

Trends in Perovskite Solar Cells and Optoelectronics: Status of Research and Applications from the PSCO conference

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Organic metal halide perovskites¹ are the subject of intensive research efforts, due to the impressive performance achieved in photovoltaic and optoelectronic devices.²⁻³ The attraction towards these materials, hereafter simply perovskites, arises from a multitude of reasons. First, they show optimal primary optoelectronic properties such as direct band-gaps, long carriers diffusion lengths, low exciton binding energies, whose incarnation is the remarkable power conversion efficiency, over 22%, that these materials already delivered in optimized photovoltaic devices. This comes together with ease of processing via solution or vapour phase (or a combination of the two) techniques, low cost and abundance of base materials, low temperature of processing leading to versatility in terms of what substrates can be used and ability to process multiple layers on-top of each other.

With the huge interest raised by perovskites, many conferences traditionally dedicated to materials science or photovoltaics are hosting an increasing number of “perovskite sessions”. As researchers working in the perovskite field from its inception, we launched a new conference entirely dedicated to perovskite solar cells and optoelectronics (PSCO) to gather the forefront of perovskite research into a unique forum. The second edition of the PSCO conference has been organized in Genova, Italy, from September 26-28 2016, following the successful launch of the conference in September 2015 in Lausanne, Switzerland. The 2017 edition will be held in Oxford, UK, from 18-20 September 2017. The 2016 conference hosted 19 invited, 52 contributed and 10 sponsor presentations, with about 400 participants. 12 “best poster” and “best contributed talk” awards were offered by major journals and publishers, including three ACS Energy Letters awards for best talks. The 2016 conference was sponsored by various industrial partners and featured by the “Destiny” Marie Curie Initial Training Network, with the organization of a dedicated seminar.

Given the rapid evolution of the field, we sought of taking advantage of the large number of invited and contributed talks, spanning diverse topics, but sharing the broad perovskite denominator. We summarize here a selection of the most relevant research topics highlighted in the PSCO

conference, which may serve as a short survey to set the state-of-the-art and the future direction of the field.

The PSCO field has been naturally driven by the applicative potential of perovskites in solar cells and optoelectronic devices. This strong application-oriented research is now reflected into a huge effort towards transitioning from laboratory cells to modules, with strong emphasis on materials engineering towards efficient and durable devices. As researchers working in academic institutions, it is rewarding to see, however, how basic studies are still attracting huge attention. Basic research is continuously casting new light on the fundamental materials properties, which are likely at key of perovskite's success, and it is constantly feeding the knowledge required to further progress towards working scalable devices.

The hot topics discussed at the PSCO conference indeed reflect this interdisciplinary/multicultural scenario and can be (somehow arbitrarily) grouped into the following three categories.

1) Fundamental understanding of perovskites

Perovskites share unconventional electronic properties related to the presence of ns^2 metals such as Sn(II) or Pb(II) which can be understood in view of structure-property relationships.⁴ In particular, there are synthetic requirements under which a halide perovskite can exist, with synthetic conditions defining the properties and structural stability of the ensuing perovskites. Using the structural features as guides, the origin of the optical and electronic properties of similar perovskites have been clarified, casting the basis for tuning and controlling them. In particular, it is now clear how the local arrangement of MX_6 octahedra plays a fundamental role in determining the perovskite band gap and transport properties, with less tilted, cubic-like perovskites showing a red-shifted band-gap and decreased carrier effective masses. This is due to the interplay of structural and electronic degrees of freedom, in particular the spin-orbit coupling associated to the heavy metal, whose magnitude is

modulated by the MX_6 octahedral tilting.⁵ The evolution of the band-gap in mixed Sn/Pb perovskites can be understood in a similar fashion, although the picture is complicated by the different stabilization of various phases with intermediate compositions. Mixed Sn/Pb perovskites are under the spotlight, because they offer an additional mean to modulate the materials band-gap in the near-IR region and to mitigate the lead content in devices, opening the pathway towards perovskite/perovskite tandem cells.⁶

As solution processed materials, perovskites have an inherently “*soft*” structure, related to both the inorganic lead-halide matrix and the mobile (organic) cations, such as cesium, methylammonium (MA) or formamidinium (FA). Notably, the organic and inorganic perovskite components are interdependent, such that a change in the orientation of the organic cations also implies structural changes in the inorganic sub-lattice.⁷ The implications of such soft dynamical behavior on the optoelectronic properties of perovskites has stimulated several hypotheses to explain their spectacularly long diffusion lengths and low recombination rates, likely representing the main reason for the success of this class of materials. Most notably, many dynamical times scales exist in perovskites, spanning from ultra-fast fs scale to ms or even minutes or hours.⁸⁻⁹

Among the astonishing properties of perovskites there are a set of apparent inconsistencies in these materials, when related to traditional semiconductors: i) their long carrier lifetimes coupled to modest mobility; ii) their apparently low trap density despite the low temperature solution preparation; and iii) the apparently flexible inorganic lattice still associated to rather sharp diffraction and optical absorption onsets.¹⁰⁻¹¹ Given the deformable nature of the crystal lattice in these materials, displacements of ions from their equilibrium positions will likely interact with the charge carriers, influencing the electronic and transport properties of the material. Thus, investigating the nature of carrier-phonon interactions in pristine perovskites is crucial to understand the carriers dynamics and transport processes. A mechanism of charge carrier protection has been proposed, such that the nascent charge carriers are screened by large-polaron formation on time scales of a few hundred fs.

This results in long-lived energetic electrons with a surprisingly long lifetime on the order of 100 ps, which is exceedingly longer than that in conventional semiconductors.¹² The formation of protected energetic carriers requires the competitive ultrafast dynamics of large-polaron formation in the dynamically disordered hybrid perovskite structure. In contrast to that of energetic carriers, the protection of long-lived band-edge carriers does not seem to require the presence of organic cations and it is thus likely an intrinsic property of the soft perovskite inorganic lattice.¹³

Given the simple processability at relatively low temperature, one can expect an intrinsic level of structural/chemical disorder of the perovskite, which will inevitably result in the formation of defects. Defect properties of a photovoltaic absorber (in solar cells) or of a recombination emitter (in light-emitting diodes) are among the most critical properties determining the final performance of devices using that material. The defect properties can be modelled by first-principles electronic structure calculations based on Density Functional Theory (DFT).¹⁴ The amenable information to DFT modeling are the defect formation energies, which define the stable states of charge of a given defect and their relative concentrations when the material is grown in equilibrium with its precursors; and the defect transition energies, whose position with respect to the band edges determines whether a defect will likely trap charge carriers or not. Both quantities are sensitive to the level of DFT employed in the calculations. An approximate DFT and/or the lack of spin-orbit coupling may hamper the quantitative description of the band energetics of the perovskite,¹⁵ which is fundamental to determine whether a defect charge transition level will fall into the material band-gap. With their low migration barriers¹⁶ and relatively low formation energies,¹⁷ iodine defects are the natural candidates to explain the rich photophysics of perovskites, including those related to light-induced material transformation. Recent electrochemical experiments have shown that iodine vacancies are the majority migrating species in lead-iodide perovskites and likely those responsible of their ionic conductivity.

The lack of understanding and control of the potential inhomogeneities related to defects and to different processing of the perovskite thin films may restrict the efficiencies of optoelectronic devices and their lab-to-lab reproducibility, which is a key step to make a perovskite based technology market appealing. The use of both confocal fluorescence lifetime imaging microscopy and scanning-probe microscopy studies may help to elucidate the origins of this local heterogeneity in terms of composition and surface defects. In particular, local photoluminescence, local electroluminescence, local conductivity and local surface potential show a correlation with the perovskite film processing methods. Specific processing additives and illumination may serve to alter the distribution of surface states. Also, the “photo-cleaning” effects repeatedly observed in perovskites can be associated with photo-induced ion migration, as indicated by time-of-flight mass spectrometry and DFT calculations.¹⁸⁻¹⁹ By using different processing methods and additives, it is possible to tailor the materials properties, and obtain carrier lifetimes and PL intensities in thin films approaching those in single crystals, opening the way for these materials to approach the Shockley-Queisser limit.

Apart from the remarkable bulk properties of this class of semiconductors, it has been realized that the interfaces between the perovskite and the selective contacts are a key aspect of the operation and also show dynamic interactions, possibly underlying the hysteresis of perovskite solar cells. Bisquert and co-workers have made a distinction between capacitive and non-capacitive hysteresis, providing an interpretation of capacitances as a function of frequency, both in dark and under light, finding that capacitance is primarily impacted by the contacts to the perovskite layer, and due to ion migration.²⁰ Capacitive hysteresis originates at the perovskite/TiO₂ interface and is related to the ability to accommodate both ionic and electronic charges in a reversible way. Non-capacitive hysteresis is on the other hand related to modification of the interface or contact reactivity caused by perovskite ionic motion and it is connected to device degradation.²¹

2) Materials processing and synthesis

A huge effort has been invested in various labs to enhance the materials properties leading both to efficient and stable devices, along with chasing effective strategies for perovskite deposition over larger areas. A winning strategy to achieve both targets seems the choice of mixed cation/mixed halide perovskites, where formamidinium, methylammonium and cesium cations are alloyed within iodine/bromine perovskites in variable percentage providing outstanding materials properties which translate into reproducible devices with 20% efficiency.²² A notable result is the report of a 22%-efficient solar cell with an external electroluminescence of 4%, and an outstanding open-circuit voltage of 1.24 V at a band gap of 1.63, eV entailing one of the smallest loss-in-potential (0.39 V) ever measured for any solar cell material. A further engineering of the perovskite deposition step (i.e. replacing an antisolvent step with a flash vacuum treatment) was shown to lead to 1 cm² devices with 19.6% efficiency.²³ Preliminary data suggested encouraging results, with retention of 95% of the initial stability over 500 hours at 85 °C under maximum power point.

A recognized issue in fabrication and long-term device use is perovskite extreme water sensitivity. 2D perovskites have been discussed as a way to impart moisture-resistance to the material through hydrophobic organic layers, while the inorganic layers could be further optimized to generate photocurrent. To overcome the use of lead into potentially widespread devices double perovskites could be considered, which are more accommodating for metal substitution than conventional perovskites and demonstrated promising photophysical properties. As an example, bismuth double perovskites such as Cs₂AgBiBr₆ further motivate the study of this class of materials for optoelectronic applications.²⁴

Along with the synthesis of bulk perovskites single crystals and thin films, nanocrystals based on the same materials have become increasingly popular²⁵ since they allow for further tuning of the materials optoelectronic properties by e.g. size or shape selection and by using optimized synthetic strategies, such as anion exchange, allow to obtain high purity and high crystalline materials. Notably, these strategies have led to the first demonstration of a fast, room-temperature synthesis of inks based

on CsPbBr₃ perovskite nanocrystals using short, low boiling-point ligands and environmentally friendly solvents. Requiring no lengthy post-synthesis treatments, the inks are directly used to fabricate conductive thin films of high optoelectronic quality. The robustness of such properties was further demonstrated by the fabrication of the first perovskite nanocrystal-based solar cell.²⁶

3) Device optimization, processing and large area / modules

Despite being a relatively young field, the industrial potential of devices based on metal-halide perovskites has motivated intense research efforts towards the realization of scalable photovoltaic solutions. Large focus has been posed on the scalability towards modules with monolithically interconnected cells, with emphasis on the required deposition processes and potential applications. Following the optimization of laboratory-scale devices based on a conventional TiO₂-based architecture with power conversion efficiency ~18% for active area <1 cm², modules with an aperture area of 156 cm² have been demonstrated. A standard interconnection geometry with three patterning steps was used to contact the sub-cells in series, ensuring low loss of the aperture area and low power loss due to contact sheet resistance, bringing the module close to its maximum performance. Focus to close the performance gap further between single cell and large area module should therefore be put on the homogeneous deposition of all layers on such large areas. Notably, this work shows the potential to use perovskite photovoltaics in high-performance stand-alone applications.

Another potential application area is created by stacking perovskite modules on top of other thin-film technologies like CIGS or crystalline silicon. The benefits of the perovskite cells in such a concept are both optical and electrical. On one side, the optical absorption edge is sharp compared to other thin film photovoltaic technologies, whilst the parasitic absorption below the edge is very low. On the other side, the ratio of open-circuit voltage to optical gap is high, a necessity for stacked or tandem concepts. A 4-terminal stacked concept was shown to be superior over the 2-terminal tandem approach. Tandem solar cells between perovskite and conventional inorganic semiconductors are seen as a major deployment area in the industrial uptake of perovskite solar cells.

In a notable effort to integrate new materials with large-scale module fabrication, graphene and related 2D materials have been used to engineer the interfaces and to improve both efficiency and stability of perovskite cells and modules.²⁷ Lithium-Neutralized Graphene Oxide (GO-Li) has been used at the interface between TiO₂ and Perovskite, allowing fast electron injection from the perovskite to the titania, eventually increasing the integrated short-circuit current. A similar effect has been observed for graphene in the mesoporous-TiO₂, with graphene-based composites being successfully employed in large area perovskite modules achieving 12.6% efficiency on an overall active area of 50.6 cm².

An alternative to conventional TiO₂-based perovskite solar cells is the so-called inverted p-i-n architecture.²⁸ These devices can be fabricated either via the conventional solution-based techniques or by using vacuum based preparation methods. These vapour-deposited p-i-n perovskite solar cells exhibit high power-conversion efficiencies, topping at 20% in a planar device.²⁹ Additionally, a multi-layer stack of different vacuum-deposited perovskite layers was used to generate tandem structures with high efficiency and open circuit voltage above 2.3 V.

High efficiency planar p-i-n solar cells were also obtained through a new two-step solution process to synthesize high quality perovskite film at room temperature. Combining with smooth PEDOT:PSS hole transport layer and high mobility electron transport (PC₇₁BM) layer, the planar solar cell achieves power conversion efficiency above 20%.³⁰ This champion cell shows no current hysteresis both in voltage scan directions and rates. The growth and purity of perovskite film can be manipulated individually, providing a simple and reproducible way to fabricate perovskite based solar cells *via* low temperature solution process in an ambient atmosphere. An important characteristic of p-i-n inverted architectures is that of being transferable on flexible (plastic) substrates. This is an important aspect that could be exploited for the uptake of perovskite solar cells and optoelectronic devices in the wide consumable electronics market.

Concluding, the PSCO conference has demonstrated that metal-halide perovskites have made giants steps towards possible market products, with several companies working on dedicated materials, instrumentation and scale-up industrial aspects. This strong application-oriented direction is however not an obstacle to basic research. On the contrary, it gives the opportunity to researchers to, in the near term, translate their discoveries into potential solutions to product improvement. The key to the future market success of perovskites is likely the realization of stable materials and devices, while continuing the investigation of the unusual optoelectronic materials properties of this astonishing class compounds.



Figure 1. From left to right: Filippo De Angelis, Henry Snaith and Annamaria Petrozza (PSCO organizers) wearing the ACS Energy Letters conference t-shirt.



Figure 2. From left to right: Alison Walker, coordinator of “Destiny”, Md. K. Nazeeruddin and Henry Snaith (PSCO organizers) during the conference awards ceremony.



Figure 3. From left to right: Henry Snaith, Md. K. Nazeeruddin, Annamaria Petrozza and Filippo De Angelis with the PSCO cake.

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