Perspective

Photonics for Photovoltaics – advances and opportunities

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Photonics for Photovoltaics – advances and opportunities

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ABSTRACT

Photovoltaic systems have reached impressive efficiencies, with records in the range of 20-30% for single-junction cells based on many different materials, yet the fundamental Shockley-Queisser efficiency limit of 34% is still out of reach. Improved photonic design can help approach the efficiency limit by eliminating losses from incomplete absorption or non-radiative recombination. This Perspective reviews nanopatterning methods and metasurfaces for increased light incoupling and light trapping in light absorbers and describes nanophotonics opportunities to reduce carrier recombination and utilize spectral conversion. Beyond the state-of-the-art single junction cells, photonic design plays a crucial role in the next generation of photovoltaics, including tandem and self-adaptive solar cells, and to extend the applicability of solar cells in many different ways. We address the exciting research opportunities and challenges in photonic design principles and fabrication that will accelerate the massive upscaling and (invisible) integration of photovoltaics into every available surface.

Keywords: Photovoltaics, nanophotonics, light trapping, emission control, solar cells, nanomaterials

1. INTRODUCTION: COST REDUCTIONS OF PV TECHNOLOGY, SCALING TO TW TECHNOLOGY

Photovoltaics (PV), the direct conversion of sunlight into electricity, can provide a major contribution to the energy transition in our society. Together with other renewable energy sources such as wind, hydroelectric and geothermal energy, it can replace the majority of conventional energy technologies based on fossil fuels that generate the CO_2 which causes climate change. Over the years, the cost of PV panels (per generated power) has consistently decreased by 40% for every doubling of the installed PV capacity (Fig. 1a). This is due to reduced materials and manufacturing costs, partly as a result of mass manufacturing, and a gradual improvement in conversion efficiency as research and technology progressed over the years. Today, the manufacturing of a solar panel costs ~0.21 US$/$W_p$, *i.e.* ~60 US$ for a standard 280 W_p PV panel.[1]

Today, less than 1% of our total energy use (electricity, fuel, heat) is generated by PV. To generate half of our energy need by PV,[2] the worldwide PV capacity must increase some ~100-fold, from the 600 GW_p installed today to ~60 TW_p in 2050. This corresponds to about 25 000 solar fields the size of the biggest solar field to date, the Bhadla Solar Park in India.[3] Spread all over the world, this is an extremely ambitious but realistic goal.[2] Developing building- and landscape-integrated PV concepts will help expand the PV capacity to very large areas. Economically, such a large expansion of PV worldwide will only be realized if the cost of PV (including storage) becomes similar to that of energy generation based on fossil fuels. This requires a further reduction in the price of PV panels by another factor 2 to ~0.1 $\$/W_p$ (Fig. 1a).[4] Total system costs are higher, still above 1 $\$/W_p$ for residential systems and above 0.5 $\$/W_p$ for utility-scale systems in most regions.[5] The price per kWh is then related to the specific yield, which depends heavily on location, weather, and specifics of the installation. Typical values vary from < 1000 kWh/kW$_p$ to almost 2500 kWh/kW$_p$.[6] Scaling up to 60 TW$_p$ requires a dramatic increase of the worldwide PV manufacturing production by a factor 14 or 40, depending on whether that goal is achieved in 2050 or 2030 (Fig. 1b). We note that there is still intense debate if *all* of the world's energy needs can be supplied by renewable energy, and the fraction that can be delivered by PV depends on the cost of storage technology that must still be developed.[2]

A key factor in reducing the costs of PV installations (per W$_p$) is to increase the conversion efficiency of the solar cells. Today, the cost of the PV cell itself constitutes only a relatively small part of the PV systems cost, and therefore an increase in efficiency is a nearly linear driver in decreasing PV energy generation costs.[7] Silicon solar cells, which constitute ~90% of the total PV market, have a record conversion efficiency of 26.7%, thin-film cells based on CdTe, $Cu(In,Ga)(S,Se)_2$ (CIGS) or perovskite have records in the range 20-25%, and the overall record for single-junction PV held by thin-film GaAs cells is 29.1%.[8] So far, none of the PV materials have reached the fundamental single-junction Shockley-Queisser (SQ) efficiency limit of 34%,[9] and therefore there is much room for improvement. We note that, to understand the drive for increasing PV efficiency by single-% values, it is important to realize that PV technology encompasses an annual market of over 100 billion US$ and hence a 1% increase in efficiency for a particular cell material/design can have a value of hundreds of millions US$.[7]

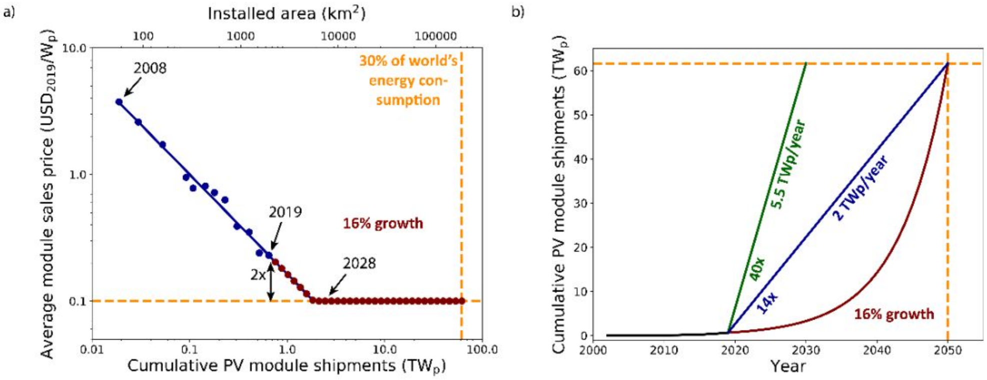

Figure 1. Cost reduction and PV installation growth. (a) Average sales price of Si solar modules normalized by the generated power under standardized conditions (W$_p$) as a function of total installed capacity since 2008. Data from the ITRPV report.[Error! Bookmark not defined.] Red data points are extrapolations of the 22% annual cost reduction at a yearly 16% growth of installed capacity, assuming that module prices will saturate at a price of 0.1 US$/W$_p$. (b) Historic realization (black) and 16% annual fixed growth scenario (red) of installed PV capacity. The linear curves correspond to increased PV installation rates (14/40× compared to today's ~0.1 TW$_p$/year to reach a 60 TW$_p$ target in 2050 or 2030.

In all PV materials, optical losses are a major reason that the Shockley-Queisser (SQ) limit is not reached: part of the sunlight is reflected from the cell, it is incompletely trapped inside the semiconductor, or it is absorbed in inactive layers of the cell; all these effects reduce the achievable photocurrent from the cell. Indeed, for the five record materials mentioned above, optical losses reduce the photocurrent by 3-7% compared to the SQ current limit.[8] Thin-film PV materials with efficiencies in the 13-17% range ($Cu_2ZnSn(S,Se)_4$ (CZTS), compound semiconductor quantum dots, organic) show even higher optical losses (20-30% compared to the SQ current limit), indicating large room for improvement. Aside from optical losses, electrical carrier recombination losses constitute a second main reason that the SQ limit is never reached. These losses are mostly determined by the electrical quality of materials and interfaces, and lower mostly the photovoltage of the cell; they form the main limiting factor to the total efficiency in most thin-film PV materials, especially CZTS.

Integration of suitably designed nanostructures that control the flow of light in the cell can help reduce the optical loss factors described above and mitigate the effects of carrier recombination on the cell voltage.[10–12] In addition to helping reach the SQ limit, photonic concepts can also bring solar cell efficiencies *beyond* the single-junction SQ limit by changing the working principle of the solar cell away from the assumptions made in the detailed-balance limit that is behind the SQ model. For example, altering the angular or spectral distribution of light entering or exiting the solar cell can lead to efficiencies well beyond 34%.[10,13,14] The most successful implementations so far involve using concentrating lenses and stacking solar cells in a tandem or multi-junction configuration where high-energy light is absorbed in the high-bandgap solar cell placed in front of a low-bandgap cell that absorbs the low-energy light.[15] More exotic concepts, including up- or down-conversion of the solar spectrum to match it better to the solar cell bandgap, are predicted to have similar limits, but so far have not been successfully implemented to improve record solar cells.[16]

This perspective reviews the state-of-the art of photonic design principles for increased PV efficiency. It first reviews light incoupling and light trapping (Section 2), the area in which light management of PV has traditionally focused most. It then describes opportunities for future research in reducing carrier recombination by enhanced light outcoupling (Section 3), enhanced spectral conversion in tandem and bifacial solar cells (Section 4),spectral shaping by down- and up-conversion (Section 5), and large-scale building and landscape integration of PV (Section 6). We conclude with a section addressing modeling of photonics for PV, adaptive PV, and large-scale fabrication. We summarize our perspective with the most important future opportunities for photonic design for PV (Section 7).

2. LIGHT INCOUPLING AND LIGHT TRAPPING

Standard wafer-based Si solar cells have excellent light coupling and trapping due to a macroscopic pyramidal surface texture that is made by chemical etching of the wafer. For thinner Si wafers and for other materials such texture cannot be easily made. As an alternative, (non)-resonant dielectric nanoscatterers or nanotexturing placed at the front of the cell can be used for efficient light coupling, either via Mie resonance interference engineering to enhance forward scattering or via optical impedance matching at the interface for example by tapered nanostructures.[17] When combined with a conventional anti-reflection coating, this creates broadband incoupling of light, minimizing reflection.[18–21] For the scattering approach, the effect is strongest with high-index materials, *e.g.* based on TiO_2 and Si. When made onto a flat electrically passivated semiconductor surface this creates “electronically flat, optically textured” solar cells with minimized surface recombination.[22] These nanophotonic approaches can exhibit similar reductions in reflection as seen for optimized planar antireflection coatings, but work over a broader range of incident angles and wavelengths.[20,21]

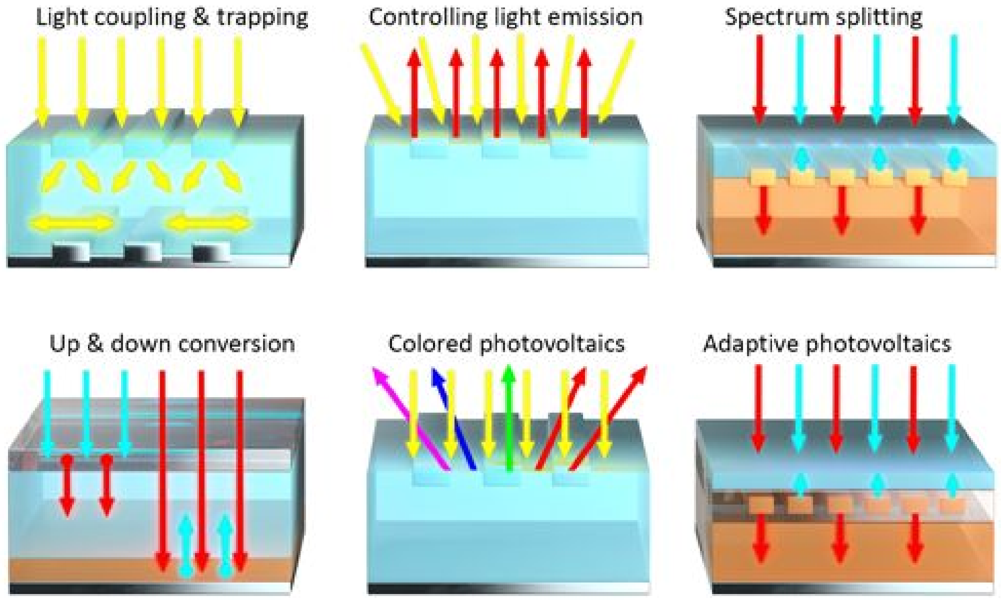

Figure 2. Schematic of light management geometries to optimize photovoltaics.

Light can be preferentially scattered into waveguide modes in a thin-film solar cell by tailoring the spatial distribution of resonant nanoscatterers.[23] In the presence of periodic patterning with dielectric structures, the guided modes of the thin silicon slab become leaky and can in- and out-couple to the incoming electromagnetic modes supported by the surrounding medium. As a result, the light path length at wavelengths near the bandgap in the film is enhanced and absorption increased. Improved light trapping enables the use of thinner semiconductor absorber layers, which reduces fabrication costs, requires less use of precious metals (e.g. indium in CIGS) and toxic elements (Pb in perovskites and quantum dots, Cd in CdTe) and enables (ultra-)thin flexible PV foils. Reducing thickness by improving light trapping also reduces bulk recombination and thereby enhances the photovoltage of the cell as long as additional recombination at the nanostructure is avoided.

Since periodic arrangement of scatterers generally works well only at discrete wavelengths and specific angles of incidence, engineered disorder has been explored to ensure that the spectrum of scattering wavevectors matches well with that of the waveguide modes of the absorber thin film.[24–30] By tailoring the resonant scattering cross sections of the scattering geometries light trapping can be optimized to match with the optical spectral range near the semiconductor bandgap. Intense research is going on to investigate what spatial frequency distributions in the scattering pattern, such as e.g. hyperuniform distributions, are best tailored for optimum light trapping in thin layers.

Ultimately, the light trapping efficiency is limited by reciprocity, as light scattering also couples light out of the cell.[31] From geometric optics, using Lambertian scattering, the maximum light path enhancement in a cell is $4n^2$, with n the refractive index. Specially engineered gratings and other narrow-band photonic designs have the potential to overcome this limit over certain spectral bands.[32–35] It will be interesting to see if recent work on non-reciprocal photonic structures can be applied to solar cells, which would provide new possibilities to further enhance light trapping.[36]

For all the nanostructures with periodic structures and engineered disorder, the effect of coherence could play an important role in the performance. Sunlight, emitted in a narrow cone to an observer on earth, exhibits a coherence length of ~80 wavelengths, *i.e.* ~40-80 microns in the wavelength range relevant for silicon.[37] For most diffractive patterns, 10 unit cells is sufficient to saturate any spatial correlation effects, such that the coherence of direct sunlight does not play a limiting role. Solar simulators have a similar coherence length, typically 20-30% lower than direct sunlight, so can provide a reasonable approximation for performance of nanopatterned solar cells under direct sunlight. However, the coherence length drops rapidly as the fraction of diffuse light increases, making solar simulators unsuitable for testing real world performance of cells with spatial correlations extending beyond a few microns expected to be deployed in regions with substantial power generation from diffuse light.[37]

Nanophotonic research on light trapping for PV initially started with using plasmonic nanoparticles as strong light scatterers.[38] It was then soon realized that dielectric nanoparticles are advantageous as they show lower optical absorption losses than the plasmonic metals.[17,20] A key feature of the plasmonic structures however, is the high electrical conductivity. Transparent Ag nanogrids were demonstrated with a tradeoff between conductivity and transmittivity that is better than for indium-tin-oxide, the conventional transparent contact layer for many solar cell designs.[39–41] Tailored metal nanowire structures can be made at large scale using soft-imprint lithography, while low-cost random geometries can be made by spray coating chemically synthesized nanowires on top of the solar cell.[40,41] A special geometry in which the combined effect of plasmonic and Mie resonance is relevant is in nanopatterned metal back or intermediate (for tandem cells) contacts, that serve both as current collectors and tailored light scattering geometries for efficient light trapping in thin cells.[23,42] Selective nanopatterning of interfaces can also reduce interface recombination at backcontacts.[43]

Effectively transparent metal contacts can also be made by reflecting light off specially shaped macroscopic metal contact fingers and busbars, directing light into the semiconductor rather than into the absorbing metal.[44] Similarly, using the principles of transformation optics, contact finger cloaking designs have been made to avoid interaction of light with the metal fingers.[45] The most modern high-efficiency Si solar cell designs are based on heterojunctions with selective and passivating electric contact layers which are very beneficial from an electronic point of view but show relatively high optical losses in the ultraviolet and/or infrared.[46,47] Tailored nanophotonic designs are required to reduce these optical losses. Reducing parasitic losses not only improves the photocurrent of the cell, it also reduces the thermal load, which is important as the cell voltage rapidly decreases for increasing operating temperature.

Finally we note several novel promising earth-abundant solar cell materials have recently been reported but show low photocurrent and could hence benefit from light management using nanopatterned interfaces. CZTS avoids the use of the scarce element indium that is used in CIGS, and has reached a conversion efficiency of 12.6%.[48] Since the main limiting factor in CZTS is the low voltage, the large optical losses (20% below S-Q limit compared to 3-7% for Si, GaAs, CdTe and halide perovskites) are often overlooked. Light management using nanopatterned interfaces will further improve the photocurrent for these cells. Most recently, $Sb_2(S,Se)_3$ has appeared as an interesting material that is fully based on earth-abundant constituents, with an efficiency of 10.0%.[49] In both materials, defect engineering and passivation will be key to further raise the efficiency, assisted by proper photonic design.

Even the strongly absorbing lead halide perovskites could benefit substantially from improving absorption near the band gap, especially in tandem cells where there is no back reflector.[50] Lead halide perovskites are also a particularly appealing material for testing nanophotonic approaches since their low surface and interfacial recombination velocities allow for increased surface area with little increase in non-radiative recombination.[51,52] Their sensitivity to a variety of solvents and process steps normally used in lithography make traditional patterning more challenging, although their soft nature and low crystallization temperature also enable direct nanoimprint lithography. [53–57] For characterization, and for applications where high light intensity is required such as lasing,[58] these lead halide perovskites are also generally more unstable than traditional materials used for photonic structures.[59] We note that any of these tests of novel photonic structures for PV devices needs to consider also how patterning and changes in the absorption and emission affect the electronic properties of the material, and ultimately the quality of the PV devices.

3. REDUCING RECOMBINATION BY CONTROLLING LIGHT EMISSION

An important strategy to increase PV conversion efficiency that has been somewhat neglected so far, is through control over light *emission* from the cell. The open circuit voltage of a PV cell is determined by the balance between radiative and non-radiative emission. Conventionally, research focuses on minimizing non-radiative decay processes through materials optimization, reducing and passivating carrier traps. However, a similar effect may be obtained if the radiative emission rate at the bandgap is enhanced by optimizing the optical density of states by photonic design.[10,60–65] This approach is most effective for PV materials with low photoluminescence (PL) quantum yield, either due to low material quality or intrinsic non-radiative losses, which is the case for all PV materials except GaAs which is a high-quality direct-gap semiconductor close to the radiative limit. In all other materials the open circuit voltage could gain >100 mV, corresponding to a >10% relative improvement in efficiency, by enhancing the radiative rate through enhanced light out coupling.[10,63–65] Also silicon solar cells, in which carrier recombination is ultimately governed by Auger recombination, could benefit substantially from a strong PL rate enhancement, as it would effectively drive it closer to the behavior of a traditional direct band gap semiconductor.

Reciprocity between light incoupling and outcoupling sets up a paradox of simultaneous optimization of light trapping and enhancing the radiative rate. Introducing photonic structures that have highly asymmetric light-matter interactions for different angle and photon energy make it possible to artificially break this in/out coupling paradox. Ideally, the cell couples in light with energy well above the bandgap over a wide angular range, while photons right at the band gap (corresponding to PL energy) only couple strongly to the direction corresponding to the sun.[10] The latter avoids the entropy increase associated with the conversion from collimated light from the sun to a broad angular distribution of light inside the cell, and allows the open circuit voltage to increase by >100 mV, while also allowing for better collection of diffuse light. One important consideration is that methods to control directional emission cannot come at the expense of photon absorption or enhance surface recombination, otherwise the efficiency gain is lost. Fortunately, nanophotonics provides the ability to engineer the angular distribution of the local density of optical states, pushing states from oblique angles to the surface normal to increase both absorption and emission towards the sun.[10,66] Such an approach should be implemented without additional parasitic absorption, for example using transparent scatterers. Highly directional PL would require solar tracking, that could be implemented in a cost-effective way if passive, or self-adapting photonic structures could be implemented to track the sun.

Controlled light outcoupling can also play a key role in the mid-infrared to improve radiative cooling. Although the concept was already demonstrated experimentally almost half a century ago,[67] it has received renewed interest recently with the development of tailored photonic structures. It has been shown to reduce surface temperature of a variety of materials, including solar cells, below ambient temperature, even under illumination.[68–73] Thermal management can have large impact: every 1 K in temperature rise reduces the cell efficiency by ~0.3-0.5%(relative).[74,75] Typical operating temperatures are 30-50 K above ambient,[75] which means that a solar cell with a certified efficiency of 25% will display an efficiency of between 19% and 23% under realistic conditions.

Pioneering work has already demonstrated that specially designed photonic structures enabled 5 K radiative cooling below ambient under full solar illumination, where a black paint control reached 60 K above ambient and aluminum 20 K above ambient.[69] Subsequent studies using metamaterial surfaces have demonstrated almost 10 K below ambient during daytime operation.[72] Efficient photonic engineering of IR emission enabling operation near ambient therefore could have large efficiency benefits under real operating conditions, approaching the gains of tandem cells, spectral conversion or pushing materials to the radiative limit.

Importantly, the lifetime of PV systems (typically guaranteed for 25-30 years for a high-quality Si solar panel) can be increased by an estimated 26-200% significantly increased by up to a factor of 2 if the operation temperature is kept low,[75] which can dramatically reduce the levelized cost of electricity. Such gains in lifetime may be even more pronounced for materials like halide perovskites, which are generally much more sensitive to elevated temperatures than standard crystalline Si and commercial thin-film materials. The primary challenge in this area of course is to apply a radiative cooling coating that does not hamper solar conversion in the visible and near-infrared.

4. TANDEM AND BIFACIAL SOLAR CELLS

A highly active research field in light management for PV addresses tandem and bifacial solar cells. In a single-junction solar cell, photons with an energy larger than the bandgap are incompletely used because the photogenerated carriers quickly thermalize to the band edge. Tandem solar cells can partially overcome these losses by absorbing the high-energy light in a semiconductor with a larger bandgap, thereby producing a larger potential from these photons. Tandem solar cells based on silicon require a material with a bandgap in the range of 1.6-1.8 eV, and materials most commonly employed are perovskite and III-V semiconductors. Si/perovskite tandems have recently demonstrated a power conversion efficiencies of 29.2%, well above the record for Si-only cells (26.7%), while Si/III-V tandems have reached 33%.[76,77]

The key light management challenge in tandem solar cells is the proper division of the low- and high-energy band of the solar spectrum over the two semiconductors. Specially structured layers placed in between the two cells should be designed to selectively reflect transmitted high-energy light back towards the top cell, while transmitting low-energy light towards the bottom cell. In the two-terminal series-connected tandem configuration, charge carriers must freely transmit through this spectrum splitting interlayer. At the same time, these layers should be optically structured to control light scattering and trapping into the desired layers. Designing and fabricating such optically rough, and electrically flat layers challenges both photonic design and fabrication. A light management challenge in Si/III-V tandems is to further reduce parasitic losses in the metal backreflector.

The market share of bifacial silicon solar panels, that absorb light from both front- and rearside is rapidly increasing, expected to exceed 70% within the next 10 years.[1] Many bifacial field test sites have now been operating for multiple years and increased annual energy yields compared to traditional monofacial designs typically range between 10-20%.[78,79] Vertically placed bifacial cells also can help with “peak shaving” the excess yield in the middle of the day, since they produce most of their power in the morning and evening, improving synchronization with electricity demand.[78,79] The light spectrum incident on a bifacial panel is strongly dependent on the light scattering spectrum and albedo of the background, and diffuse light incoupling plays an important role. Light scattering sheets placed on the ground could be designed with optimized angular and spectral light scattering properties to maximize the light coupling into both sides of the panel. In parallel, plant biologists could help design directional and spectrally selective light scattering properties of plants placed in PV fields with bifacial panels.

Shading effects, that reduce efficiency in series-connected tandem solar cells play a more dominant role in bifacial panels. Bifacial tandem solar cells should be operated in a three- or four-terminal configuration to avoid these problems. Adaptive photonics may be realized that control the division of light over the tandem top- and bottom cells during the day, thereby optimizing current matching for bifacial tandems. Such adaptive scattering would need to be light-, current-, or voltage-induced and wavelength-selective. Micro/nanomechanical actuators or orientation-control of scatterers embedded in liquid crystals could provide such futuristic properties.

6. SPECTRAL SHAPING

An alternative strategy towards better use of the high-energy part of the solar spectrum is to down-convert the high-energy light so that it matches the semiconductor bandgap energy. One solution that is being extensively investigated uses singlet fission[80] in an organic layer or quantum cutting in an inorganic material[81–83] to convert a high-energy photon into two photons at half the initial energy each.[84] The photonics challenge is then to preferentially direct these photons towards the underlying solar cell.[85] When the organic down-conversion layers are placed on a silicon cell, the refractive index contrast already causes direction of a major fraction of the downconverted light into the silicon solar cell. One could imagine a photonic layer that locally enhances the optical density of states near the solar cell to further increase the fraction of light emitted towards the cell. Alternatively, the down-conversion emitter could be integrated with anisotropic nanophotonic light scatterers to create directional emission. Such geometries could be created as down-conversion "foils" that can be placed as an add-on onto existing PV panels. Anything placed onto existing solar panels should not introduce additional losses, so ultra-high optical quality of the organic layers and guaranteed long lifetime are essential to make this a viable downconversion solution.

In the past, trivalent lanthanide ions have also been considered as down- and up-conversion systems. A key problem with these ions is their low optical absorption and emission cross sections, due to the parity-forbidden nature of the intra-4*f* transitions.[86] This leads to the requirement of very high lanthanide concentrations as well as undesired optical saturation effects due to the long PL lifetime of the lanthanides. A recent development, that does point towards a potential practical application of lanthanide downconversion is in Yb-doped perovskites, where recombination of a single exciton in a high-bandgap perovskite leads to the creation of two excited Yb^{3+} ions.[81–83,87] Challenges for integration into solar cells include the power saturation that reduces the efficiency at high incident flux, and the need for directional photon emission towards the solar cell. Photonic engineering may be used to accelerate the Yb^{3+} decay and tailor its angular emission to enable practical application as a solar cell add-on. Another, more moderate way to improve solar cell efficiency is by downshifting a single UV photon to a single lower-energy photon above the bandgap.[88] While this is not a photon multiplication effect, it can be advantageous, as solar cells typically have higher internal quantum efficiency for carrier collection in the near-infrared than in the visible spectral range.

Upconversion of low-energy photons that are transmitted through the solar cell is a complementary way to better convert the solar spectrum. Absorbing two photons and emitting one photon of roughly twice the energy can be achieved, for example, by quantum-dot sensitized triplet exciton annihilation.[89] These systems have to be optically thick for light with photon energy below the solar cell bandgap, and at the same time transparent for the upconverted emission. So far, quantum-dot sensitized schemes have suffered from strong self-absorption above the bandgap, but photonic methods to tailor the modal field distribution in the films may help alleviate this problem. Alternatively, the upconverted excitation could be transferred to the cell by direct electrical coupling of the upconverted excitation state to the solar cell.

Luminescent solar concentrators (LSCs) are a special type of spectral shaper where transparent or colorful windows absorb, downshift and redirect part of the solar spectrum to small solar cells placed at the window sides, or invisibly integrated in the window itself.[90,91] The small required solar cell area enables the use of expensive but more efficient GaAs solar cells, and concentration effects enhance the conversion efficiency. So far efficiencies are low, and improving LSCs requires control of the directional emission from the downconverting quantum dots or dyes so that emission within the escape cone is minimized. This can be realized using anisotropically shaped Mie resonators that are coupled to the emitters.

6. LARGE-SCALE INTEGRATION OF PV: BUILDING AND LANDSCAPE INTEGRATION

As PV systems are becoming deployed at a very large scale it is essential to integrate them into our landscape, built environment and infrastructure in a practical and appealing way. In rural landscape integration, bifacial solar panels that collect sunlight from both sides are becoming increasingly popular. Here it becomes increasingly relevant to engineer the photonics of metal contact grids to minimize contact shading, see also Section 4.[44,45]

In building integration, ideally, a PV panel is directly used as a building material, thereby reducing the effective costs of the PV system. Building materials range from transparent windows, to colorful facades and curvy tile roofs. The development of colored PV panels merged with appealing architectural designs is essential to take advantage of the large area potential offered by building infrastructure. Resonant light scattering by dielectric Mie scatterers creates well-defined colors which maintain high cell efficiency.[92–96] Integration of PV systems in roads and other infrastructure will also require the development of efficient flexible and light-weight PV systems, as do roofs that are not designed to carry the heavy weights of conventional panels, such as the corrugated sheet metal roofs that are common in many parts of the world. Efficient light coupling and trapping in ultrathin foils (Section 2) is key to reach this goal.

Electrochromic windows are electrically powered devices with controlled color tinting and enable control of building temperature by controlling the transmission of infrared light. These windows could further enhance the application space for PV technologies. Integrating PVs in these designs would create self-powered or even power-generating smart windows for a range of new applications. Adaptive light management geometries would enable optimized harvesting of the proper bands of the solar spectrum at the right moment of the day.

7. MODELING, ADAPTIVE PV, LARGE-SCALE FABRICATION

Traditionally, nanophotonic designs are guided through finite-difference time-domain (FDTD) computer simulations that solve Maxwell's equations for complex 3D geometries. As optical constants are typically accurately known, these simulations give precise guidelines to optimize nanoscale cell architectures. Integrating electronic simulations at the same length scales then helps optimize the electronic properties of the cell. Initial designs are often made based on educated guesses and simulations then optimize the structure to perfection. Inverse design, or machine learning, is emerging as a powerful technique to find solutions for problems that are hard to solve by intuition, and genetic algorithms help find solutions to these problems much more effectively than by brute-force random optimization. Inverse design takes advantage of the time reciprocity that is inherent to Maxwell's equations. For example, instead of calculating the full angular emission pattern of a lensing structure in every step of an optimization for highly directional emission, it is possible to simply maximize the electric field intensity at the emitter position for a normal incident plane wave, drastically increasing simulation efficiency and enabling discovery of non-intuitive geometries.[66] It may even be possible to take such an inverse design principle a step further where the desired angular emission pattern is used as an input to synthesize emitters in the required positions, providing a simple self-aligned process.

While solar cells are typically optimized for efficiency under a single standard spectrum (*e.g.* AM1.5G), in practice the spectrum changes significantly over timescales from minutes to months.[97] Variations in spectrum, intensity, temperature and angular distribution of the incoming radiation all affect the energy yield and appearance, but current photovoltaic modules cannot adapt to these varying conditions for practical, cost and aesthetic reasons.

For example, tandem solar cells would benefit from a spectrum-splitting layer between the top- and bottom-cells of which the optical characteristics are tuned depending on the solar spectrum as it varies over time. Micromechanical actuation may be designed to achieve such adaptive spectrum splitting, powered by thermo-electric effects or by power from the solar cell itself. Alternatively, light, temperature or electric field-induced phase changes, photo-induced ionic migration (such as seen in halide perovskite solar cells) or programmable refractive index changes in optical resonators can create adaptive functionalities. The challenge here is to find a robust working mechanism across a range of temperature and illumination conditions that does not cause other module degradation mechanisms over a period of several decades.

The output of bifacial solar cells would also strongly increase if the reflection of light from the ground were optimized as the sun moves along the hemisphere over the course of the day. Here too, adaptive directional emitters can be envisioned to perform this task, potentially self-aligned by photostriction or thermo-mechanical forces. Such adaptive methods are inspired by concepts from natural phototropism, which then in turn inspires the question if plants could also be tailored, through phenotypical plasticity, to adopt adaptive scattering properties that are optimized for light capture in bifacial solar fields throughout the day.[98–101]

Adaptive photonics are also interesting to achieve tracking of cells that require control over the incident direction of light, such as concentrating photovoltaics, and geometries for directional emission such as described in Section 3. Recent developments in the design of micro-mechanical metamaterials and robotics may further inspire progress in the field of adaptive optics for PV.[98–102]

Finally, to reach the full potential of PV for large-scale power generation it is essential to develop fabrication strategies that allow multi-scale control (from nano to millimeter) with large throughput and at low cost. For Si PV technology these concepts are all well advanced to a very high level. Yet, for most other PV materials these developments have just begun. New fabrication strategies based on soft-nanoimprint and roll-to-roll concepts to fabricate nanostructures are particularly interesting for the large-scale PV requirements.[103,104] Solution or vapor processing such as for perovskites and quantum dots can easily be integrated into roll-to-roll processes. In all these developments, achieving low cost is essential. While prototyping photonic structures is often expensive, several routes exist to mass-produce them cheaply. These include self-assembly, roll-to-roll nanoimprint lithography, or even injection molding, as is done for DVDs, for example.[104–108] If similar manufacturing techniques could be developed at low costs for photonic PV structures, they could help create the more than ten-fold enhancement in PV manufacturing capacity that is needed to create PV technology on earth at the tens of TW level.

Table I. Photonics opportunities for photovoltaics with improved efficiency. Present status, promise for applications, photonics opportunities, and corresponding improvements for different solar cell materials (green) and various generic PV concepts (blue).

Material/ concept	Status/promise/applic ations	Photonics opportunity	Dominant improvement
Si	Large scale, lowest-cost PV for solar fields, rooftop integration	• Effectively transparent contacts to reduce absorption and reflection at fingers and busbars • Bandgap emission rate enhancement to enhance PL emission quantum efficiency • Nanopatterned selective contact layers to reduce parasitic UV absorption • Micropatterend surfaces with tailored infrared emission for radiative cooling	Current Voltage Current Temperature
III-V	Ultrahigh efficiency, light-weight, flexible applications in space, automobiles, etc., ..	• Enhanced light absorption in active layer • Reduced parasitic absorption in buffer layers	Current Current
Thin film PV: perovskite, organic, CIGS, CdTe, quantum dot, CZTS, ...	Light-weight, flexible roll-to-roll fabrication for building, landscape, and infrastructure integration	• Light incoupling and light trapping to enhance near-band edge absorption • Bandgap emission rate enhancement to enhance PL emission quantum efficiency	Current Voltage
Bifacial solar cells	High energy yield, landscape integration, daily solar power peak shaving	• Effectively transparent contacts on both sides to reduce absorption and reflection at fingers and busbars • Optimize light scattering spectrum and albedo from the environment • Adaptive light scattering from ground plane	Current Current Current
Tandem solar cells	Ultra-high efficiency for solar fields, building, landscape, and infrastructure integration, space, automobiles, etc.	• Electrically flat/optically textured spectrum splitting layers to optimize layer integration for tandem solar cells based on Si • Low-absorbing structured light trapping backreflectors/contacts for Si bottom cells • Bandgap emission rate enhancement to enhance PL emission quantum efficiency at two wavelengths • Adaptive current matching	Current Current Current Current

Down- and up-conversion layers, LSCs	High efficiency, flexible integration on all PV modules and foils	• Directional emission towards the solar cell • Emission rate enhancement to avoid saturation for lanthanide-doped systems • Minimize self-absorption of up- and down-converted light in organic layers • Direct energy transfer from upconverted dopants to semiconductor	Current Current Current Current
Flexible, colored, tailored PV	Building, landscape and infrastructure integration, automobiles, etc.	• Color and directional emission control for better integration (buildings, landscapes), architectural designs • Adaptive optical properties for tailored architectural designs, electrochromic windows.	Integration, architecture Integration, architecture

CONCLUSION

Solar energy by photovoltaic conversion has to be employed on a massive scale to succeed in a transition to a renewable energy supply. Photonic design can aid this transition by improving existing solar cells closer to their efficiency limit, eliminating losses from incomplete absorption or non-radiative recombination. Beyond the state-of-the-art solar cells, photonic design plays a crucial role in next-generation photovoltaics based on tandem solar cells. Photonic structures and metasurfaces help to increase absorption, ease fabrication, and improve efficiency. Looking further ahead, solar cells could look radically different when photonic design is used in a smart way: photonic layers that prepare a spectrum that is better matched to the solar cell absorber, solar cells that reflect light with a tailored color spectrum while maintaining a majority of the power conversion efficiency, or solar cells that adapt themselves to solar irradiation conditions. These concepts may be futuristic now, but with the rapid advance of photonic theory and experimental techniques in recent years there is good reason to believe that some of these designs will accelerate the massive upscaling and (invisible) integration of photovoltaics into our society.

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Notes

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REFERENCES

1. *International Technology Roadmap for Photovoltaic (ITRPV): 2019 Results, 11th Edition*. https://itrpv.vdma.org/ (2020).

2. Energia - The International Network on Gender & Energy. Renewables 2020 Global Status Report (REN21) Released. *REN21's Renewables 2020 Global Status Report (GSR)* (2020).

3. Climate Investment Funds website. https://www.climateinvestmentfunds.org/CIF10/india/bhadla.

4. Sivaram, V. & Kann, S. Solar power needs a more ambitious cost target. *Nat. Energy* **1**, 16036 (2016).

5. Jaeger-Waldau, A. PV Status Report 2019. *Publ. Off. Eur. Union* (2019) doi:10.2760/326629.

6. ESMAP, SOLARGIS, WB & IFC. Global Solar Atlas. *Global Solar Atlas* (2019).

7. Peters, I. M., Rodriguez Gallegos, C. D., Sofia, S. E. & Buonassisi, T. The Value of Efficiency in Photovoltaics. *Joule* **3**, 2732–2747 (2019).

8. See webpage for updated efficiencies and cell parameters for record solar cells of different materials. http://www.lmpv.nl/sq/.

9. Shockley, W. & Queisser, H. Detailed balance limit of efficiency of p-n junction solar cells. *J. Appl. Phys.* **1640**, 510–519 (1961).

10. van der Burgt, J. S. & Garnett, E. C. Nanophotonic Emission Control for Improved Photovoltaic Efficiency. *ACS Photonics* **7**, 1589–1602 (2020).

11. Richter, J. M. *et al.* Enhancing photoluminescence yields in lead halide perovskites by photon recycling and light out-coupling. *Nat. Commun.* **7**, 13941 (2016).

12. Pazos-Outon, L. M. *et al.* Photon recycling in lead iodide perovskite solar cells. *Science* **351**, 1430–1433 (2016).

13. Araújo, G. L. & Martí, A. Absolute limiting efficiencies for photovoltaic energy conversion. *Sol. Energy Mater. Sol. Cells* **33**, 213–240 (1994).

14. Peters, M., Goldschmidt, J. C. & Bläsi, B. Angular confinement and concentration in photovoltaic converters. *Sol. Energy Mater. Sol. Cells* **94**, 1393–1398 (2010).

15. Green, M. A. *et al.* Solar cell efficiency tables (Version 55). *Prog. Photovoltaics Res. Appl.* **28**, 3–15 (2020).

16. Nair, G., Chang, L.-Y., Geyer, S. M. & Bawendi, M. G. Perspective on the Prospects of a Carrier Multiplication Nanocrystal Solar Cell. *Nano Lett.* **11**, 2145–2151 (2011).

17. Brongersma, M. L., Cui, Y. & Fan, S. Light management for photovoltaics using high-index nanostructures. *Nat. Mater.* **13**, 451–460 (2014).

18. Jeong, S., McGehee, M. D. & Cui, Y. All-back-contact ultra-thin silicon nanocone solar cells with 13.7% power conversion efficiency. *Nat. Commun.* **4**, 2950 (2013).

19. Savin, H. *et al.* Black silicon solar cells with interdigitated back-contacts achieve 22.1% efficiency. *Nat. Nanotechnol.* **10**, 624–628 (2015).

20. Spinelli, P., Verschuuren, M. A. & Polman, A. Broadband omnidirectional antireflection coating based on subwavelength surface Mie resonators. *Nat. Commun.* **3**, 692 (2012).

21. Zhu, J. *et al.* Optical absorption enhancement in amorphous silicon nanowire and nanocone arrays. *Nano Lett.* **9**, 279–82 (2009).

22. Spinelli, P., Macco, B., Verschuuren, M. A., Kessels, W. M. M. & Polman, A. Al_2O_3/TiO_2 nano-pattern antireflection coating with ultralow surface recombination. *Appl. Phys. Lett.* **102**, 233902 (2013).

23. Ferry, V. E. *et al.* Optimized Spatial Correlations for Broadband Light Trapping Nanopatterns in High Efficiency Ultrathin Film a-Si:H Solar Cells. *Nano Lett.* **11**, 4239–4245 (2011).

24. Martins, E. R. *et al.* Deterministic quasi-random nanostructures for photon control. *Nat. Commun.* **4**, 2665 (2013).

25. Oskooi, A. *et al.* Partially disordered photonic-crystal thin films for enhanced and robust photovoltaics. *Appl. Phys. Lett.* **100**, 181110 (2012).

26. van Lare, M.-C. & Polman, A. Optimized Scattering Power Spectral Density of Photovoltaic Light-Trapping Patterns. *ACS Photonics* **2**, 822–831 (2015).

27. Yu, S. *et al.* Design of Non-Deterministic Quasi-random Nanophotonic Structures Using Fourier Space Representations. *Sci. Rep.* **7**, 3752 (2017).

28. Pratesi, F., Burresi, M., Riboli, F., Vynck, K. & Wiersma, D. S. Disordered photonic structures for light harvesting in solar cells. *Opt. Express* **21**, A460 (2013).

29. Bozzola, A., Liscidini, M. & Andreani, L. C. Broadband light trapping with disordered photonic structures in thin-film silicon solar cells. *Prog. Photovoltaics Res. Appl.* **22**, 1237–1245 (2013).

30. N. Tavakoli, R. J. Spalding, P. Koppejan, Gkantzounis, C. Wan, R. Rohrich, R. Sapienza, F. Koenderink, M. Florescu, E. Alarcon-Llado. Over 65% sunlight absorption in 1µm thick Si slab. *Submitted* (2020).

31. Yablonovitch, E. & Cody, G. Intensity enhancement in textured optical sheets for solar cells. *IEEE Trans. Electron Devices* **ED-29**, 300–305 (1982).

32. Garnett, E. & Yang, P. Light Trapping in Silicon Nanowire Solar Cells. *Nano Lett.* **10**, 1082–1087 (2010).

33. Yu, Z., Raman, A. & Fan, S. Fundamental limit of nanophotonic light trapping in solar cells. *Proc. Natl. Acad. Sci.* **107**, 17491–17496 (2010).

34. Yu, Z., Raman, A. & Fan, S. Fundamental limit of light trapping in grating structures. *Opt. Express* **18**, A366 (2010).

35. Wang, K. X. *et al.* Light trapping in photonic crystals. *Energy Environ. Sci.* **7**, 2725 (2014).

36. Green, M. A. Time-Asymmetric Photovoltaics. *Nano Lett.* **12**, 5985–5988 (2012).

37. Divitt, S. & Novotny, L. Spatial coherence of sunlight and its implications for light management in photovoltaics. *Optica* **2**, 95 (2015).

38. Atwater, H. A. & Polman, A. Plasmonics for improved photovoltaic devices. *Nat. Mater.* **9**, 205–213 (2010).

39. Wu, H. *et al.* A transparent electrode based on a metal nanotrough network. *Nat. Nanotechnol.* **8**, 421–425 (2013).

40. Lee, J.-Y., Connor, S. T., Cui, Y. & Peumans, P. Solution-Processed Metal Nanowire Mesh Transparent Electrodes. *Nano Lett.* **8**, 689–692 (2008).

41. Garnett, E. C. *et al.* Self-limited plasmonic welding of silver nanowire junctions. *Nat. Mater.* **11**, 241–249 (2012).

42. Adhyaksa, G. W. P., Johlin, E. & Garnett, E. C. Nanoscale Back Contact Perovskite Solar Cell Design for Improved Tandem Efficiency. *Nano Lett.* **17**, 5206–5212 (2017).

43. Yin, G. *et al.* Optoelectronic Enhancement of Ultrathin CuIn 1-x Ga x Se 2 Solar Cells by Nanophotonic Contacts. *Adv. Opt. Mater.* **5**, 1600637 (2017).

44. Saive, R. *et al.* Effectively Transparent Front Contacts for Optoelectronic Devices. *Adv. Opt. Mater.* **4**, 1470–1474 (2016).

45. Schumann, M. F. *et al.* All-Angle Invisibility Cloaking of Contact Fingers on Solar Cells by Refractive Free-Form Surfaces. *Adv. Opt. Mater.* **5**, 1700164 (2017).

46. Bullock, J. *et al.* Efficient silicon solar cells with dopant-free asymmetric heterocontacts. *Nat. Energy* **1**, 15031 (2016).

47. Yoshikawa, K. *et al.* Silicon heterojunction solar cell with interdigitated back contacts for

a photoconversion efficiency over 26%. *Nat. Energy* **2**, 17032 (2017).

48. Wang, W. *et al.* Device Characteristics of CZTSSe Thin-Film Solar Cells with 12.6% Efficiency. *Adv. Energy Mater.* **4**, 1301465 (2014).

49. Tang, R. *et al.* Hydrothermal deposition of antimony selenosulfide thin films enables solar cells with 10% efficiency. *Nat. Energy* **5**, 587–595 (2020).

50. Leijtens, T., Bush, K. A., Prasanna, R. & McGehee, M. D. Opportunities and challenges for tandem solar cells using metal halide perovskite semiconductors. *Nat. Energy* **3**, 828–838 (2018).

51. Yang, Y. *et al.* Low surface recombination velocity in solution-grown CH3NH3PbBr3 perovskite single crystal. *Nat. Commun.* **6**, 7961 (2015).

52. Khoram, P., Oener, S. Z., Zhang, Q., Fan, Z. & Garnett, E. C. Surface recombination velocity of methylammonium lead bromide nanowires in anodic aluminium oxide templates. *Mol. Syst. Des. Eng.* **3**, 723–728 (2018).

53. Wang, H. *et al.* Nanoimprinted perovskite metasurface for enhanced photoluminescence. *Opt. Express* **25**, A1162 (2017).

54. Wang, H. *et al.* Nanoimprinted Perovskite Nanograting Photodetector with Improved Efficiency. *ACS Nano* **10**, 10921–10928 (2016).

55. Schmager, R. *et al.* Light coupling to quasi-guided modes in nanoimprinted perovskite solar cells. *Sol. Energy Mater. Sol. Cells* **201**, 110080 (2019).

56. Kim, H. *et al.* Optically Pumped Lasing from Hybrid Perovskite Light-Emitting Diodes. *Adv. Opt. Mater.* **8**, 1901297 (2020).

57. Brittman, S. *et al.* Controlling crystallization to imprint nanophotonic structures into halide perovskites using soft lithography. *J. Mater. Chem. C* **5**, 8301–8307 (2017).

58. Brenner, P. *et al.* Highly stable solution processed metal-halide perovskite lasers on nanoimprinted distributed feedback structures. *Appl. Phys. Lett.* **109**, 141106 (2016).

59. Lou, S., Xuan, T. & Wang, J. Stability: A desiderated problem for the lead halide perovskites. *Opt. Mater. X* **1**, 100023 (2019).

60. Kirchartz, T., Staub, F. & Rau, U. Impact of Photon Recycling on the Open-Circuit Voltage of Metal Halide Perovskite Solar Cells. *ACS Energy Lett.* **1**, 731–739 (2016).

61. Rau, U., Paetzold, U. W. & Kirchartz, T. Thermodynamics of light management in photovoltaic devices. *Phys. Rev. B* **90**, 035211 (2014).

62. Kosten, E. D., Kayes, B. M. & Atwater, H. A. Experimental demonstration of enhanced photon recycling in angle-restricted GaAs solar cells. *Energy Environ. Sci.* **7**, 1907 (2014).

63. Mann, S. A., Grote, R. R., Osgood, R. M., Alù, A. & Garnett, E. C. Opportunities and Limitations for Nanophotonic Structures To Exceed the Shockley–Queisser Limit. *ACS Nano* **10**, 8620–8631 (2016).

64. Cui, Y. *et al.* Boosting Solar Cell Photovoltage via Nanophotonic Engineering. *Nano Lett.* **16**, 6467–6471 (2016).

65. Zhang, D. *et al.* Increasing Photoluminescence Quantum Yield by Nanophotonic Design of Quantum-Confined Halide Perovskite Nanowire Arrays. *Nano Lett.* **19**, 2850–2857 (2019).

66. Johlin, E., Mann, S. A., Kasture, S., Koenderink, A. F. & Garnett, E. C. Broadband highly directive 3D nanophotonic lenses. *Nat. Commun.* **9**, 4742 (2018).

67. Catalanotti, S. *et al.* The radiative cooling of selective surfaces. *Sol. Energy* **17**, 83–89 (1975).

68. Zhu, L., Raman, A., Wang, K. X., Anoma, M. A. & Fan, S. Radiative cooling of solar cells. *Optica* **1**, 32 (2014).

69. Raman, A. P., Anoma, M. A., Zhu, L., Rephaeli, E. & Fan, S. Passive radiative cooling below ambient air temperature under direct sunlight. *Nature* **515**, 540–544 (2014).

70. Hsu, P.-C. *et al.* Radiative human body cooling by nanoporous polyethylene textile. *Science* **353**, 1019–1023 (2016).

71. Rephaeli, E., Raman, A. & Fan, S. Ultrabroadband Photonic Structures To Achieve High-Performance Daytime Radiative Cooling. *Nano Lett.* **13**, 1457–1461 (2013).

72. Hossain, M. M., Jia, B. & Gu, M. A Metamaterial Emitter for Highly Efficient Radiative Cooling. *Adv. Opt. Mater.* **3**, 1047–1051 (2015).

73. Li, T. *et al.* A radiative cooling structural material. *Science* **364**, 760–763 (2019).

74. Green, M. A. *Solar Cells: Operating Principles, Technology and System Applications*. (University of New South Wales, 1982).

75. Silverman, T. J. *et al.* Reducing Operating Temperature in Photovoltaic Modules. *IEEE J. Photovoltaics* **8**, 532–540 (2018).

76. Jošt, M., Kegelmann, L., Korte, L. & Albrecht, S. Monolithic Perovskite Tandem Solar Cells: A Review of the Present Status and Advanced Characterization Methods Toward 30% Efficiency. *Adv. Energy Mater.* **10**, 1904102 (2020).

77. Cariou, R. *et al.* III–V-on-silicon solar cells reaching 33% photoconversion efficiency in two-terminal configuration. *Nat. Energy* **3**, 326–333 (2018).

78. Liang, T. S. *et al.* A review of crystalline silicon bifacial photovoltaic performance characterisation and simulation. *Energy Environ. Sci.* **12**, 116–148 (2019).

79. Guerrero-Lemus, R., Vega, R., Kim, T., Kimm, A. & Shephard, L. E. Bifacial solar photovoltaics – A technology review. *Renew. Sustain. Energy Rev.* **60**, 1533–1549 (2016).

80. Smith, M. B. & Michl, J. Singlet Fission. *Chem. Rev.* **110**, 6891–6936 (2010).

81. Luo, X., Ding, T., Liu, X., Liu, Y. & Wu, K. Quantum-Cutting Luminescent Solar Concentrators Using Ytterbium-Doped Perovskite Nanocrystals. *Nano Lett.* **19**, 338–341 (2019).

82. Kroupa, D. M., Roh, J. Y., Milstein, T. J., Creutz, S. E. & Gamelin, D. R. Quantum-Cutting Ytterbium-Doped $CsPb(Cl_{1-x}Br_x)_3$ Perovskite Thin Films with Photoluminescence Quantum Yields over 190%. *ACS Energy Lett.* **3**, 2390–2395 (2018).

83. Milstein, T. J. *et al.* Anion Exchange and the Quantum-Cutting Energy Threshold in Ytterbium-Doped $CsPb(Cl_{1-x}Br_x)_3$ Perovskite Nanocrystals. *Nano Lett.* **19**, 1931–1937 (2019).

84. Rao, A. & Friend, R. H. Harnessing singlet exciton fission to break the Shockley–Queisser limit. *Nat. Rev. Mater.* **2**, 17063 (2017).

85. Futscher, M. H., Rao, A. & Ehrler, B. The Potential of Singlet Fission Photon Multipliers as an Alternative to Silicon-Based Tandem Solar Cells. *ACS Energy Lett.* **3**, 2587–2592 (2018).

86. Polman, A. Erbium implanted thin film photonic materials. *J. Appl. Phys.* **82**, 1–39 (1997).

87. Pan, G. *et al.* Doping Lanthanide into Perovskite Nanocrystals: Highly Improved and Expanded Optical Properties. *Nano Lett.* **17**, 8005–8011 (2017).

88. van Sark, W. G. J. H. M., Meijerink, A., Schropp, R. E. I., van Roosmalen, J. A. M. & Lysen, E. H. Modeling improvement of spectral response of solar cells by deployment of spectral converters containing semiconductor nanocrystals. *Semiconductors* **38**, 962–969 (2004).

89. Tayebjee, M. J. Y., McCamey, D. R. & Schmidt, T. W. Beyond Shockley–Queisser: Molecular Approaches to High-Efficiency Photovoltaics. *J. Phys. Chem. Lett.* **6**, 2367–2378 (2015).

90. Meinardi, F., Bruni, F. & Brovelli, S. Luminescent solar concentrators for building-integrated photovoltaics. *Nat. Rev. Mater.* **2**, 17072 (2017).

91. McKenna, B. & Evans, R. C. Towards Efficient Spectral Converters through Materials

Design for Luminescent Solar Devices. *Adv. Mater.* **29**, 1606491 (2017).

92. Neder, V., Luxembourg, S. L. & Polman, A. Efficient colored silicon solar modules using integrated resonant dielectric nanoscatterers. *Appl. Phys. Lett.* **111**, 073902 (2017).

93. Uleman, F., Neder, V., Cordaro, A., Alù, A. & Polman, A. Resonant Metagratings for Spectral and Angular Control of Light for Colored Rooftop Photovoltaics. *ACS Appl. Energy Mater.* **3**, 3150–3156 (2020).

94. Wen, L. *et al.* Multifunctional Silicon Optoelectronics Integrated with Plasmonic Scattering Color. *ACS Nano* **10**, 11076–11086 (2016).

95. Park, H. J., Xu, T., Lee, J. Y., Ledbetter, A. & Guo, L. J. Photonic Color Filters Integrated with Organic Solar Cells for Energy Harvesting. *ACS Nano* **5**, 7055–7060 (2011).

96. Soman, A. & Antony, A. Colored solar cells with spectrally selective photonic crystal reflectors for application in building integrated photovoltaics. *Sol. Energy* **181**, 1–8 (2019).

97. U.S. National Renewable Energy Laboratory. National Solar Radiation Database (NSRDB). *NREL.gov* (2017).

98. Zagolla, V., Dominé, D., Tremblay, E. & Moser, C. Self-tracking solar concentrator with an acceptance angle of 32°. *Opt. Express* **22**, A1880 (2014).

99. Baker, K. A., Karp, J. H., Tremblay, E. J., Hallas, J. M. & Ford, J. E. Reactive self-tracking solar concentrators: concept, design, and initial materials characterization. *Appl. Opt.* **51**, 1086 (2012).

100. Tremblay, E. J., Loterie, D. & Moser, C. Thermal phase change actuator for self-tracking solar concentration. *Opt. Express* **20**, A964 (2012).

101. Qian, X. *et al.* Artificial phototropism for omnidirectional tracking and harvesting of light. *Nat. Nanotechnol.* **14**, 1048–1055 (2019).

102. Svetozarevic, B. *et al.* Dynamic photovoltaic building envelopes for adaptive energy and comfort management. *Nat. Energy* **4**, 671–682 (2019).

103. Verschuuren, M. A., Knight, M. W., Megens, M. & Polman, A. Nanoscale spatial limitations of large-area substrate conformal imprint lithography. *Nanotechnology* **30**, 345301 (2019).

104. Agrawal, H. & Garnett, E. C. Nanocube Imprint Lithography. *ACS Nano* acsnano.0c04793 (2020) doi:10.1021/acsnano.0c04793.

105. Boles, M. A., Engel, M. & Talapin, D. V. Self-Assembly of Colloidal Nanocrystals: From

Intricate Structures to Functional Materials. *Chem. Rev.* **116**, 11220–11289 (2016).

106. Zhang, J., Li, Y., Zhang, X. & Yang, B. Colloidal Self-Assembly Meets Nanofabrication: From Two-Dimensional Colloidal Crystals to Nanostructure Arrays. *Adv. Mater.* **22**, 4249–4269 (2010).

107. Xia, Y. & Whitesides, G. M. Soft Lithography. *Annu. Rev. Mater. Sci.* **28**, 153–184 (1998).

108. Ahn, S. H. & Guo, L. J. High-Speed Roll-to-Roll Nanoimprint Lithography on Flexible Plastic Substrates. *Adv. Mater.* **20**, 2044–2049 (2008).