Photon Management in Perovskite Solar Cells

Thomas Kirchartz[1,2*]

[1]IEK5-Photovoltaik, Forschungszentrum Jülich, 52425 Jülich, Germany

[2]Faculty of Engineering and CENIDE, University of Duisburg-Essen, Carl-Benz-Str. 199, 47057 Duisburg, Germany

Metal-halide perovskites as absorber materials for solar cells have captured the attention of an increasing number of scientists over the last years. As is common for most photovoltaic technologies, early stages of solar cell development were dedicated to the understanding and tuning of the properties of materials and interfaces in the perovskite-based solar cells to ensure efficient charge separation and suppress un-wanted recombination channels.[1,2] However, when photovoltaic technologies become more mature, the design of optical properties of materials, interfaces and complete device stacks becomes increasingly important in order to come even closer to the efficiency limits dictated by thermodynamics.[3] This viewpoint is dedicated to briefly discuss the how photons can be absorbed more efficiently, can be reused, distributed between different cells in tandem devices and finally be used as a means to characterize the electronic properties of materials and interfaces. More detailed discussions of these topics can then be found in a series of perspective articles on topics related to the optical properties and the optical design of perovskite-based solar cells that are introduced in this viewpoint.

How important optical design is for a specific solar cell technology mostly depends on the optical properties of the chosen absorber layer and in particularly on its complex refractive index. The real part of the refractive index affects how important antireflective coatings will be, because the reflectance of an interface will depend on the refractive index contrast. In addition, the refractive index will control the amount of total internal reflection and thereby the pathlength enhancement for weakly absorbed light achievable by light scattering. In the limit of ray-optics, the average pathlength enhancement will ideally be given by the so-called Yablonovitch limit[4,5] of $4n^2$, where n is the real part of the refractive index. This means that if weakly absorbed light is scattered isotropically, the path of the photons in the absorber will have an average length of $4n^2d$, where d is the thickness of the absorber layer. For materials like Si $4n^2 \approx 50$,[6] implying that light absorption could ideally be ~25 times better than in a comparable cell with a perfect back mirror but no means of scattering the light and using total internal reflection. In Si, the high refractive index and the fairly low absorption coefficient above the indirect band gap ensure that optical design is of huge importance for photovoltaic functionality. Managing the photons is a necessity to decouple optical thickness (up to $4n^2d$) from electrical and mechanical thickness (d) of the Si wafer. In direct semiconductors and in semiconductors with a lower real part of the refractive index, such as lead-halide perovskites, both the necessity and the potential of photon management are reduced. However, there is still potential left, for instance for light scattering to improve photocurrent generation.[7] In order to bring any photovoltaic technology to market, the technological potential for improving efficiency has to be exploited. Therefore, it is understandable why managing photons has so far not received much attention in the context of lead-halide perovskites but also why it has to receive more attention in the future. This is true in particular given how good the electronic properties and the suppression of non-radiative recombination have already become. The open-circuit voltage has already approached levels [8-12] where further progress also requires efforts in optical design rather than only efforts to reduce non-radiative recombination in the bulk and at interfaces. These high open-circuit voltages imply that the luminescence quantum efficiencies of the solar cells are high, which in turn makes photons a useful tool for analyzing the properties of the absorber material, the interfaces towards the contacts and the whole device.

Photon Recycling. The efficiency limit of single junction solar cells is given by the Shockley-Queisser (SQ) model[13] which assumes among other things that all recombination is radiative. Heat is still created by thermalization of charge carriers to the band edges and by thermalization during charge collection but not by recombination. Silicon solar cells cannot fully approach this limit,[14] because of Auger recombination which converts the energy of recombining electron-hole pairs into kinetic energy of a third carrier and subsequently in thermal energy, when the accelerated charge carrier thermalizes back to the band edge. Semiconductors like GaAs or the lead-halide perovskites have much higher radiative recombination coefficients as compared to Si (roughly 5 orders of magnitude higher [15-18]) leading to the situation that Auger recombination is much less relevant at charge carrier densities encountered during photovoltaic operation at intensities around one sun.[19] Thus, GaAs and lead-halide perovskites should in principle be able to come much closer to the limit imposed by the SQ model up to a point where the internal luminescence quantum efficiencies approach unity.[20] In this situation, recombination mostly leads to the creation of a photon as opposed to the creation of heat. However, without any optical measures, the external luminescence quantum efficiency will be substantially lower than the internal one. Only a small percentage of photons will be created with an angle that allows them to avoid total internal reflection. In addition, the photons will be created with photon energies where the absorber and the contact layers absorb light. To approach the thermodynamic limit, photons can be emitted or reabsorbed in which case the process starts again. This full cycle of emission and reabsorption is called *photon recycling* and becomes relevant in all semiconductors where a high percentage of recombination is radiative. However, if the photons are absorbed in one of the contact layers, typically the photon energy is lost. This may be due to the fact that the contact layers are not of sufficient electronic quality to harvest the created electron-hole pair or because the photon absorption happens without the creation of electron hole pairs but by free carrier absorption. These lost photons that are parasitically absorbed will become a highly significant loss process once a solar cell approaches the limit given by the SQ model.[21-24] This loss has had practical significance for GaAs-based solar cells where efficiency improvements were possible in the past by removing the GaAs substrate and replacing it with a mirror. This led to strongly reduced parasitic absorption and additional improvements in efficiency were then achieved by further optimization of the mirror reflectivity. Given that photons may bounce back and forth in a thin solar cell many times, approaching a mirror reflectivity of one close to the radiative limit leads to substantial improvements in the open-circuit voltage of the solar cell.[25] The perspective by Pazos-Outon et al.[20] uses simulations to illustrate how perovskite solar cells are benefitting from photon recycling and how efficiency and optical design are interrelated. Figure 1a shows how the external luminescence quantum efficiency $Q_{e,lum}$ depends on the internal luminescence quantum efficiency $Q_{i,lum}$ and the probability p_a of parasitic absorption using the equation

$$Q_{e,lum} = \frac{p_e Q_{i,lum}}{1 - p_r Q_{i,lum}}. \quad (1)$$

Here, p_e is the outcoupling efficiency, p_r is the probability of reabsorption and the sum of the three probabilities is equal to 1 ($p_e + p_a + p_r = 1$). The denominator in eq. (1) represents the effect of photon recycling, i.e. the higher $p_r Q_{i,lum}$, the more $Q_{e,lum}$ is increased by photon recycling. We note that due to the low outcoupling efficiency $p_e = 6\%$ assumed (that is representative of flat perovskite solar cells[21]) an increase in parasitic absorption (y-axis) leads to a substantial decrease of $Q_{e,lum}$. Because[26]

$$qV_{oc} = qV_{oc,rad} + kT \ln\left\{\frac{p_e Q_i^{lum}}{1 - p_r Q_i^{lum}}\right\}. \quad (2)$$

this loss in $Q_{e,lum}$ directly leads to a loss in open-circuit voltage. Here, $V_{oc,rad}$ is the radiative limit to the open-circuit voltage, which is about 1.325 V for $CH_3NH_3PbI_3$.[9]

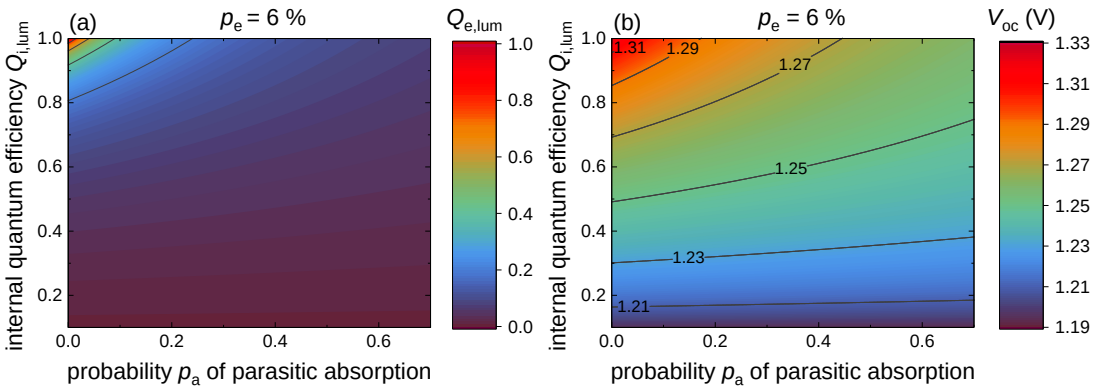

Figure 1: Illustration of the importance of parasitic absorption for improving (a) the external luminescence quantum efficiency and (b) the open-circuit voltage V_{oc} of a perovskite solar cell. Simulations are performed using Eq. (1) and using an outcoupling efficiency of p_e = 6%, which is representative for a flat perovskite solar cell with a reflecting mirror on one side. 94% of the photons generated by radiative recombination are kept inside the device by total internal reflection and are then absorbed eventually either by the absorber (with probability $p_r = 1 - p_e - p_a$) itself leading to photon recycling or by any of the contact layers leading to parasitic absorption with probability p_a.

Managing Photons in Tandem Solar Cells. While optical management is important in single junction solar cells to maximize absorption in the absorber layer and minimize parasitic absorption in the contact layers, this challenge becomes even more important in tandem solar cells where photons have to be absorbed in different absorber layers based on their wavelength. In research, various tandem solar cell geometries are investigated (see Figure 2) that either have two terminals (as a normal single junction solar cell), four terminals (two independent solar cells coupled optically) or even three terminals (grown monolithically on top of each other but still two separate circuits for top and bottom solar cells). While four terminal tandem solar cells are easiest to implement they require one opaque contact usually made of metal and three transparent contacts. In contrast, two terminal tandem cells would only require one opaque and one transparent contact. Thus, the choice between two or four terminal tandem cell relies heavily on how much the parasitic absorption loss caused by the two additional transparent contacts is outweighing the additional degrees of freedom in design (e.g. choice of band gap or absorber layer thickness) that the four-terminal approach offers.

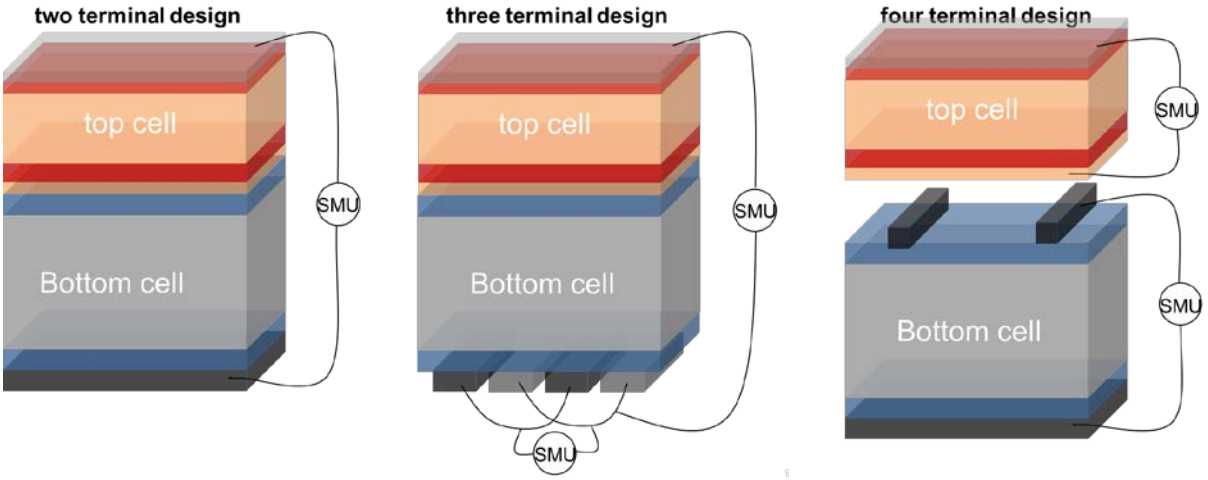

Figure 2: Schematic comparison of the three different geometries for tandem solar cells, two-terminal, three terminal and four-terminal tandem solar cells. SMU stands for source measure unit and indicates where in a normal current-voltage measurement, voltage and current would have to be measured. Each contact that is connected to the SMU and is not the opaque back contact, has to be realized by a transparent contact, based e.g. on transparent conductive oxides and/or metal grids, that leads to parasitic absorption. In case of the two- and three terminal designs, one transparent contact is needed, while the four-terminal design requires three transparent contacts.

Two terminal tandems therefore seem to have an intrinsic advantage when it comes to higher efficiencies. However, there are additional optical challenges related to two terminal tandem cells in particular that have to be tackled. First, in a two-terminal tandem the challenge is not only to absorb as much light as possible in either the bottom or the top part of the tandem cell but to absorb exactly equal photon densities in each subcell. This is necessary because otherwise the current densities at the maximum power point are likely imbalanced. Due to the series connection between the two subcells, the lower of the two current densities will limit the current density of the tandem solar cell. This can become challenging in real outdoor applications, in particular if the absorptance of the subcells become dependent on the angle of the incident light which naturally varies over the course of a day. This effect is negligible in fully textured tandem solar cells with light scattering interfaces such as amorphous Si/microcrystalline Si devices[27] but it may become an issue in flat perovskite-based tandem solar cells. Hence, measures to circumvent any strong angular or spectral dependence of two-terminal tandem cells, i.e. based on intermediate reflectors[28,29] or scattering elements, might become increasingly important.

The perspective by Jacobs et al.[30] discusses optical aspects of tandem solar cell design in detail with a specific focus on parasitic absorption in transparent contact layers, the effect of texturing the silicon wafer, and the impact of diffuse and direct illumination on two- vs. four terminal tandem concepts.

Optical Measurement Techniques. Due to their frequently quoted and discussed defect tolerance,[31,32] lead-halide perovskite layers and devices are often exceptionally well luminescing and therefore measurement techniques that use photons emitted by luminescence have always been particular popular in the community. There are typically two major variants of photoluminescence measurements done, namely steady state measurements and transient measurements. In steady state measurements, based on the spectrum or just the intensity (often as a function of position on the cell) of the luminescence, information on the internal quasi-Fermi level splitting is derived.[33] If done on a cell, these measurements deliver an optically measured analogue of the open-circuit voltage and are therefore sometimes called implied V_{oc} measurements. If done on layer stacks that lack certain layers

used in a solar cell, conclusions can be drawn on the loss in open circuit voltage caused by the addition of certain interfaces that are necessary to extract charge but often lead to additional recombination.[34-36]

Transient photoluminescence measurements are helpful to identify the type of recombination present. While tr-PL has proven to be successful in determining recombination coefficients for different types of recombination (radiative, defect assisted and Auger recombination) in perovskite films on glass,[37] problems start once charge extracting interfaces are applied to the sample.[38] While steady-state photoluminescence has proven to be useful at quantifying the losses due to interfacial recombination, photoluminescence transients have so far been substantially more challenging to interpret.[39]

While different types of photoluminescence measurements have been instrumental to better understand recombination, transport of charge carriers cannot easily be measured by photoluminescence. The perspective by Braly et al.[40] therefore combines steady-state photoluminescence with photoconductivity measurements to determine the photoluminescence quantum yield and the diffusion length at the same time. This combination of methods is fast and simple allowing transport and recombination to be tracked as a function of photoactivation (initial improvement of photovoltaic performance often observed in perovskite-based films and devices) or during longer term degradation.

In order to fully understand optical characterization methods, measuring and understanding the shape of the absorption onset including the effect of excitions, i.e. Coulomb-interactions between electrons and holes is crucial. While the exciton binding energies in lead-halide perovskites are smaller than the thermal energy *kT*, excitonic effects still affect the shape of the absorption coefficient around the band edge, in particular for higher band gap perovskites. In addition, excitonic effects control the signal measured in transient absorption and transient reflectance measurements. The perspective by Chen et al.[41] gives an overview of excitonic effects in methylammonium lead halide perovskites with a particular focus on their influence on the shape of the absorption coefficient as a function of energy and on their influence on transient optical characterization methods and on the recombination coefficients for radiative and Auger recombination.

Outlook. There are still plenty of open scientific and practical technological questions related to photon management and optical characterization techniques that have to be addressed in the context of lead-halide perovskites in order to maximize the efficiency of these devices and provide the basis for future commercial exploitation of the technology. The topics discussed in this viewpoint and in the perspectives that have been introduced here will hopefully provide a basis for understanding the current understanding and technological state-of-the-art of these methods and technologies and will be useful for interested readers from different disciplines.

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