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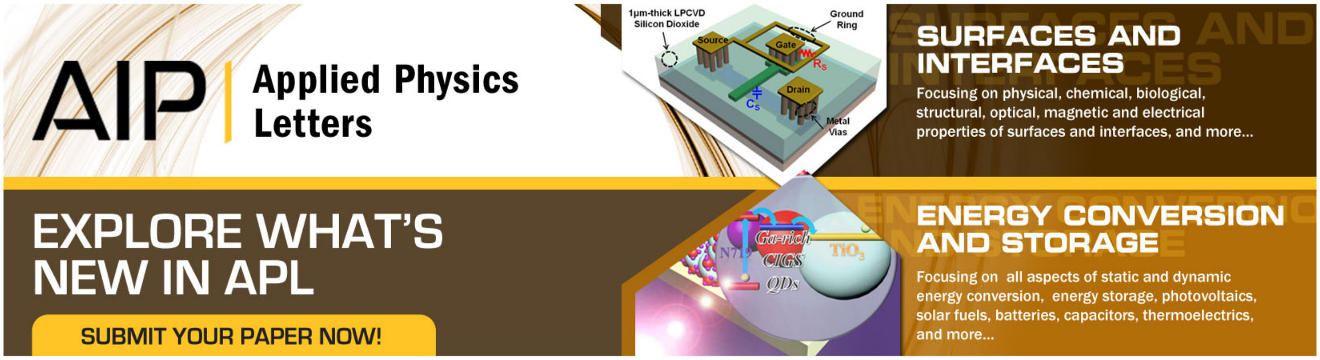

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Light trapping with total internal reflection and transparent electrodes in organic photovoltaic devices

Kristofer Tvingstedt, Zheng Tang, and Olle Inganäs
Biomolecular and Organic Electronics, Center of Organic Electronics, Department of Physics, Chemistry and Biology, Linköping University, SE-581 83 Linköping, Sweden

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Herein, we demonstrate a method to build highly efficient light trapping structures for printed organic solar cells and modules, compatible with roll to roll manufacturing. Echelle grating structures in combination with semitransparent electrodes allow for efficient light trapping via means of total internal reflection. With this method, we demonstrate an increased cell photocurrent response up to 24%, compared to a standard cell configuration with a planar reflector. The demonstrated light trapping approach is expected to be even more useful for photovoltaic modules, where light hitting "dead areas" in between the sub-cells comprising the module will now be utilized. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4759125]

Organic photovoltaics (OPV) are emerging as a potential alternative inexpensive route to convert solar energy into electricity due to encouraging recent progress in the field.[1–3] In particular, polymer-based material devices are expected to be more cost-effective as they can be easily processed from solution, which enables the use of simple printing techniques. Low cost of production is envisaged, but increased efficiency is paramount for making these devices attractive. Absorption of solar radiation typically requires absorber layer thicknesses where charge recombination becomes a severe limitation, lowering the internal quantum efficiency (IQE) of the cell, whereas in thinner films, charge generation and collection can be almost complete. This conflict between optical absorptance and charge recombination in organic photovoltaic films calls for new optical strategies.

In this work, we demonstrate a promising route for increasing the active layer absorptance in thin solar cells with high internal quantum efficiency through means of light trapping and photon recycling via total internal reflection (TIR). Light trapping techniques are essential in indirect band gap inorganic photovoltaics such as silicon and have been implemented for a long time.[4,5] The direct transfer of these geometries to solution processed thin polymer solar cells is however not possible. An efficient light scattering element, working in the incoherent (ray) domain, should be somewhat larger than the wavelength of light to effectively alter the photon propagation direction. When incorporating such refractive or reflective structures in combination with a ~100 nm thick organic film, difficulties are obviously inherent. The electrical properties are compromised when including light scattering elements in direct electrical contact with such thin active layers.[6] Hence, engineering approaches may be required, where the light trapping element is separated from the light absorbing layers to maintain favorable multiple absorption events but to avoid the possibility of electrical defects.

Light trapping techniques compatible with thin film polymer cells and solution processability have previously also been suggested. Arrays of micro lenses and apertures collectors have been demonstrated,[7] and folded organic solar cells have shown considerable improvements in light collection as well as photocurrent.[8,9] Backscattering structures with intermediate aspect ratios coated conformally with active layer from solution can cause light trapping and a positive effect on photocurrent has been confirmed.[10] These light trapping configurations however all suffers from various drawbacks. The array of lenses and holes is only working for light entering from a very small set of angles. The folded cells are working efficiently for most incidence angles[11] but are on the other side consuming more material and are more difficult to realize on an industrial scale. The geometrical requirement of the intermediate aspect ratio structures and the needed high conformal active layer coating homogeneity renders these approaches difficult to scale. A further drawback of active layers in direct contact with structured electrodes is that the possible surface recombination will be increased, as the surface to volume ratio increases.[12] Recently, proposed light confinement methods[13–15] operating in the wave domain and suitable for planar films have theoretically indicated that very strong field enhancement in the active layer is possible. Such methods could be suitable for organic solar cell as the dielectric function of the organic active layer is rather low, and the absorbing layer is rather thin, which is a necessity in these methods. Experimental verification of such strong light confinement routes, able to beat the Yablonovitch limit, however remains to be demonstrated. Recent approaches of coherent light management have further proven that a thin film solar cell electrode does not necessarily have to be very transparent.[16] In this work, we however stay in the ray domain approach and incorporate large tilted reflective structures of a ladder type "echelle" or blazed grating which are able to shift the propagation direction of light into angles of total internal reflection. Other light trapping approaches using blazed grating, holographic, or others rely on multimodal waveguide coupling resulting from the diffraction of fine structured patterns.[17,18] The structures employed here are much larger, with a pitch of 55 μm and a height of 40 μm, and are optically more related to an array of tilted mirrors.

Reflection of light from shallow angles at the inside of a dielectric media has the advantage of being completely lossless, unlike the reflection of light by metals. As the same dielectric media, in our case glass (or plastic), is used as a highly transparent encapsulation lid and effectively lets light into the solar cell, all that is required to utilize the light trapping property of TIR is of course to change the direction of light propagation *inside* the cell. This is also the standard way to trap light in thick inorganic solar cells. The structures required to effectively alter the light propagation direction into angles enabling TIR are, as stated above, not easily compatible with conventional OPV geometries and solution based coating techniques. However, by employing two planar transparent electrodes, we can realize light trapping via TIR without interfering electrically in the cell, as cell planarity is undisturbed. A conceptually similar approach employing only one large inverted pyramid behind a small cell was recently demonstrated.[19] To realize the devices outlined here, well performing semitransparent solar cells where the major optical losses are primarily found in transmission, and no longer in reflection, are required. Such semitransparent solar cells were also recently demonstrated[20] and are predominantly expected to be incorporated in windows for building integrated photovoltaics. We now go beyond that approach and place a directionally reflecting mirror in direct optical contact behind the semitransparent solar cell, removing its transparency. This is conceptually shown in a module with 4 separated cells in optical contact with reflecting echelle microstructures (Fig. 1(a)). An additional benefit is obtained for module configurations with multiple subcells, as the areas in between the subcells will no longer be unutilized area. Light impinging here will be redirected, possibly after a few internal reflections, into regions of the module subcells where photocurrent is generated. No such modules as in Fig. 1(a) were manufactured in this work, but merely single cells.

The large reflecting echelle grating is laminated on the backside of a semitransparent solar cell with the aid of either optical adhesives or cured poly(dimethylsiloxane) (PDMS). The latter enables conformal optical contact between the cell and the echelle reflector, while still keeping the option to simply remove the structures from the cell, enabling easy comparison between cells with or without different reflectors. The echelle grating microstructures responsible for the light redirection are manufactured by means of micro embossing of an acrylic resin in a roll to roll process by Microsharp Ltd (UK). These structures were then metallized twice, from two different angles such that good structure coverage was obtained, through thermal sublimation into an at least 200 nm thick Ag layer. The asymmetrical structure of a blazed grating, originally proposed for light trapping by Heine and Morf,[5] was preferred over a symmetrical grating, as it does not couple out light after one TIR event, in the way reciprocity forces a symmetrical structure to do.[5,19] It is however far from a Lambertian reflector we employ here, and the expected improvement is therefore lower than what is predicted from the Yablonovitch limit ($4n^2$) for light scattering under such perfectly randomizing conditions. The path length enhancement would in that limit amount to around 9 as the refractive index of the adhesive is what determines the trapping. To get a better understanding of how the employed asymmetrical structures operate we implemented ray tracing calculations which are included as supplementary material.[21] The employed blazed echelle structures are depicted in a SEM image in Fig. 1(b).

Reflectance measurements performed by placing three types of devices at the backside opening of an integrating sphere in a Perkin Elmer Lambda 950 spectrophotometer reveal the pronounced optical darkening (also obvious by simple visual inspection) that occurs when the TIR enabling structures are laminated on the backside of the semitransparent cell. A semitransparent cell is first measured in both transmission and reflectance mode. A planar Ag mirror, or an echelle Ag mirror, is then laminated in optical contact on the backside of the cell, and the device reflectance is measured. These latter samples hence show no transmission. To allow for a relevant comparison, we plot the complete device absorptance A (=100-R-T) for the three compared configurations in Fig. 2. A pronounced increase in device absorption is achieved for the two reflecting devices. This increase in device absorptance is however only partly due to an actual

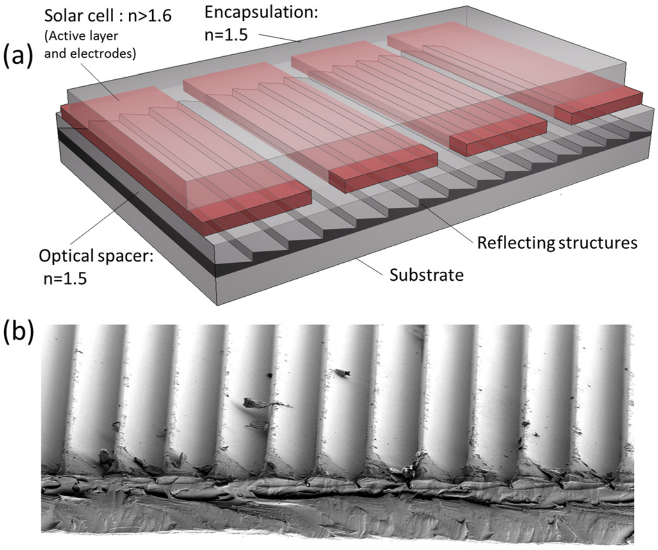

FIG. 1. (a) Sketch of the TIR echelle light trapping configuration in a proposed module arrangement. Light hitting the "dead area" in between the (four) sub cells in such a module will be redirected into photocurrent generating regions. (b) SEM image of the reflecting echelle structures on a plastic substrate. The structure pitch is 55 μm and the height is 40 μm.

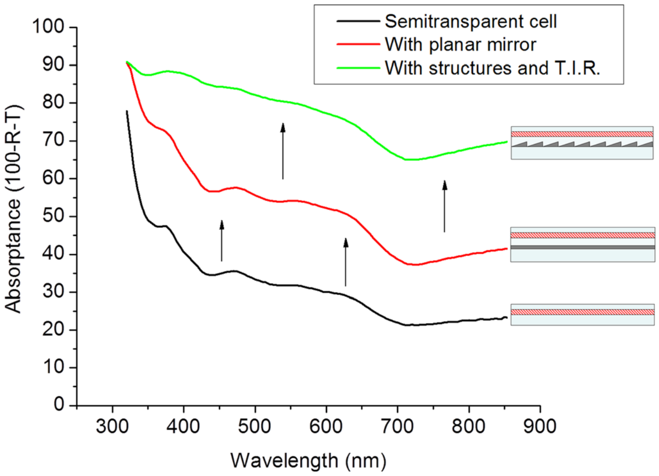

FIG. 2. Device absorptance as determined by reflectance and transmission measurements for the three compared configurations.

increase of absorption in the active layer of the solar cell, as we may have pronounced parasitic absorption in the reflectors or the cell electrodes.

The effect of the echelle reflector on active layer absorption is best shown by improvements in solar cell performance. Our semitransparent solar cells were made from TQ1/PCBM70 in ratio of 2:5 and manufactured as described recently in Ref. 18. The active layer thickness was ∼45 nm. The current voltage characteristics of the cell under 100 mW/cm^2 simulated AM 1.5 solar irradiance and the cell external quantum efficiency (EQE) is presented in Figs. 3(a) and 3(b). By comparing the response of a semitransparent cell without any reflecting structures on the backside to the *same* semitransparent cell, but with a planar reflecting mirror or the echelle reflector laminated in optical contact behind, we clearly see the large impact of the different reflectors. To avoid possible overestimation of the effect of the echelle reflector employed for a single solar cell, it is critical to verify that the full illuminated sample area is not redirecting a disproportionate amount of light into the active cell region, defined by the cross section of ITO/active layer/PEDOT. The dominant absorbers (active layer and PEDOT electrodes) was therefore coated on the entire substrate to assure that light hitting the sample outside the active cell region is exposed to similar absorption events as those inside the cell region. Hence, absorption and scattering that occurs outside or inside the defined active cell region are very close to identical. That a planar mirror improves the photocurrent from a semitransparent solar cell with ≈70% is not unusual. The doubled beam path generally allows for an improvement factor smaller than 2, since light intensity is attenuated in the first pass through the cell and cannot be as high in the equally long second path. For the cell with echelle reflector, we however see an improvement of 109% revealing that more light is actually absorbed in the second and higher order passes compared to what is absorbed in the first pass. The more justified comparison with the planar mirror behind the cell (non-transparent device comparison) gives then that the echelle reflector is generating 24% more photocurrent. Spectrally resolving the effect of the light trap with EQE measurements (Fig. 3(b)) further demonstrates that the improvement is also strongly dependent on wavelength. EQE measured with structured and planar mirror is divided by the EQE of the semitransparent cell, as inset in Fig. 3(b), to elucidate at what wavelengths the improvement occurs. Although an improvement is occurring for all photon energies, it is clear that in the range where absorption is weakest, closer to the band edge, we indeed also have the most pronounced enhancement. At around 700 nm where active layer absorption is very weak, we have an improvement in EQE of as much as 3.3 times that of the semitransparent cell. As this is a region where the solar photon flux is quite strong, it hence contributes quite significantly to the improved J_{sc}. Just below 400 nm, it is also observed that the improvement is lower for the structured mirror cell compared to the planar mirror cell. This is attributed to a more pronounced parasitic absorption in the echelle reflectors (vide infra).

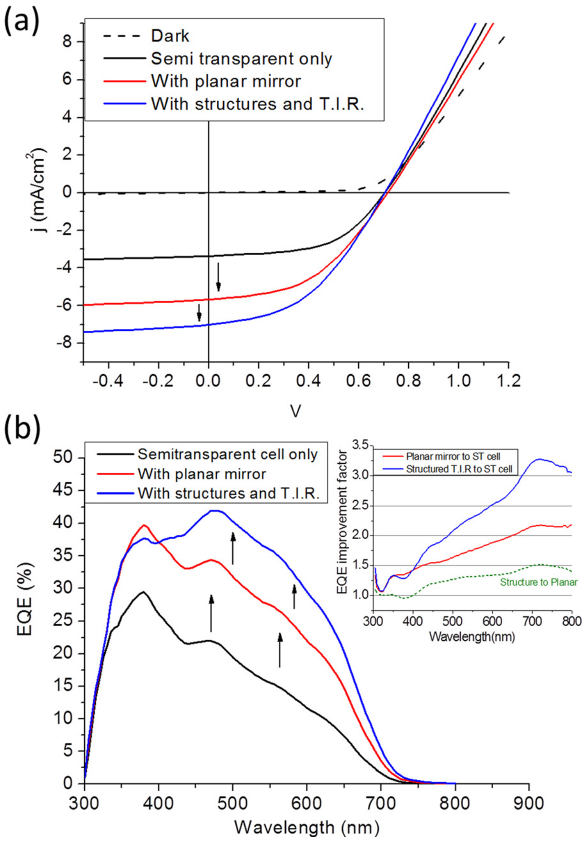

FIG. 3. (a) I-V characteristics of one cell with or without planar and structured echelle reflectors laminated behind. (b) EQE of the same cell with the same structures. Inset shows the ratio of EQE between the different cells, spectrally resolving the effect of the structures.

The separation of external quantum efficiency into active layer photon dissipation and internal quantum efficiency is important to identify the distribution between optical and electrical losses in a solar cell. We here therefore employ the transfer matrix model[22] (TMM) in combination with transmission and reflectance measurements to disentangle the active layer absorptance from parasitic optical losses at the electrodes or the reflectors. As the TMM model is not compatible with multiple light directions in its interference pattern calculations, we cannot employ the model for the cell with the scattering structures. However, as the same cell was used, with and without the scattering structures laminated behind, we can employ the TMM model for the planar semitransparent cell to correctly calculate the internal quantum efficiency of the cell and use that value (which must remain the same) to "back calculate" the active layer absorptance for the other two configurations, after we have measured the external quantum efficiencies of those. The calculated IQE for the semitransparent cell itself is presented in Fig. 4(a) and approximated with a wavelength independent value of ∼83%. The dissipation in this device for the different layers is included as a large inset in Fig. 4(a). The red line represents the active layer dissipation which is used for the IQE calculations. The set value of 0.83 is then used as input to obtain the active layer dissipation (*EQE*/*IQE*) for the two other configurations (Fig. 4(b)). Here, it is clear that there indeed is a pronounced increase of photon absorption in the desired active layer, but not as pronounced as first anticipated. The difference between the measured complete device absorptance of Fig. 2 above and those of active layer absorptance in Fig. 4(b) reveals that there is actually a quite

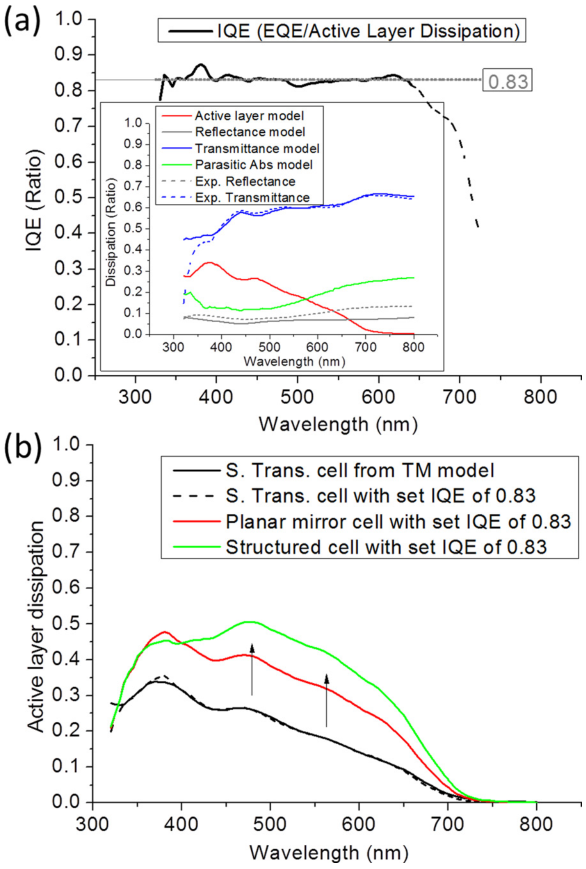

FIG. 4. (a) Calculated photon dissipation in the different layers of the semitransparent cell. Internal quantum efficiency can then be determined to be approximately 83% for the cell. (b) With the known cell IQE of 83%, we can back calculate the active layer dissipation as EQE/IQE when the cell is used in combination with the planar or structured mirror laminated behind.

pronounced increase in parasitic device absorption as well. This is currently assigned to occur primarily in the actual reflecting structures, which therefore need to be further optimized to demonstrate the full potential of this light trapping approach.

To evaluate the relevance of this light trapping approach under daylight illumination conditions, experiments with varying angle of incident illumination were also performed. The sample was tilted around its center axis (parallel or perpendicular to the linear grating structures) in both directions. Fig. 5 reveals that an increase is obtained for a large range of incident angles and further that the increase is not the same when rotating in the two different directions. This is not surprising as the grating structures are asymmetrical. What is important to note is that the structured cell benefitting from light trapping actually never performs worse than the cell employing a planar mirror.

Comparing the performance of this light trap with that of others employed for OPVs, we note that the improvement of photocurrent density by 24% here reported is similar to what is obtained for micro lenses[7] but lower than what is found for folded cells.[8] These comparisons are not for the same thickness, nor the same material, and should therefore be taken as indications only. The angle dependence, material usage, and ease of manufacturing however speak to the advantage of the here proposed method. A recent demonstration relying on wrinkles and folds at the micrometer scale in organic photovoltaic cells reported a significant photocurrent improvement of as much as 47%.[23] Accounting for the improvement in terms of the EQE, there showed very large enhancements at sub band gap wavelengths which contributed largely to the photocurrent.

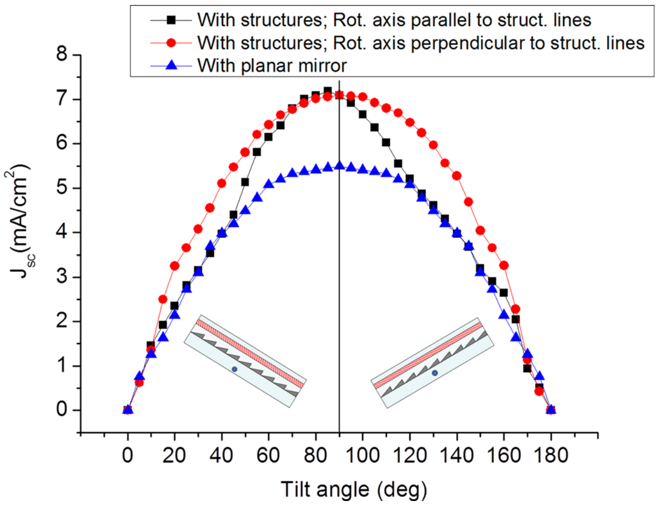

FIG. 5. Angle dependence along two rotational axes, parallel and perpendicular to the structure lines, of the structured cell compared to a cell with a planar mirror. The improvement in photocurrent attributed to light trapping is significant for a large range of angles. During a full day with solar illumination incident from many angles, the structured cell is hence clearly outperforming the cell with a planar mirror.

In conclusion, we have demonstrated a light trapping configuration suitable for large scale module manufacturing of solution printed organic solar cells. We demonstrate significant improvement in both absorption and photocurrent when redirecting light into angles of total internal reflection at the air interface of the cell. We further verify that the configuration is suited also under full day illuminations with varying angles of incidence. We however note that parasitic absorption in the employed metal structures remains problematic and propose that alternative structures may minimize these losses.

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