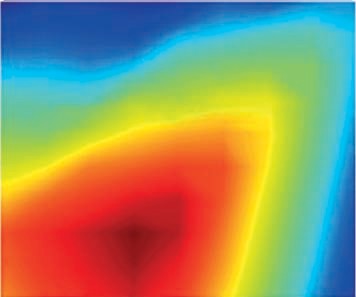
29.4

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**e**

1.7



TMM optimization Perovskite–perovskite–Si

Bottom cell bandgap (eV)

1.6

1.5

1.4

PCE (%)

35.3



32.3

29.3

26.2

1.4 1.6

1.7

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1.9 2.0 2.1 2.2

2.3

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1.9

2.0 2.1 2.2

Top cell bandgap (eV) Top cell bandgap (eV) Top cell bandgap (eV)

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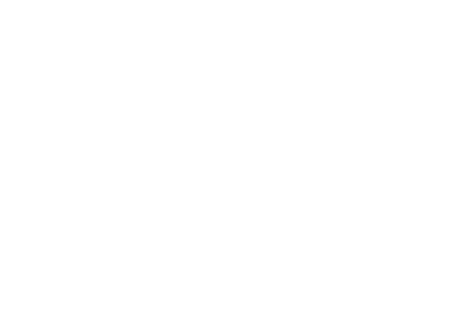
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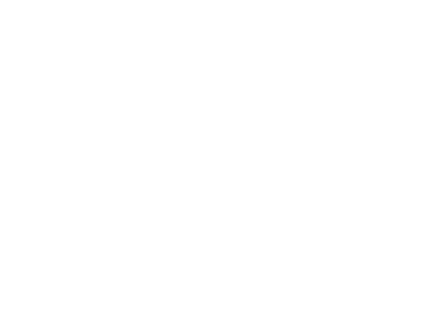
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1.1 eV Si cell

1.44 eV Perov. cell

1.95 eV Perov. cell



1.1 eV Si cell

1.44 eV Perov. cell

1.95 eV Perov. cell

Combined

17.5

Current density (mA cm–2)

15.0

12.5

10.0

7.5

5.0

2.5

0.0

400 600 800 1,000 1,200

0.0

0.5 1.0 1.5 2.0 2.5 3.0 3.5

Wavelength (nm) Voltage (V)

Figure 8 | **Triple-junction solar cells.** Calculated Shockley–Queisser limits for triple-junction solar cells containing a base absorber with a 1.22 eV (part **a**) or 1.1 eV (part **b**) bandgap. The dashed line traces the peak power conversion efficiency (PCE) for the whole range of front-cell bandgaps, and the white circles indicate the bandgap combination where efficiency is highest in each case. Maximum PCE of double-junction perovskite-on-perovskite solar cells with optimized thicknesses (part **c**), determined by shifting bandgaps and using optical transfer matrix and single-diode device modelling based on realistic perovskite diode characteristics. Maximum PCE of triple-junction perovskite-on-perovskite-on- perovskite (part **d**) and perovskite-on-perovskite-on-silicon (part **e**) solar cells with optimized thicknesses, determined by using a fixed perovskite base absorber with a 1.22 eV bandgap (part **d**) or a silicon heterojunction base absorber with a

1.1 eV bandgap (part **e**) and shifting the bandgaps of the middle and front absorbers. Calculated external quantum efficiencies (EQEs) (part **f**) and current–voltage curves (part **g**) of an optimized perovskite-on-perovskite-on-silicon solar cell with an ideal bandgap combination of 1.95, 1.44 and 1.1 eV. TMM, transfer matrix method. Parts **a**, **b**, **c**, **d**, **e**, **f** and **g** are adapted with permission from REF. 90, American Chemical Society.

electronically and optically improving the wide-gap perovskite sub-cell. In addition to these losses, some voltage loss (~0.1 V) results from the interconnecting layers, along with the same optical losses as the other tandem cells, which should be addressed to boost the performance of these devices.

Obtaining perovskite solar cells with optimum per- formance may require the implementation of more intri- cate optical-loss avoidance strategies, such as texturing or plasmonic structuring of the perovskite or contact layers, which is already employed in silicon cells.

The physical connection of the sub-cells in 4T architectures is currently challenging. At present, no 4T modules of any technology have been commercially installed. The use of power inverters, necessary forconnect- ing a module to the grid, accounts for a substantial propor- tion of the overall module cost91, which makes connecting each sub-cell with its own inverter a less attractive option in terms of cost. One possible solution would be for the sub- cells to be connected in series at the module level. Current matching between the front and rear cell is necessary in this approach and could be achieved by adjusting the

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# REVIEWS

Roll-to-roll processing

The process of creating devices on a continuous roll of flexible plastic or metal foil. If solar cells could be fabricated in this way, it is thought that the production cost could be a fraction of that for current wafer-based and

module-based production processes.

relative size of the cells36. However, shadowing on the bot- tom cells caused by wiring and interconnections of the upper cells brings new concerns. It remains to be seen if this challenge could be addressed or if it proves to be a deal breaker for the real-world application of 4T architectures. A further, longer-term, challenge is to scale the pro- cessing of high-quality layers to a large area. As the size of a cell increases, the likelihood of physical defects, such as pinholes, which hinder the performance, increases. There are many large-scale solution-coating options — including slot-die coating, inkjet printing, blade coating and spray coating — but it remains to be seen whether reproducible large-area devices with a very low level of non-uniformities can be fabricated by any of these meth- ods. Promisingly, the most efficient 2T hybrid tandem cells have been implemented in 1 cm2 devices, which is larger than most lab-scale solar cells. This result suggests that the deposition methods used could be scaled-up to the industrial level without losing performance53. The good performance over a larger area was attributed to the conformal coating of the ALD-based interlayers used. Even the more challenging 2T all-perovskite tandem cells showed fairly small performance drops when mov- ing from the 0.2 cm2 test cells to 1 cm2 devices23. If solu- tion processing of the perovskite layers ultimately proves unsuitable for the significantly larger area scale-up needed to fabricate modules, the option of physical vapour dep- osition may become more attractive. However, physical vapour deposition does present further challenges related to attaining compositionally uniform films of the multi- component alloyed perovskite materials necessary to

achieve the optimum bandgaps61.

A critical concern for the deployment of perovskite- based solar cells is stability. A key metric when selling panels is the cost per kilowatt hour of produced elec- tricity over the panel’s lifetime: the longer the module can be guaranteed for, the lower the LCOE. For all-per- ovskite tandem cells, the stability of Sn–Pb alloys has not been fully confirmed, although the stability appears closer to that of neat Pb perovskites than that of neat Sn perovskites, which is promising. The stability of Pb-based perovskites is unproved, even after years of real-world testing, although encouraging recent stability data show that they should be able to match the stability of Si92. Using industry-standard packaging, semi-transparent perovskite sub-cells and full 2T hybrid tandem cells have been reported to be able to survive the stringent damp heat test prescribed by the International Electrotechnical Commission (IEC), which is a key metric that commer- cial modules must pass53,93. The success of tandem cells in passing this test bodes well for their long-term stability

and real-world utility. This is a very important point to prove — one half of the module failing before the other would clearly be a show stopper.

### Conclusions

Multi-junction solar cells incorporating perovskites have emerged as an exciting field of research, which has shown rapid progress and has high commercial interest. The best cells are already more efficient than single- junction perovskite solar cells and getting very close to the best Si cells, despite only a few years of intensive research efforts in comparison with many decades for Si. The impressive efficiencies and tunable bandgaps of perovskite semiconductors make them ideal candidates for use in multi-junction architectures, either as hybrid cells incorporating a more established technology or as all-perovskite multi-junction cells. They offer an unprec- edented opportunity for improving upon the power per cost of commercially available PVs.

Perovskite products are not yet on the market, though we anticipate that to change in the next few years. The strongest selling point of the hybrid tandem concept is that it allows perovskites to potentially leverage an estab- lished technology in order to enter the competitive PV market. We anticipate that hybrid tandems will likely be the first iteration of perovskite-containing modules to enter the market. They will effectively be a ‘boost’ to Si modules, providing significant improvements for minimal added cost and leveraging the scaled production of the silicon industry. Once the large-scale manufacturing of perovskites has been established and the inevitable prob- lems solved, all-perovskite tandem cells andpossibly triple- junction cells will likely be the more attractive options as an even more inexpensive technology with lower capi- tal expenditure for scale-up. Owing to the removal of the rigid Si substrate, it will be possible to address new applications with all-perovskite multi-junction modules, promoting the possibility of lightweight and flexible form factors. Flexible substrates would also allow the option of using roll-to-roll processing to increase production through- put. In addition to requiring less expensive raw materials, we expect that, owing to a reduction in the transport and installation costs, the total cost per unit power will be sig- nificantly lower than that of Si once scaled-up, providing a low-cost high-energy-yield PV technology to power the demanding world of the future.

There are many challenges that still need to be addressed to leverage the full potential of perovskite-based multi-junction devices, but given the explosion of interest and rapid progress in perovskite research, we anticipate such challenges to be rapidly overcome.

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**Author contributions**

All authors contributed equally to the preparation of this manuscript.

**Competing interests statement**

The authors declare no competing interests.

**Note added in proof**

During the production of this article, a new efficiency record of 18.5% for all-perovskite 2T tandems was published by Rajagopal *et al.*, who employed new contact materials to reduce voltage losses: Rajagopal, A., Yang, Z., Jo, S. B., Braly,

* + - 1. L., Liang, P. W., Hillhouse, H. W., & Jen, A. K. Y. Highly efficient perovskite–perovskite tandem solar cells reaching 80% of the theoretical limit in photovoltage. *Adv. Mater.* <http://dx.doi.org/10.1002/adma.201702140>(2017).

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