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Photonic Strategies for Photovoltaics: New Advances Beyond Optics

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Abstract: Photonic structures with dimensions comparable to sunlight wavelengths are now regarded as the preferential solutions to enhance the efficiency of photovoltaic devices via light trapping. These photonic microstructures operate in the wave-optics regime. Electromagnetic models were thus developed to determine the optimal parameters for application in the front contact of different photovoltaic technologies, namely in thin-film solar cells based in silicon or perovskite materials. In this way, distinct photonic cell architectures were obtained, showing efficiency improvements up to 50% with respect to planar reference solar cells. The results demonstrate that the advantages in the application of the photonic structures are not just limited to optical gains related with light absorption enhancement, but also enable other important benefits such as: electrical gains due to the improvement of the front contact conductance, and better environmental/outdoor performance due to an advanced micro-structured encapsulation that even allows self-cleaning properties.

Key words: solar energy, thin-film photovoltaics, photonics, light management

1. Introduction

Recent R&D trends in photovoltaics (PV) have focused in novel concepts capable of reaching higher efficiencies while reducing fabrication costs. This can be accomplished by exploring new materials, developing cheaper techniques and reducing the solar cells' absorber thickness. Thin-film PV is a promising avenue to improve the market-share of solar electricity, as it allows lighter, flexible solar cells while reducing raw material usage and production costs. As thickness decreases, light-management plays a critical role in ensuring that thin absorbers are capable of converting as much energy as possible, bringing forth high efficiencies.

Conventional light-trapping approaches are based on textured surfaces which provide: 1) anti-reflection, via geometrical refractive-index matching caused by the front facets, improving the short-wavelength (above absorber bandgap) photocurrent; and 2) light scattering which increases the longer-wavelength (near-bandgap) absorption via optical path-length amplification and coupling with confined waveguided modes. However, the main drawback is that texturing increases roughness (hence defect density) in the PV material, which deteriorates its electrical transport via recombination.

Alternative strategies (Fig. 1) have been investigated by the authors [1-6] and several other groups worldwide [7-10]. Such solutions are based in advanced nano/micro-photonic effects that can provide stronger interaction between sunlight and thin-film PV materials, achieving broadband harvesting of sunlight in ultra-thin absorbers without affecting their electrical performance. In this way, optically thicker but physically thinner devices are capable of delivering high electrical power (enhanced efficiency), with

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prolonged stability, while being supported on mechanically-bendable inexpensive substrates. These remarkable features open the way for the market implementation of solar electricity in a broad range of portable consumer applications.

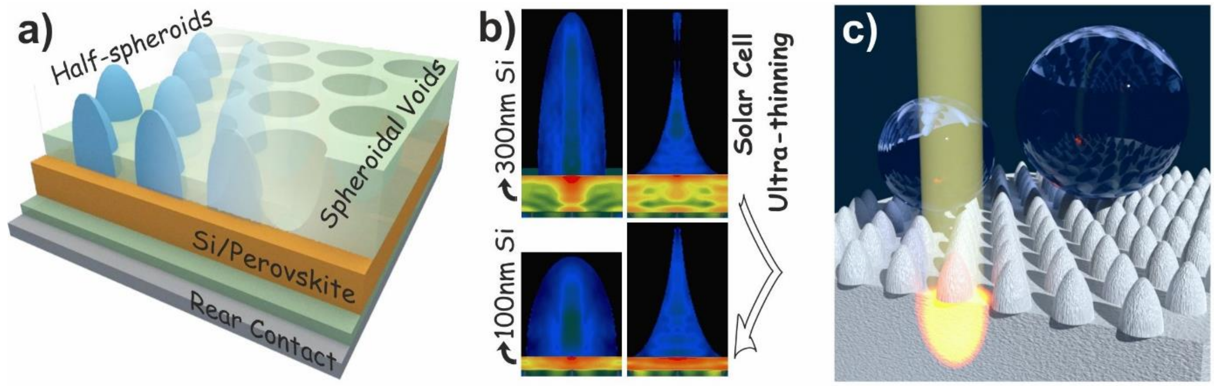

Fig. 1 a) Illustration of two types of optimized geometries for light trapping in silicon or perovskite solar cells. Such optical schemes allow reducing the thickness of the absorber material and, at the same time, improve the photocurrent and efficiency of the cells (see profile of carriers photo-generation in b) [11] c) Representation of photonic microstructures formed by colloidal lithography on the surface of an encapsulating film, which makes it possible to repel water droplets and, therefore, enables a useful functionality of self-cleaning the cell surface.

1.1 The New Age of Bendable Photovoltaics for Consumer-oriented Products

Thin-film PV has been discussed as a potential high-impact market, which could follow the progress and growth of mobile electronics [12]. The need for reliable sources of power that can be integrated into mobile electronics by being flexible/bendable makes the research and development of such PV technologies enticing. It is expected that, in this decade, the PV market will witness an astonishing expansion in distributed autonomous systems supported on a variety of platforms that can be rigid but curved (e.g., in buildings [13] and vehicles [14]) or flexible (e.g., polymers [15], paper [16], metal foils [15]); enabling a plethora of consumer-oriented applications (Fig. 2). In addition, flexible solar cells offer important cost-saving solutions to industry since they: 1) have reduced material costs due to their small thickness, 2) can be integrated on a variety of inexpensive substrates, 3) can be manufactured in large-area roll-to-roll processes, and 4) have lower installation costs. However, this can only be achieved with thin-film cells able to operate on curved/bendable substrates with high stabilized efficiency, far beyond the state-of-art, which is presently one of the main goals of the PV community. In addition, since flexible solar cells operate under bending, their optical response for different incidence angles becomes paramount.

Despite astonishing advances in the past decade in flexible thin-film PV, such this technology is still far from achieving its full market potential. This is mainly due to the fact that only modest efficiencies have been reached with thin solar cells (10-15%), compared to those (20-25%) of conventional rigid c-Si wafer-based cells. Optics is the chief reason for such reduced performance, as the thin (< 10 μm) PV devices required for mechanical bendability exhibit reduced light absorption (particularly in near-infrared, NIR) in comparison with the thicker (100-200 μm) wafer-based cells. Light-trapping methods have risen as an effective approach to this problem [5, 11].

Light-trapping (LT) enables physically-thin but optically-thick devices, implying cheaper and faster fabrication, lightweight and improved flexibility. It can also provide roll-to-roll manufacturing and application in the aforementioned bendable substrates aimed for consumer products: building-integrated PV (BIPV), wearables, smart-packing, mobility sector, etc. (Fig. 2). Additionally, thickness reduction can lead to higher open-circuit voltage due to lower bulk-recombination.

Fig. 2 Light-trapping (LT) can improve the performance of thin-film PV, enabling high-power-density sources at affordable cost for citizens; while their bendability and lightweight allows conformal integration on any platform of daily consumer applications. This will catalyze an explosion of new commercial products such as: facile and more aesthetic implementation of PV modules on curved building surfaces, or on the coverage of vehicles (e.g., for electric cars) [14], wearable PV, solar-powered mobile electronics (IoT [17], medical diagnostic portable devices [18], smart-packaging [19]), among many other applications.

1.2 Light-Trapping by Wave-optical Photonic Structures

High refractive index particles and periodic structures with wavelength-scale dimensions are considered the preferable LT solutions for integration in the illuminated face of PV devices [8, 20-22], due to their remarkable ability for light in-coupling (via geometric anti-reflection) plus scattering towards the absorber medium. Such optical strategy in the regime of wave-optics requires pyramidal-like features made of low-loss materials with strong light interaction capability, i.e., micro-structured dielectric media with high n and small k in its complex refractive index ($N = n+ik$). This can be realized, for instance, with photonic-structured metal-oxides (e.g., TiO_2 or a transparent conductive oxide, TCO) front-patterned on the cells. The authors have shown that this allows strong light harvesting (up to ~50% photocurrent gain shown with TiO_2 structures on thin-film Si cells) [23, 24] without affecting the solar cells' electrical performance (i.e., similar efficiency gain), among other advantages as described below:

1) Optical — their combined anti-reflection and forward-scattering effects can pronouncedly increase absorption at UV-visible and near-infrared (NIR) wavelengths, respectively; resulting in broadband photocurrent enhancement superior to conventional LT approaches (e.g., texturing). The scattered light can be particularly trapped within the cell when coupled with resonant wave-guide modes related with the structure's geometry, leading to pronounced peaks in the absorption spectrum. For periodic structures, this increase can even surpass the theoretical limit — the *Tiedje-Yablonovitch* limit — for specific

wavelengths related with the LT structure's pitch [25-27].

2) Electrical — can be incorporated on top of planar thin-film cells without increasing the surface area and defect density in the cell layers, therefore not degrading the electric performance (charge transport) via carrier recombination/trapping.
3) Stability — the photonic micro-structures on the cell front can lead to improved operational stability of the devices by two means: 1) blocking the harmful higher-energy photons of UV radiation, which degrade the materials' electronic properties; and 2) contributing to the device encapsulation as they can exhibit super-hydrophobic water-repellent properties [28] that allow a practical self-cleaning functionality. Both mechanisms are particularly advantageous to assist in the stability of less matured PV technologies such as perovskite-based solar cells [29, 30].
4) Industrial attractiveness — the dielectric-based materials of the photonic structures can be made of Earth-abundant metal-oxide compounds or even of the same material of the cells' absorber (as in texturing). In this respect, they can have an extremely lower material cost than the plasmonic-based LT approaches that rely on expensive noble metals. Besides, the wave-optical patterning can be performed via low-cost and highly scalable soft-lithography processes such as the colloidal-lithography methods described further below [21, 31, 32].

2. Results and Discussion

2.1 Electromagnetic Modelling

The resonant nature of wave-optical structures substantially limits the parameter space in which their optical effects can provide exceptional improvements in the absorption of the PV layers, across the relevant sunlight spectrum. The photonic front features need to provide a gradually varying effective refractive-index, from air towards the PV absorber, to minimize reflection. At the same time, their geometry must interact with the incoming light to produce strong scattered fields preferentially directed into the higher-index absorber. Therefore, prior to experimental implementation, we performed a rigorous screening based on modelling, to understand the influence that the parameters of the LT structures have on such effects, and then to appropriately search for the best parameter set that allows the highest photocurrent enhancement in the devices [11, 23, 33].

The simulation of the electromagnetic field propagation in these structures with wavelength-scale dimensions was performed employing an exact 3D numerical formalism (finite-difference time domain, FDTD) method. This is one of the preferential approaches to solve electromagnetic problems in the wave-optics regime, due to its conceptual simplicity and versatility. Furthermore, since it is a time-domain method, the solutions can cover a wide frequency range with a single simulation run [10, 20, 27, 34, 35]. When coupled with a "*smart-search*" optimization algorithm, FDTD allows an effective screening of the sets of parameters (i.e., material, geometry) of the LT features that maximize the broadband light absorption in the PV layer (e.g., Si in the case of Fig. 3a-c) while minimizing the optical losses (i.e., reflection and absorption occurring in the other materials of the device). For that, a *particle swarm* optimization algorithm [36] was employed to iteratively look for the optimal parameters of different LT designs, using the photocurrent density produced by the cell as figure-of-merit. As an example, this enabled the determination of the optimized LT structures for thin-film Si-based solar cells, presented in Fig. 3a-c [11, 23], composed of semi-prolate voids in a layer of either TiO_2 or AZO material.

The reasons for the choices of these materials are the following. TiO_2 was identified as a preferential medium for front dielectric LT structures on thin-film PV, due to its relatively high real part (n) of the refractive-index, which favors anti-reflection and scattering effects, and low imaginary part (*k*) in the visible-NIR wavelength range which implies reduced parasitic absorption [8, 37, 38]. Despite these advantageous optical properties, the poor electric conductivity of TiO_2 makes it difficult to engineer a good-performing front contact for the LT-enhanced devices. Therefore, despite the lower *n* and higher *k* of the most relevant transparent conductive oxide (TCO) materials, they can constitute preferential photonic materials for the cells' front in view of the electrical performance. In particular, the abundance, non-toxicity and excellent transparency and electrical properties of ZnO-based TCOs (e.g., AZO) emphasizes their importance as the best alternatives to the standard TCOs based on ITO [39-41].

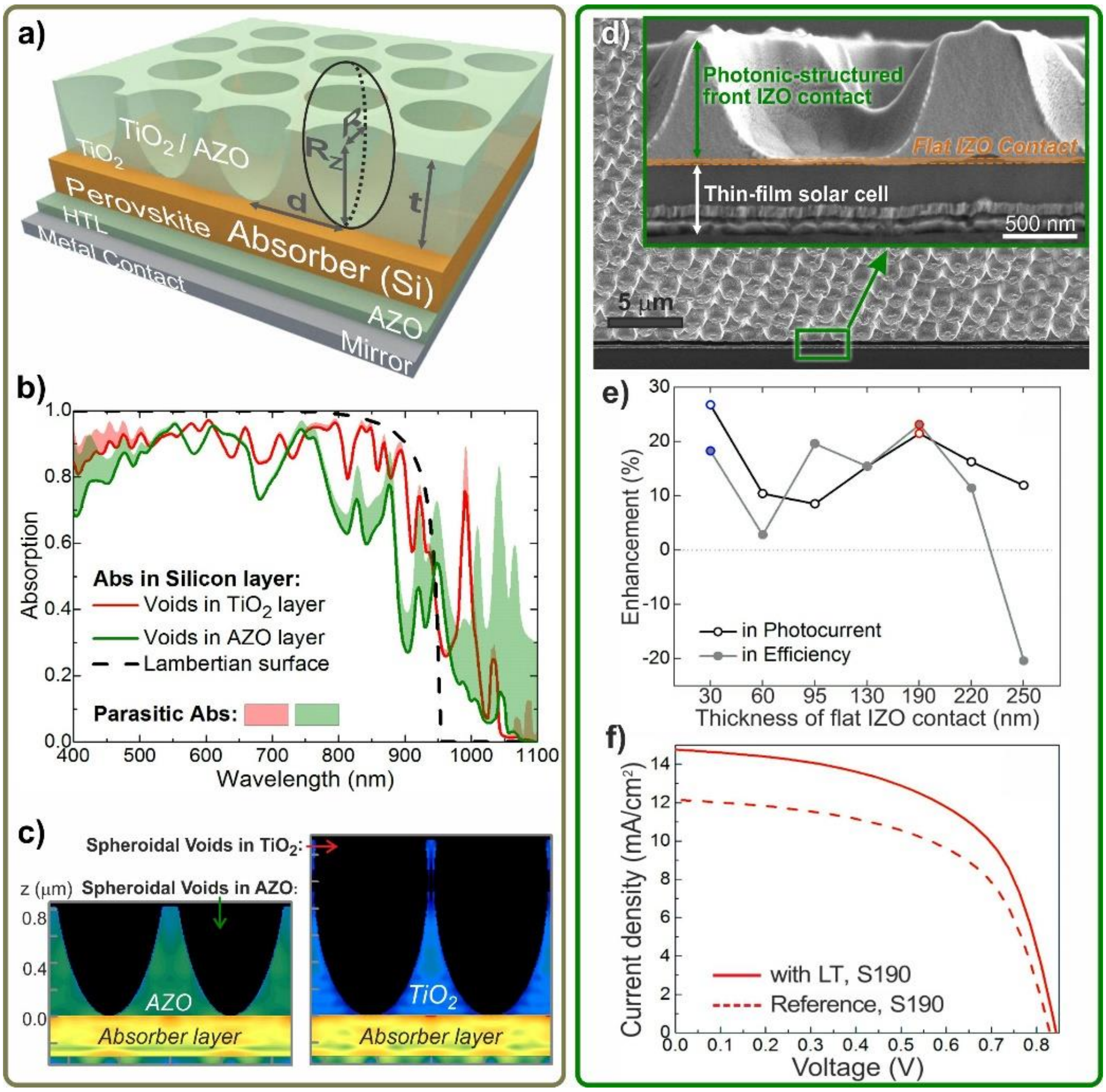

Fig. 3 a-c) Electromagnetic modelling results of hexagonal arrays of TiO_2 half-spheroids (a-c) or semi-spheroidal voids in a TiO_2 or AZO layer (d-f), both integrated in thin-film (300 nm) Si solar cells [11]. The results show the light absorption, Abs, spectra (b) and photo-generation rate, G, profiles (c) of the optimized photonic front structures. d) SEM of a solar cell, composed of the layer structure: glass (substrate)/Al+AZO (rear contact)/a-Si:H (n-i-p absorber)/IZO (front contact), coated with a photonic-structured IZO front contact. e) Evolution of the enhancement of the solar cells' J_{SC} and efficiency with the thickness of the flat IZO layer, marked in orange in d), separating the a-Si absorber from the front LT structures. c) 1-Sun J-V curves obtained for the flat IZO thickness (190 nm), marked by the red circles in e), that allows the highest efficiency enhancement caused by the LT features, as compared with the respective flat solar cell (reference) without the photonic front structures [6]. ***Reproduced with permission from Elsevier.***

2.2 Photonic-enhanced Thin Film Solar Cells

The authors have shown that effective wave-optical structures can be made of low-cost materials and can be straightforwardly integrated at industrial scale via inexpensive and large-area soft-lithography processes. One of the preferential methods for high-throughput photonic patterning is known as colloidal lithography (CL). It consists in depositing a monolayer of close-packed colloidal polystyrene (PS) microspheres by wet-coating [42]. The microspheres are then reduced and shaped by applying a reactive ion etching (RIE) process, thus resulting in a non-closed-packed hexagonal array of spheroids that acts as a mask for the deposition of additional material. Finally, a lift-off step removes the PS colloids, leaving only the microstructured material on the front surface (Fig. 3d). CL is a particularly advantageous technique for PV application, as it can be adapted to precisely pattern any material with the dimensions appropriate for efficient LT (as those of Fig. 3d), and can be implemented in any type of solar cell with different absorbers (based in perovskites [1, 2, 43], CIGS [44], tandems [45], etc.).

The authors developed a CL approach to engineer optically-enhanced TCO materials that can perform the double role of front electrode and LT medium [46]. Besides the optical benefits due to LT effects, such solution is also electrically advantageous since the use of a photonic-structured TCO allows higher volume of electrode material without optical losses, thus enabling lower sheet resistance of the front contact. Furthermore, it avoids the use of another distinct material for the LT medium, since the front TCO can assume such role, thus reducing the process complexity and costs. To explore such advantages, photonic-structured TCO front contacts (made of IZO) were applied on a-Si:H test solar cells [6]. Fig. 3d shows the micro-patterned IZO top contact layer patterned on the cells, following the aforementioned CL process [21]. The resulting hexagonal arrays of ~850 nm tall pyramidal-shaped features uniformly cover the active area of the solar cells, with a pitch defined by the initial diameter (1.6 μm) of the PS spheres. In addition of the geometrical parameters of the photonic features, another critical parameter here is the thickness of the flat IZO layer separating the base of the pyramidal-like features from the Si absorber layer. The thinner the separation the higher can be the optical gains due to the LT effects, but the presence of such flat layer is always necessary to allow a sufficiently low sheet resistance at the front contact. Therefore, we carefully studied this compromise [6], looking for the optimum separation between the photonic and absorber media.

As shown in Fig. 3e, two thicknesses of such flat IZO layer lead to particularly prominent results: 30 nm (S30 cells) and 190 nm (S190 cells - IVs shown in Fig. 3f). In S30, the LT features are placed very close to the a-Si layer, so the attained J_{SC} is high, but the electrical performance (V_{OC}, FF) of the devices is poor due to the ultra-thin flat IZO layer connecting the front contact. The application of the IZO-based LT structures reinforces the conductance of such contact due to the incorporation of additional IZO material, so the efficiency of this cell is highly enhanced due to both optical and electrical improvement. In the case of S190, the larger separation between the LT features and the a-Si is compensated by both the electrical reinforcement of the IZO top contact and the optimized anti-reflection action of the flat IZO layer, thereby enabling the highest efficiency gain (23.1%) and highest absolute values in J_{SC} (14.8 mA/cm^2) and efficiency (7.2%) attained in this study.

2.3 Additional Advantages: Higher Angular Acceptance and Self-Cleaning

The previously analyzed opto-electronic response of the solar cells was performed under normally incident illumination. Nevertheless, it is also important to analyze the cells' response for oblique incidence, particularly with thin-film devices that are usually not mounted on sun-tracking platforms. In addition, if the cells are integrated on flexible substrates, they can operate in a bent/curved state having a range of incidence angles shining throughout their active area. Therefore, if their response is not omnidirectional, a J_{SC} reduction in one portion of the active area can cause an overall drop in the total current supplied by the cells. In view of this, it is advantageous to implement LT structures able to provide high J_{SC} values as independent as possible of the incidence angle [11, 47].

The analysis of the angle-resolved opto-electronic response of the S30 and S190 cells in our work [6] showed that the maximum absolute values of photocurrent and voltage occur at 0º (light impinging normally to the cell surface) and minimum for 90º (light impinging parallel to the surface). As expected, when the incidence angle increases there are increased losses by reflection which reduce the power output. However, when observing the angular response of the enhancement values, it was observed that the LT structures are even more beneficial for oblique illumination. For S190, the gains in efficiency and J_{SC} are quite similar and increase for angles away from the normal incidence, reaching respectively 52% and 53.2% at ±70º angle. For S30, the gain in J_{SC} follows a similar trend, but the efficiency enhancement becomes even higher with increasing angle, peaking in 52.2% at ±40º angle, which is mainly due to the fact that the efficiency of the planar reference cell decreases more with increasing angle than that of the LT-enhanced cell.

Similar pronounced broadband and broad-angle LT enhancements were attained with a quite different wave-optical structure having a dome-like geometry, also fabricated via CL [4]. In this work, a parylene film was micro-patterned with an hexagonal array of cones (see SEM in Fig. 4c) after being coated on the front TCO of nc-Si:H solar cells. Fig. 4a displays the JV curves with illumination angle ranging from 0° to 90°. The first aspect here reported is the pronounced J_{SC} enhancement, relative to the uncoated reference, not only for normal incidence (23.6% gain) but even higher for oblique illumination. The polar plot of Fig. 4b shows the influence of the illumination angle on the efficiency and J_{SC} enhancement, relative to the flat reference cell without the front microstructures. These enhancement values increase with angle until ~50°, with peak enhancements up to 52% and 61% in J_{SC} and efficiency, respectively; while the V_{OC} increased about 5%, and FF was marginally reduced (~1%) with increasing angle. Consequently, it is estimated that such photonic-structured coatings can allow substantial enhancements of > 35% of the average daily power supplied by the cells.

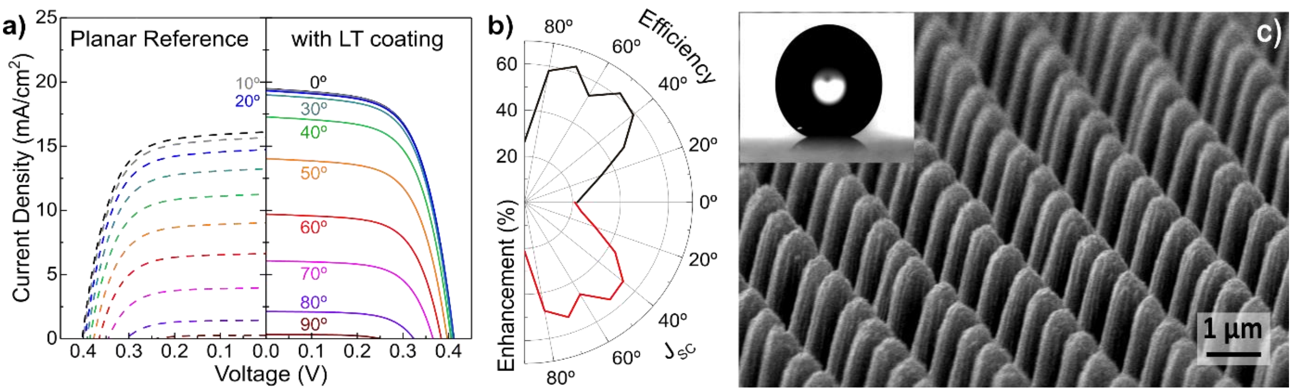

Fig. 4 a) 1-Sun JV curves of the solar cells before (uncoated reference, *left curves*) and after (*right curves*) coating with the photonic-structured parylene shown in images c, d), for illumination angles varying from 0° (normal to cell's surface) to 90° (parallel to surface). b) Polar plot presenting the angular dependence of the gain in efficiency (top quadrant) and photocurrent (bottom) of the LT-enhanced solar cell relative to the planar reference. c) Artistic image of the photonic coating on the transparent front contact of the solar cells. The microstructured parylene provides LT while also acting as a water-repellent protective layer which allows an effective self-cleaning functionality [4]. ***Reproduced with permission from Wiley.***

Interestingly, when developing the micro-structured photonic coatings of Fig. 4c for LT, it was observed that the resulting micro-roughness and surface-chemistry modification provided super-hydrophobicity, with water contact angle of up to 165.6° with extremely low adhesion, thus enabling self-cleaning effect. It is well-known that the accumulation of dust, snow or other particles on solar panels hinders light capture and promotes faster degradation (e.g., via hot-spot formation) [48], leading to severe unpredictability in the devices' response and to efficiency losses [49-52]. This represents a significant cost in large-scale PV installations, as not only is the power generated by the dusty panels affected, but periodic cleaning is also mandatory. To solve this problem, the extreme tuning of the surface wettability properties via super-hydrophobic coatings is regarded as one of the most cost-effective approaches. Inspired by plant leaves (e.g., *Lotus* flower) with self-cleaning capability [53], super-hydrophobic coatings possess conspicuously low surface energy [54], allowing water droplets to easily roll down the coatings' surface, carrying away any existing dust particles, thus producing a biomimetic self-cleaning effect [55]. Moreover, the micro-structures required for the geometric-based super-hydrophobic behavior are well in range with those studied for LT purposes in the wave-optics regime, thence permitting improvements in the solar cells' optical as well as outdoor environmental performance [4, 23, 56, 57].

Parylene-C [poly(chloro-p-xylylene] was the preferred coating material for this study, since it is an extremely stable polymer with excellent barrier properties for encapsulation [58-63] and low surface energy. Moreover, it presents excellent optical transparency [64] and an appropriate refractive-index for anti-reflection purposes, combined with great flexibility and mechanical strength [55]. The parylene's hydrophobicity was controlled by simultaneously adjusting the surface corrugations (i.e., roughness, patterned features) and also the chemical composition of the surface [1, 54, 55, 65-69] with the CL patterning process.

3. Conclusions

The authors have demonstrated that the use of wave-optical photonic structures, integrated in the illuminated contact of solar cells, is an attractive concept capable of boosting the photocurrent supplied by the devices, as a consequence of the broadband light absorption enhancement that they provide in the PV material, as well as their angular acceptance. The fundamental LT limits in the wave-optics regime are not yet fully understood [20], but what has become clear is that this type of LT structures already showed higher optical potential than that of conventional geometric optics [5]. It is imperative to pursue further theoretical research in wavelength-scale structures for a better understanding of their light-management potentialities, which is crucial for most opto-electronic technologies (PV, photonic computation, LEDs, optical sensing, etc).

Also, importantly, when applied in the top contact of solar cells with flat PV layers, the integration of wave-optical structures does not harm the devices' electrical performance (as occurs with conventional LT based in texturing), so the attainable gains in the power supplied by the cells can be as high (or even higher) than the photo-carrier generation gains, as seen in previous results. Nonetheless, as a final remark, we underline that recent breakthrough research has revealed that the use of wave-optical structuring can enable several other types of advantages for PV technology besides improving the photocurrent supply:

1) Physically-thinner but optically-denser solar cells also allow:

- Lower material usage — leading to savings in costs and manufacturing time, but is also particularly crucial for PV absorbers with critical/hazardous materials (e.g., lead-containing perovskite solar cells, PSCs) [43].

- Electrical gains — caused by two effects: 1) a reduced thickness of the PV material implies lower bulk carrier recombination, and 2) if the photonic structures integrated are made of a TCO material they can lead to reduced sheet resistance of the front contact [6].
- Higher Flexibility — the flexural rigidity scales with the third power of the films thickness, which is crucial for driving the novel generation of solar-powered portable electronic and BIPV systems [68, 69].

2) Stability improvements, which is particular crucial for less-matured PV technologies, due to:

- UV Protection — the front-located LT structures absorb most of the UV light in their material, thus preventing the ingress of such higher-energy photons towards the sensitive cell layers. This is particularly useful for PSCs, since UV penetration is one of the main effects that accelerate the cells degradation [1, 2].
- Water repulsion — the micro-scale dimensions and high aspect-ratio of the photonic features can lead to a super-hydrophobic water-repellent front surface, enabling the water droplets to easily roll-off carrying away dust or other debris on the devices. In addition to an important self-cleaning functionality, this effect prevents humidity accumulation on the cells which can improve the environmental stability of water-sensitive PV technologies such as PSCs [4].

Acknowledgements

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