

Scalable Ways to Break the Efficiency Limit of Single-Junction Solar Cells

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Solar photovoltaics will play a dominant role in the power generation of the zero-carbon future¹. Today, the market of large-scale solar power generation is dominated by silicon solar cells, where high-performance lab-scale devices are reaching their detailed-balance efficiency limit². With decreasing module prices³, cell efficiency becomes ever-more valuable as it becomes an increasingly important driver for further reductions in the levelized cost of electricity.

The next generation of solar cells thus strives to surpass the efficiency limit of single-junction solar cells. Multi-junction solar cells, such as those based on III-V materials, are already very efficient, yet these materials have remained prohibitively expensive for large-scale deployment⁴. In this special topic, we instead focus on nascent techniques and materials that offer the possibility to exceed the single-junction efficiency limit in a scalable (multi-km²) way in the future.

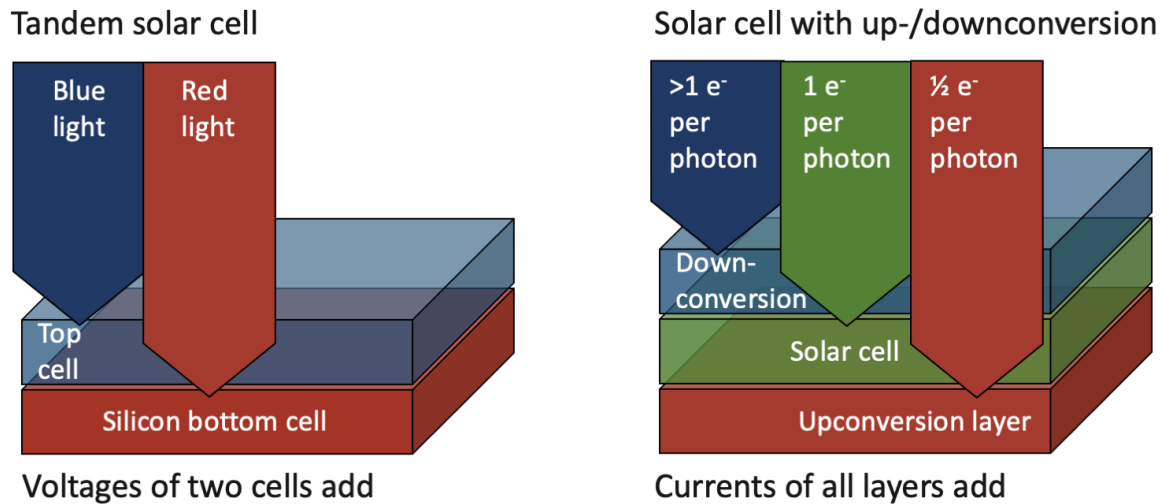


Figure 1. Working principle of a tandem solar cell (left) and an up- and downconversion-based solar cell (right). The tandem cell consists of two separate sub-cells, often the bottom cell is made of silicon and absorbs the low-energy part of the solar spectrum, and the top cell absorbs the higher-energy photons. The up- and downconversion cell often only consists of one solar cell junction, while the up- and downconversion layers transform the incident spectrum so that it can be better converted by the cell.

The detailed-balance single-junction efficiency limit assumes that the solar cell consists of a single absorber layer, in which an incoming photon generates a single electron-hole pair that can be extracted to perform work⁵. All common schemes that go beyond this limit break at least one of these assumptions: they use two or more absorber layers with different optical bandgaps or enable the generation of more than a single electron-hole pair, so that the broad solar spectrum can be used more efficiently. Broadly speaking, the resulting solar cells discussed in this special topic collection can be categorized into two main categories, namely *tandem solar cells* and *solar cells that make use of up- or downconversion* (Figure 1). Here, we bring together reports on scalable ways to overcome the limitations of single-junction solar cells using these approaches.

Tandem solar cells

A tandem solar cell consists of two individual sub-cells, each having its own bandgap and its own junction to aid electron-hole separation⁶. There are different ways to connect the two sub-cells. For instance, they can be electrically independent and only optically connected (four-terminal cell, 4T), permitting the independent power developed by each cell to be combined externally. Alternatively, they can be connected electrically in series (two-terminal cell, 2T), where the voltages intrinsically sum, but the current needs to be matched. Other geometries such as three-terminal cells are less common. Tandem cells could be highly relevant in the fast deployment of solar PV in the near future.

Solar PV technologies target the generation of electricity without the emission of significant greenhouse gasses, but to reach the goals of the Paris Agreement⁷, the transition towards this low-carbon generation of electricity must be rapid⁸. This gives a deployment advantage to new technologies that are compatible with existing technologies, production facilities and markets. Kamaraki et al. show that perovskite/silicon tandem solar cell fulfil many of these requirements⁹. The authors work for the company Oxford PV, which is commercializing the technology. They claim that the most commercially viable tandem cell is a 2T configuration because the 4T cells would require doubling the number of inverters, which is cost-prohibitive in the current market. Si solar cells are chosen as the most promising bottom cell because of their current dominance, proven long-lasting performance, and low cost. For the top cell, it is not enough to be efficient or low-cost, the cell must be both—particularly of comparable efficiency to the bottom cell. Thus, they describe how a metal halide perovskite top cell potentially fulfils these requirements since high-quality films can be formed by a variety of simple, low-cost deposition techniques. In addition, the technological compatibility of perovskite cells with the existing and highly developed Si industry allows these tandems to leverage the decades of development in Si cell and module fabrication, as well as systems and business development. An important commercial consideration is that perovskite/Si tandem modules offer a higher efficiency than traditional Si-only modules, which allows the producer to charge a premium in applications where space is limited, such as in rooftop systems. After market introduction, they emphasize that the learning curve of the tandem cells needs to keep pace with Si-only cells to remain competitive. They are optimistic that large efficiency potential and the connection to the established, large-scale Si industry should make this possible.

There are still several materials challenges to overcome before perovskite-based tandem cells become a reality. Several of these are addressed in this special topic collection. A high photoluminescence quantum yield (PLQY) is critical for all solar cell materials. Perovskites often show high PLQY, but it can be diminished by sample morphology and material purity. Screening material compositions and fabrication techniques therefore requires a rapid method to study the PLQY of a perovskite sample. Akhundova et al. show that the spectral shape of the photoluminescence (PL) from wide-bandgap perovskites correlates with the PLQY of a sample¹⁰. The authors assign this correlation to the observation both the PLQY and the spectral shape reflect the domain size dispersion because of its role in the funnelling of carriers into

trap sites. The size dispersion is greatly affected by processing parameters such as annealing conditions, and these results highlight the importance of carefully controlling fabrication.

The fabrication of perovskite thin films needs to be scalable without compromising luminescence efficiency. Ishteev et al. demonstrate a new deposition technique for all-inorganic, large-bandgap perovskite thin films¹¹. They fabricate the precursors, CsBr and PbBr₂, by dry, mechanical milling before deposition in a single-source CVD process under mild vacuum (0.02–1.00 mBar). All these techniques are in principle scalable, and the authors are able to vary the ratio between the precursors (*i.e.* CsBr:PbBr₂) to search for the most luminescent composition. They observe greatest brightness from a 35:65 ratio, a composition that forms both the CsPb₂Br₅ and the CsPbBr₃ perovskite. They demonstrate that this technique can be used to conformally coat textured surfaces, but identify that the stability and PLQY will require further improvements.

For the full device stack, perovskite tandem cells require transparent electrodes (4T) and interconnection layers (2T). These layers have received considerable attention because of their importance for the overall performance. The interconnection layer between the two subcells of a 2T tandem cell ideally offers good recombination properties, while transmitting all low-energy light. Koç et al. calculate the optical transmission of an interconnection layer consisting of thin Ag between two transport layers in a perovskite-perovskite tandem cell¹². They report transfer matrix calculations that explore the transmission of light into the low-bandgap bottom cell when the thicknesses of the silver layer and the adjacent transport layers are varied. They find two trade-offs. Firstly, the refractive index of the transfer layers is important to the transmission of light. Ideally the index would be larger than 1.9, however, by the Kramers-Kronig relationship these high indices will also lead to larger parasitic absorption. Secondly, the silver layer should be as thin as possible for good light transmission (even 3 nm already reduces the photocurrent from the bottom cell), but thicker layers lead to larger lateral conductivity.

For 4T tandem cells, the top cell needs to be fully transparent to below-bandgap light. However, the transparent conductive electrode required induces large parasitic absorption and often used indium-tin oxide (ITO) is too brittle to be implemented in flexible solar cells. As in the previous example for interconnection layers, thin metal layers can also perform as excellent transparent conductive electrodes. Spinelli et al. sandwich a sputtered layer of Ag or co-sputtered Ag/Cu between two layers of ITO, and these ITO-Cu/Ag-ITO electrodes show a NIR transmission similar to ITO when the layer thicknesses are optimized¹³. They perform this using transfer-

matrix modelling, identifying an optimal transmission for thicknesses of 40 nm for the 1st ITO layer, 5 nm for the metal, and 65 nm for the 2nd ITO layer. The layers were then processed in a full device stack with a large-bandgap perovskite as the active layer and performed slightly better than the same device with an ITO contact. In addition, they find that the initial sheet resistance of the ITO-metal-ITO contact is about half that of pure ITO (200 nm), and that it retains this value upon repeated bending cycles, while the ITO resistance doubles after about 1000 cycles. The authors work for Saule Technologies, a company commercializing perovskite solar cell products, and they are confident that this contact architecture can provide a scalable and efficient solution for applications.

While perovskite/silicon and perovskite/perovskite tandem cells receive significant attention from the research community because of their potential for implementation at scale in large-area photovoltaics, different tandem configurations offer interesting properties for other applications. For example, III-V semiconductor/Si tandem solar cells combine a low-cost and well-established Si bottom cell with a highly efficient III-V top cell. While too expensive for large-area applications, these cells might suit the high-performance end of the market, for example for vehicle-integrated PV. Whitehead et al. fabricate a 4T GaAs/Si tandem cell that is mechanically stacked by combining various thicknesses of the GaAs with a Si cell that features interdigitated back contacts¹⁴. The 4T configuration is necessary here because the bandgap of GaAs is too low for a current-matched 2T combination with Si. The back-contacted silicon cell makes the stacking and contacting easier. The authors find that the tandem cell efficiency does not depend too heavily on the GaAs thickness as long as it is thicker than 1.9 μm . All tandem cells show excellent performance with >30% power conversion efficiency, and their cell with 2.8 μm GaAs shows the highest tandem efficiency (32.57%). The performance for the various layer thicknesses is confirmed with transfer matrix modelling for the optical transmission and absorption, Lambertian light-trapping for the Si cell, and the Hovel method to study the effect of photon recycling. The low refractive index of the glass interlayer ensures that the GaAs cell benefits from the effect of photon recycling by trapping the emitted light inside the top cell.

Up-and downconversion

Like tandem solar cells, two or more effective bandgaps are also required for the up- and downconversion schemes. However, only one of them needs to form a semiconductor junction to extract charge. The other bandgap can then be used to down-convert the energy if the bandgap is higher than the one that forms the junction, or upconvert if it is lower. There are several mechanisms for both up- and downconversion. In this special topic collection, we find

contributions on singlet fission and multiple exciton generation as downconversion processes, and triplet-triplet annihilation and intermediate-gap solar cells as upconversion processes.

Singlet fission is a process that can be used in photon downconversion, and the most thoroughly studied materials are crystalline, conjugated small molecules such as the linear acenes. Photon absorption in these materials initially generates spin-singlet excitons. These are converted by fission into pairs of lower-energy spin-correlated triplet excitons¹⁵. The process is spin-allowed and is hence can be rapid and efficient in materials where it is energetically favourable¹⁶. In that respect, singlet fission appears to be an ideal downconversion mechanism when combined with a low-bandgap solar cell. In practice, however, multiple challenges arise when implementing these organic materials in a practical solar cell architecture. Cheung and Kaake investigate the effect of the exciton binding energy and endothermicity on the potential solar cell efficiency¹⁷. The binding energy comes from the Coulombic interaction between the electron and hole of the exciton. The endothermicity, also called entropic gain, is observed in many singlet fission materials and stems from the greater number of microstates possible when one singlet exciton converts into two triplet excitons¹⁸. The authors use both the optical gaps of the singlet fission material and the low-bandgap solar cell as free parameters in a detailed-balance based efficiency calculation and assume charge transfer from the triplet exciton to generate current. They find that in simple two-bandgap systems, the efficiency drop from the exciton binding energy can be partially recovered by the endothermicity. However, at a high exciton binding energies (*e.g.* 0.5 eV), the maximum achievable efficiency drops from 43.9% to 31.0% even assuming an effective endothermicity of 0.25 eV. Then, a double heterojunction is introduced, with a bridge molecule that serves to accept electrons from the triplet excitons, much like in ternary blend organic solar cells¹⁹. This bridge molecule reduces recombination and aids charge separation. The result is that the maximum achievable solar cell performance remains high even at high exciton binding energies. Revisiting the example mentioned previously, with 0.5 eV exciton binding energy and 0.25 eV endothermicity, they show that the bridge molecules would lift the efficiency potential from 31.0% to well above 40%. While charge separation from singlet fission-generated triplet excitons may lead to high efficiency²⁰, the most promising experimental realizations with singlet fission and silicon cells so far rely on energy transfer²¹.

Many downconversion processes are technically multiple-exciton generation processes, but conventionally the term *multiple exciton generation* (MEG) refers to the inverse Auger process in inorganic semiconductors that generates two or more band-edge electron-hole pairs from

one high-energy photoexcitation. This process is inefficient in bulk materials both energetically and in terms of quantum yield, but prospects are improved in colloidal quantum dots due to factors that include improved momentum matching²². Solar cells utilizing MEG to achieve greater-than-unity photon-to-carrier yield at select wavelengths have been demonstrated,²³ but have not yet reached high energy-conversion efficiencies. Writing in this collection, Pusch et al. show that equilibrium-only models are insufficient in assessing the efficiency potential of such cells, because of the voltage dependence of the inverse process of MEG, Auger recombination²⁴. As voltage increases, Auger recombination starts to play a larger role. In contrast, all relevant processes in conventional solar cells are well separated in time. In these conventional cases, the external quantum efficiency measured at short circuit can be used to calculate the photocurrent, and the superposition principle applies. However, when the efficiency potential of MEG solar cells is assessed, microscopic reversibility must be explicitly considered to properly treat the interplay between MEG and Auger recombination. Pusch et al. devise a model that incorporates these considerations for a quantum dot-based device assuming equidistant, discrete energy levels. This model reproduces experimentally observed EQE curves, and they show that the quasi-Fermi level splitting varies across the device due to the microscopic reversibility. The MEG process thus depends on the applied voltage and, as a result, the calculated IV curves show a lower open-circuit voltage compared to a model that assumes equilibrium conditions. Inclusion of trion states to the Auger recombination rate further reduces the efficiency potential, so preventing these charged states in devices will be critical to achieve significant efficiency gains over single-junction solar cells.

MEG and singlet fission both harness high-energy photons to generate multiple excitons of lower energy. In principle, direct extraction of the energy of *hot carriers* is also possible. There are major challenges, however, which are in part related to the generally rapid relaxation of hot carriers to the band-edge. Esmailpour et al. investigate the hot-carrier relaxation in type-II InAs/AlAsSb multi-quantum well structures²⁵. They study the effect of different barrier thickness on hot carrier relaxation by measuring the PL spectrum at different excitation intensities. By measuring the excess temperature from the PL spectrum for different barrier layer thicknesses, they find that the barrier layer thickness has an effect of the effective temperature because of changes in the cooling rates. The authors explain this difference by a difference in phonon scattering rates. Supported by DFT calculations, they find that the larger barrier layers (higher InAs/AlSb ratio) show a reduced phonon density of states which leads to an increase in the phonon scattering time.

While all the previous examples aim to make better use of the high-energy part of the solar spectrum, *upconversion* does the opposite. In such schemes, below-bandgap photons are absorbed in an upconverter material and transformed into higher-energy photons or excitations. An attractive strategy for incoherent photon upconversion efficiencies is based on triplet-triplet annihilation (TTA), also known as triplet fusion, where pairs of spin-triplet excitons are combined to form a higher-energy, emissive spin-singlet exciton. The most efficient schemes have relied on external sensitizers that absorb incident light and then transfer this energy to annihilator/emitter molecules. Traditionally these schemes relied on metal-organic complexes to serve as sensitizers²⁶, and more recently colloidal quantum dots^{27,28} and lanthanide-doped nanoparticles sensitizers combined with organic emitters²⁹. With the rise of perovskites as the dominant thin-film semiconductor (at least in the research community), perovskite nanocrystals³⁰ and films³¹ have also now been used as sensitizers for TTA upconversion. Surprisingly, even bulk perovskites performed very efficiently thanks to their high charge carrier mobility and long charge carrier lifetimes. For application in solar cells, these upconversion systems need to work efficiently at sub-solar fluxes, and under continuous illumination. Writing in this collection, VanOrman et al. study the effect of illumination on the efficiency of a (MAFA)PbI₃ perovskite/rubrene upconversion bilayer³². They show that the continued illumination improves the upconversion efficiency, which correlates with an increase in the total perovskite PL that is associated with the relative suppression of short-lifetime decay channels. This is consistent with trap filling within the perovskite layer, because populating the triplet excitons in rubrene by the perovskite excitations competes with trapping within the perovskite and at the interface. Under continued illumination, they observe that the upconversion PL from rubrene rises more rapidly following additional pulsed excitation, providing further evidence that the transfer of energy into the rubrene triplet state is improved. The work highlights that the upconversion efficiency reported in these systems strongly depends on not only the incident power, but also the illumination history of the sample.

A theoretically elegant but technically challenging way to overcome the efficiency limit of single-junction solar cells are *intermediate-gap solar cells*^{33,34}. Here, a state within the bandgap of the absorber layer is used to provide a second absorption gap. The intrinsic challenge in this architecture is that the reciprocity of absorption and emission processes mean that charges in the intermediate gap typically recombine faster than photons can be absorbed at solar flux to generate excitations across the bandgap. This issue can be overcome by “parking” electrons in a second level slightly lower than the intermediate gap to prolong their lifetime. Huang et al.

investigate theoretically such a system where a bilayer of Sn-doped AgAlTe_2 and LiInTe_2 is used to also aid charge separation in a type-II heterojunction³⁵. They calculate the band structure of both pure materials and those doped with various group-IV metals. The combination of the Sn-doped materials provides good lattice matching and maintains the type-II heterojunctions of the undoped materials.

Concluding Remarks

Overall, this special topic collection shows the breadth of studies, the novelty, and the creativity that the research community is applying to circumvent the single-junction efficiency limit for solar cells. While some concepts, such as perovskite/silicon tandems, are close to market entry, others while demonstrated, are not yet commercially viable for large-scale terrestrial applications, such as the III-V based technologies. The up- and downconversion technologies on the other hand have enormous potential, both for high efficiency and for low cost, but are further away from commercial implementation. We embrace this wide range of technologies at different stages of research, and we see a bright future for high-efficiency, large-scale solar cell applications.

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