APP Applied Physics

Two-dimensional finite-element modeling of periodical interdigitated full organic solar cells

P. Granero, V. S. Balderrama, J. Ferré-Borrull, J. Pallarès, and L. F. Marsal

Citation: J. Appl. Phys. **113**, 043107 (2013); doi: 10.1063/1.4788819 View online: http://dx.doi.org/10.1063/1.4788819 View Table of Contents: http://jap.aip.org/resource/1/JAPIAU/v113/i4 Published by the AIP Publishing LLC.

Additional information on J. Appl. Phys.

Journal Homepage: http://jap.aip.org/ Journal Information: http://jap.aip.org/about/about_the_journal Top downloads: http://jap.aip.org/features/most_downloaded Information for Authors: http://jap.aip.org/authors

ADVERTISEMENT





Two-dimensional finite-element modeling of periodical interdigitated full organic solar cells

P. Granero, V. S. Balderrama, J. Ferré-Borrull, J. Pallarès, and L. F. Marsal^{a)} Nano-Electronic and Photonic Systems (NePhoS), Department of Electronic, Electrical and Automatic Control

Engineering, Universitat Rovira i Virgili, Av. Països Catalans 26, 43007 Tarragona, Spain

(Received 22 November 2012; accepted 7 January 2013; published online 25 January 2013)

By means of finite-element numerical modeling, we analyze the influence of the nanostructured dissociation interface geometry on the behavior of interdigitated heterojunction full organic solar cells. A systematic analysis of light absorption, exciton diffusion, and carrier transport, all in the same numerical framework, is carried out to obtain their dependence on the interface geometrical parameters: pillar diameter and height, and nanostructure period. Cells are constituted of poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl- $(6,6)C_{61}$. Results show that light absorption is maximum for pillar heights of 80 nm and 230 nm. However, due to the short exciton diffusion length of organic materials, the analysis of the exciton diffusion process reveals that the 80 nm thickness gives rise to a higher photocurrent, except for the smaller pillar diameters. In terms of efficiency, it has been observed that the charge carrier transport is weakly dependent on the geometric parameters of the nanostructured interface if compared with the exciton diffusion process. The optimal cell is a device with a pillar height of 80 nm, a structure period of 25 nm, and a ratio of the nanopillar diameter to the period of 0.75, with an efficiency 3.6 times higher than the best planar bilayer reference device. This structure is such that it reaches a compromise between having a high proportion of P3HT to increase light absorption but preserving a small pillar diameter and interpillar distance to ensure an extended exciton dissociation interface. © 2013 American Institute of Physics. [http://dx.doi.org/10.1063/1.4788819]

I. INTRODUCTION

Since the advent of organic solar cells (OSC), there has been an intense search to make them at least as profitable as traditional polluting energy sources. However, this objective is still far since only recently a power conversion efficiency (PCE) of 10% (the minimum required for an OSC to be, at least, as competitive as an inorganic device) has been achieved.^{1,2}

Unlike inorganic solar cells, when photons are absorbed in organic devices they do not generate free charges but excitons. This quasiparticle, that is a bound state of an electron and a hole, must diffuse and reach a dissociation interface to generate free charges.³ However, organic materials have a short exciton diffusion length, typically 5–15 nm, so only those excitons generated close enough to a dissociation interface will contribute to photocurrent while the rest will recombine.^{3,4} In OSC, the dissociation interface corresponds to the donor-acceptor (D-A) union. Hence an extended dissociation interface is crucial to improve the efficiency of OSC.

One promising solution is the interdigitated heterojunction approach. This method provides devices with a widespread D-A interface. The advantage over the bulk heterojunction approach is that the interdigitated cells provide uninterrupted direct paths for charge carrier collection to the electrodes.⁵ However, this advantage is achieved at the expense of a reduced D-A interface area in comparison with the bulk heterojunction structure. Previous works have shown that the improvements in the electrical behavior of the interdigitated heterojunction cells can lead to an increase in their efficiency. $^{6-11}$

Kim *et al.*⁶ reported an increase of 6.6 times in the efficiency of poly(3-hexylthiophene) (P3HT)-C60 interdigitated devices if compared with a planar bilayer reference cell. Their cells had 50 nm of nanopillar diameter, 150 nm of nanopillar height, and 100 nm of structure period. He *et al.*⁹ increased the efficiency from 1.36% (planar) to 3.25% (interdigitated) in P3HT-1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C₆₁ (PCBM) devices with cells that had 25×25 nm of nanorod size, 80 nm of nanorod height, and 50 nm of structure period. Other authors have also reported improvements in cells efficiency in devices of different materials, such as Zheng *et al.*¹⁰ with two-fold efficiency increase (20 nm diameter and 40 nm long nanopillars) and Wiedemann *et al.*¹¹ with an improvement of about 80% (40 nm-width and 80 nm-periodicity structures).

The interdigitated D-A interface can be obtained with the template-assisted synthesis method since it allows obtaining nanometer scale structures.^{6,12–15} In the previous works, we demonstrated the possibility of obtaining polymer nanopillars onto indium-tin-oxide (ITO)/coated glass substrates from nanoporous anodic alumina templates (NAAT).¹⁴ Once the NAAT are made,¹⁶ the polymer is inserted in the template with a combination of the spincoating and the melt-assisted template wetting methods. Finally, the NAAT is dissolved in a solution of sodium hydroxide (NaOH), obtaining the polymer nanopillars.¹⁴ The geometrical features of the NAAT such as pore diameter, interpore distance, porosity, degree of hexagonal pore

0021-8979/2013/113(4)/043107/7/\$30.00

a)lluis.marsal@urv.cat.

arrangement, and thickness are rather controllable by the anodization parameters (anodization voltage, temperature, and type and concentration of the acid electrolyte).¹⁶ In Figure 1, we have two environmental scanning electron microscopy (ESEM) images of P3HT nanopillars obtained under different anodization conditions. Figure 1(a) shows nanopillars with an average height of 380 nm, a diameter of 180 nm, and an interpillar distance of 490 nm. The nanopillars are standing on a 220 nm thick P3HT base layer, which is in contact with the ITO-coated glass. Figure 1(b) shows P3HT nanopillars on a 260 nm thick base layer and with 100 nm height, 70 nm diameter, and 100 nm interpillar distance. Nevertheless, an optical and electrical modeling of such structures is crucial to evaluate a-priori the performance of this interdigitated cells and the dependence of such performance on the different geometrical and material parameters.

Several authors have previously presented simulation approaches to model the behavior of interdigitated heterojunction OSC.^{17–19} Yang and Forrest¹⁷ have simulated the photocurrent generation in nanostructured organic solar cells by using a dynamical Monte Carlo model that includes the



FIG. 1. (a) ESEM cross section image of P3HT nanopillars with 380 nm high, 180 nm diameter, and 490 nm interpillar distance and (b) of P3HT nanopillars with 100 nm high, 70 nm diameter, and 100 nm interpillar distance.

generation and transport properties of both excitons and free charges. Despite their extensive study, only efficiencies for the internal and external quantum efficiencies are calculated while the achievable energy conversion efficiencies are only estimated by using typical experimental values for the open circuit voltage and the fill factor. In simulations of Meng *et al.*,¹⁸ excitons are created at randomly chosen sites in either the hole or the electron conducting polymer at constant rate, neglecting light wave features such as diffraction and interferences. A more accurate model has been presented by Kim et al.¹⁹ In their approach, the absorbed light in the active layer was computed by considering the wave nature of light with Maxwell's equations and the photocurrent density was calculated by considering the generation, diffusion, and dissociation of excitons following the exciton diffusion equation. However, the electron and hole transport in the polymer materials were computed using an equivalent circuit model.

In this work, we develop a numerical model, based on the finite elements method, for the complete simulation of interdigitated heterojunction full organic solar cells. We integrate in the same model all the steps of the solar energy conversion process: light propagation and absorption, exciton diffusion, and charge carrier transport. The donor and the acceptor materials of our devices are P3HT and PCBM, respectively. Simulations are carried out by using COMSOL MULTIPHYSICS (R). This program is a finite-element analysis software which allows evaluating optical and electrical properties of the devices in detail.²⁰ The main advantages of such approach is that it allows evaluating all the relevant magnitudes as a function of position and that the result of each step can be used seamlessly as the input for the next step. In this work, we determine the influence of the different D-A interface geometrical parameters (pillar diameter and height, and the interpillar distance) on cells efficiency. The obtained results are compared with a reference planar bilayer cell.

II. MATERIALS AND METHODS

A. The computational domain

Our model is a 2D simplification of a real 3D device. It consists of a structure of ITO, poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS), P3HT, PCBM, and a back contact of aluminum (Al) as it is shown in Figure 2. This simplification implies that the cell is nanostructured in the form of interpenetrating P3HT and PCBM grooves. The ITO, PEDOT:PSS, and Al thicknesses are fixed while the variables are the P3HT groove width α , (equivalent to the nanopillar diameter in a 3D nanostructure), the period β (equivalent in 3D to the interpillar distance), the ratio of the groove width to the period $(\gamma = \alpha/\beta)$, and the nanopillar height including the fix base layer (T). In our case, β ranges from 25 nm to 4000 nm, γ takes the values of 0.25, 0.50, and 0.75, and T is in the range from 30 to 250 nm in intervals of 10 nm with an additional value of 500 nm. The optical properties of the materials have been modeled by using the refractive index (n) and the extinction coefficient (k) of the complex index of refraction ($\tilde{n} = n + ik$), which have been obtained from the literature.^{21–24} As reference, the results are



FIG. 2. Schematic unit cell model of the structure ITO/PEDOT:PSS/P3HT/ PCBM/Al showing the fixed parameters and the variables under study.

compared with an equivalent planar bilayer cell with the same amount of the different materials for each case.

The incident light is assumed to be normal to the surface of the devices and incident from the ITO side. It has been modeled as the superposition of a set of monochromatic linear polarized waves with a planar wavefront, and with a direction that is parallel to the nanopillars. The amplitudes of these plane waves follow the standard AM1.5 spectral distribution of the solar radiation. To obtain the complete spectrum, simulations have been carried out for wavelengths in the range from 280 to 650 nm in intervals of 10 nm. We have chosen this wavelength range because it corresponds to the absorption spectrum of P3HT, where most of the excitons will be generated.

B. Optical and electrical modeling

The finite-element method allows the simulation of light propagation inside the cell nanostructure and the computing of the absorbed light power as a function of the position. This magnitude, obtained by solving the complete Maxwell equations, is expressed as the *total power dissipation density* (Q). At any position \vec{r} in the structure, such dissipation $Q(\vec{r};\lambda)$, expressed in W/m³, for a monochromatic wave of wavelength λ is defined as follows:²⁵

$$Q(\vec{r};\lambda) = \langle -\vec{\nabla} \cdot \vec{S} \rangle, \tag{1}$$

where \vec{S} is the Poynting vector, which represents the energy flux (in W/m²) of an electromagnetic field.²⁶ The *total power dissipation density* that will contribute to exciton generation is obtained by integrating Q over the entire active area.

However, we only have considered the excitons generated in the P3HT layer since the contribution to the photocurrent of the ones generated in the PCBM will be very small. The exciton diffusion length in this material is very short so most of the excitons will recombine before reaching the dissociation interface.^{27,28} This happens even for the case of the bulk heterojunction approach where the internal quantum efficiency (IQE) is very low for the wavelengths where PCBM absorbs and P3HT does not (around 700 nm).^{29,30} In addition, light absorption in the PCBM is weak at these wavelengths.²¹ Hence, in order to reduce computational times, we have neglected the excitons generated in the PCBM. So only the *total power dissipation density* in the P3HT layer will be analyzed

$$Q(\lambda) = \int_{P3HT} Q(\vec{r}; \lambda) dV.$$
(2)

Then, by summing it for all wavelengths, we obtain the total light absorption of a device for the incident light source

$$Q_{Total} = \sum_{i} Q(\lambda_i). \tag{3}$$

The total exciton generation rate can be obtained at every point of the P3HT layer by summing the exciton generation rate corresponding to each wavelength. However, in OSC, only those excitons that reach the dissociation interface will contribute to photocurrent, hence it is also necessary to estimate the exciton flux to this interface. After being generated, excitons diffuse around their neighborhood and eventually dissociate into free charges or recombine following the *exciton diffusion equation*. So the exciton density as a function of time at a position \vec{r} can be set as follows:

$$\frac{\partial exc}{\partial t} = D_{exc} \frac{\partial exc^2}{\partial \vec{r}^2} + \sum_i \left(\frac{Q(\vec{r}; \lambda_i)}{hc/\lambda_i} \right) - \frac{exc}{\tau}, \qquad (4)$$

where $D_{exc} = L^2/\tau$ is the diffusion constant of the excitons, being L and τ the exciton diffusion length and mean lifetime, respectively, h is the Planck constant, and λ_i is the wavelength of the considered incident plane wave.

The photogenerated current density (J_{PHOTO}) is proportional to the exciton flux at the dissociation interface $(J_{EXC})^{31}$

$$J_{PHOTO} = q\theta J_{EXC},\tag{5}$$

where θ is the efficiency of exciton dissociation at the interface and q is the elementary charge. By assuming a 100% efficiency in exciton dissociation and in charge collection, we can obtain the maximum attainable J_{PHOTO} . This magnitude gives a first approximation of the amount of absorbed light that it is actually effective.

To obtain the complete current density–voltage (J–V) characteristics, we have used the drift-diffusion model. This model uses the general semiconductor drift-diffusion equations for electrons and holes to describe charge transport.^{32–34} We assume that the transport properties of the organic materials can be modeled by mobility, DOS, and doping levels together with a free charge generation and a bimolecular recombination terms.

The continuity equations for electrons and holes set as follows:

$$\frac{\partial n}{\partial t} = \frac{1}{q} \frac{\partial J_n}{\partial \vec{r}} + G(\vec{r}) - R(\vec{r}), \tag{6}$$

$$\frac{\partial p}{\partial t} = -\frac{1}{q} \frac{\partial J_p}{\partial \vec{r}} + G(\vec{r}) - R(\vec{r}).$$
(7)

In Eqs. (6) and (7), the current density equations for electrons and holes, J_n and J_p , respectively, are

$$J_n = -qn\mu_n \frac{\partial \psi}{\partial \vec{r}} + qD_n \frac{\partial n}{\partial \vec{r}},\tag{8}$$

$$J_p = -qp\mu_p \frac{\partial \psi}{\partial \vec{r}} - qD_p \frac{\partial p}{\partial \vec{r}},\tag{9}$$

where *n* and *p* are the electron and the hole densities, *D* is the diffusion constant of the free carriers, μ is the carriers mobility, and ψ is the electrostatic potential.

The free charge generation rate $G(\vec{r})$ is given by the exciton flux able to reach the dissociation interface. Since in OSC excitons only dissociate in the D-A interface, it is in this region where carriers will be generated. We have modeled a very thin generation zone in this interface to model this effect. For the recombination rate $R(\vec{r})$, we have used the bimolecular recombination definition since in OSC it is one of the most crucial charge carrier loss mechanisms³⁴

$$R(\vec{r}) = \frac{q(\mu_n \mu_p)}{\varepsilon} (np - ni^2), \qquad (10)$$

where ni is the intrinsic carrier density of electrons or holes, and ε is the product of the vacuum permittivity ε_0 and the relative permittivity ε_r of the organic materials. All the parameters have been obtained from the literature.^{3,4,35,36}

Together with the continuity equations, the Poisson's equation also needs to be solved

$$\frac{\partial^2 \psi}{\partial \vec{r}^2} = \frac{q}{\varepsilon} (n-p). \tag{11}$$

Equations (6), (7), and (11) are solved in an iterative loop. To reduce the number of iterations needed to solve the three dependent equations, an initial value for ψ for the Poisson's equation must be supplied

$$\psi_{init} = \begin{cases} -V_t \log(p_{init}/n_i) - \chi - 0.5E_{gap} \text{ for } N_{doping} < 0\\ +V_t \log(n_{init}/n_i) - \chi - 0.5E_{gap} \text{ for } N_{doping} \ge 0 \end{cases}$$
(12)

where V_t is the thermal voltage, n_{init} and p_{init} are the charge concentration at the interfaces of the active layer and the electrodes, respectively, N_{doping} is the doping concentration, χ is the electron affinity, and E_{gap} is the effective band gap between the lowest unoccupied molecular orbital (LUMO) of the acceptor and the highest occupied molecular orbital (HOMO) of the donor. In our case, n_{init} and p_{init} are set as follow:

$$n_{init} = \begin{cases} (|N_{doping}|/2 + \sqrt{(N_{doping}^2/4 + n_i^2)}) \text{ for } N_{doping} \ge 0\\ n_i^2/(|N_{doping}|/2 + \sqrt{(N_{doping}^2/4 + n_i^2)}) \text{ for } N_{doping} < 0, \end{cases}$$
(13)

$$p_{init} = \begin{cases} (|N_{doping}|/2 + \sqrt{(N_{doping}^2/4 + n_i^2)}) \text{ for } N_{doping} < 0\\ n_i^2/(|N_{doping}|/2 + \sqrt{(N_{doping}^2/4 + n_i^2)}) \text{ for } N_{doping} \ge 0. \end{cases}$$
(14)



FIG. 3. Total absorbed light power (Q_{Total}) in the P3HT layer as a function of the pillar height *T* for different interpillar distances β .

III. RESULTS AND DISCUSSION

Figure 3 shows the total absorbed light power (Q_{Total}) in the P3HT layer as a function of the period β for $\gamma = \alpha/\beta = 0.5$. All the curves show a similar behavior with two local maxima for *T* around 80 nm and 230 nm. A local minimum for *T* close to 130 nm is also present in all cases. The maximum absorption is achieved for $\beta = 25$ and 4000 nm while the lower absorption is clear for $\beta = 250$ nm. Since it can be expected that a higher light absorption will lead to a higher photocurrent, we focus the following calculation on T = 80 nm and on T = 230 nm.

Figure 4 shows the obtained J_{PHOTO} as a function of β , for these two thicknesses, for the interdigitated devices. The J_{PHOTO} for equivalent planar bilayer cells (cells with the same amount of P3HT and PCBM than the interdigitated ones but in planar configuration) are indicated as horizontal lines for reference. For all the range of considered β , except for $\beta = 25$ nm, J_{PHOTO} for T = 80 nm is bigger than for T = 230 nm, even though T = 230 nm corresponds to the



FIG. 4. Maximum attainable photogenerated current density (J_{PHOTO}) as a function of β for the two pillar heights at which the absorbed light is maximum, T = 80 nm and T = 230 nm, and for $\gamma = 0.50$. The horizontal lines correspond to the J_{PHOTO} of the equivalent planar bilayer cells.

Downloaded 30 Sep 2013 to 193.144.16.69. This article is copyrighted as indicated in the abstract. Reuse of AIP content is subject to the terms at: http://jap.aip.org/about/rights_and_permissions

highest light absorption. We can also observe that as β increases the difference between J_{PHOTO} for T = 80 and for T = 230 nm also increases. This fact can be explained by the short exciton diffusion length of organic materials.

To this end, Figure 5 depicts the steady state exciton concentration map of an interdigitated cell for $\alpha = 250 \text{ nm}$, $\beta = 500$ nm, and T = 250 nm (a), and of its equivalent planar bilayer form (b). It can be seen that in the interdigitated cell excitons reach up to a certain depth in the P3HT, limited by the exciton diffusion length. In the planar equivalent cell, this limitation results in a low concentration gradient of excitons near the donor-acceptor interface, which leads to the low J_{PHOTO} observed. This argument supports the results that can be observed in Figure 4, where an improvement in J_{PHOTO} with respect to the bilayer cell can be observed only for β below 100 nm. A further examination of the concentration map in Figure 5(a) explains also why J_{PHOTO} decreases with increasing β : as the width of the pillar becomes bigger, less excitons are generated close to the lateral interface, where most of them dissociate.

Figure 6 shows the complete current density–voltage (J–V) curves for several β with $\gamma = 0.5$, and for the reference planar bilayer cell for the two thicknesses of interest (T = 80 and 230 nm). In this graph, we can see that the open circuit voltage (V_{OC}) of the different devices is in a range from 0.5 to 0.6 V. Another important parameter of solar cells, the short circuit current density (J_{SC}), shows a higher variation between cells, ranging from 0.25 to more than 4.5 mA/cm². These values of J_{SC} show a good correlation with the J_{PHOTO} displayed in Figure 4, especially for T = 80 nm. This indicates a high charge carrier collection efficiency and that this process is not strongly influenced by the geometrical characteristics of the interdigitated interface.

Figure 7 shows the J–V characteristics for T = 80 nm for several β and for different $\gamma = \alpha/\beta$. As in Figure 6, the V_{OC} for the different cells ranges from 0.5 to 0.6 V while the J_{SC}



FIG. 5. (a) Exciton concentration map of an interdigitated cell for $\beta = 500$ nm, $\gamma = 0.50$, and T = 250 nm, and (b) of its equivalent bilayer form.



FIG. 6. Current density-voltage (J–V) curves for several β with a fix $\gamma = 0.50$. The J-V curves of the reference planar bilayer cells for the two thicknesses of interest (T = 80 and 230 nm) have also been included.

values are distributed over a wider range and show a different trend for different β . For $\beta = 25$ nm, when γ increases J_{SC} increases in a similar proportion. However, for bigger β , the increase of J_{SC} with γ is less noticeable. A higher γ implies a higher ratio of P3HT, and therefore, more absorbed light. However, not in all cases, all this supplementary amount of absorbed light will be actually effective because of the short exciton diffusion length of P3HT.

From the J–V characteristics, we have also obtained the fill factor (FF), which is shown in Figure 8 as a function of $\gamma = \alpha/\beta$ for T = 80 nm and several β . We can see that while for $\beta = 25$ nm the FF decreases when γ increases, there are no significant changes for the rest of β when γ varies. If we compare the *FF* between the different β , we can observe that it decreases as the period of the nanostructures increases. This parameter is one of the most representing factors in the performance of a solar cell since it gives information on the charge transport and recombination mechanisms. The analysis of this parameter reveals that, in the case of study, the exciton diffusion is a more limiting factor than the charge transport, since when the exciton dissociation starts becoming a limitation (large β and γ) the FF decreases. These are in good agreement with our previous results



FIG. 7. J–V curves for T = 80 nm for several β and for different $\gamma = \alpha/\beta$.

Downloaded 30 Sep 2013 to 193.144.16.69. This article is copyrighted as indicated in the abstract. Reuse of AIP content is subject to the terms at: http://jap.aip.org/about/rights_and_permissions



FIG. 8. Fill factor (*FF*) as a function of $\gamma = \alpha/\beta$ for T = 80 nm and several β .

interpretations from the analysis of the J_{PHOTO} as a function of the geometrical parameters.

Table I shows the efficiency (η) corresponding to the devices referred to in the previous figures. We can observe that a bigger quantity of polymer does not always mean better results. Efficiencies are higher for T = 80 than for T = 230 nm for the same γ , and a higher γ only represents a noticeable increase in the efficiency for $\beta = 25$ nm. The increase in β causes a decrease in the efficiency leading to for β as big as $\beta = 500$ nm, the interdigitated cell efficiencies are lower than that of the equivalent planar bilayer. The maximum efficiency is achieved for $\beta = 25$ nm, T = 80 nm, and $\gamma = 0.75$, with an efficiency 3.6 times higher than the best planar bilayer reference device. This optimal planar cell is the equivalent, in amount of each organic material, for an interdigitated cell of T = 60 nm and $\gamma = 0.50$ and presents an efficiency of the 0.69%.

The results of our work show a good agreement with previous reported studies. Yang and Forrest¹⁷ reported that the absorption by the organic layers limits the optimal cell thicknesses to between 200 and 300 nm, a range that includes one of our optimized nanopillar height. They also achieved the highest external quantum efficiencies with an interdigitated network when the domain size was equal to or smaller than the exciton diffusion length. Meng et al.¹⁸ found that the optimal energy conversion efficiency can be achieved when the feature size is around 10 nm, a value close to its exciton diffusion length. In study of Kim et al.,¹⁹ they state that for a sufficient light absorption the active layer thickness should be greater than the 50% of the photon mean free path, which is the inverse of the absorption coefficient ($\sim 100 \text{ nm}$ for organic materials³). However, as in our results, extremely high pillars for a complete light absorption do not give the highest efficiencies.

The results of our study are also in good agreement with the reported experimental work in the introduction. Kim *et al.*⁶ reported an increase of 6.6 times in the efficiency of P3HT-C60 interdigitated devices if compared with a planar bilayer reference cell. Although this improvement factor is bigger than the one we obtain, they do not report details about their reference device. He *et al.*⁹ increased the effi-

TABLE I. Efficiencies of representative cells obtained from the J–V curves from Figures 6 and 7. These values should be compared with the maximum efficiency that have been obtained with a planar bilayer reference cell, which is $\eta = 0.69\%$.

	Cells efficiency (%)			
	$T = 80 \text{ nm},$ $\gamma = 0.25$	$T = 80 \text{ nm},$ $\gamma = 0.50$	$T = 230 \text{ nm},$ $\gamma = 0.50$	$T = 80 \text{ nm},$ $\gamma = 0.75$
$\beta = 25 \text{ nm}$	1.18	1.76	1.72	2.49
$\beta = 100 \mathrm{nm}$	0.85	0.95	0.81	0.95
$\beta = 500 \text{ nm}$	0.34	0.35	0.21	0.38
Best planar bilayer reference cell ($T = 60 \text{ nm}$)			0.69	

ciency 2.4 times with cells of 25×25 nm of nanorod size, 80 nm of nanorod height, and 50 nm of structure period, also in good agreement with our results. Furthermore, these authors also consider a wide range of nanorod sizes and they find the same trend for the J_{SC} and the efficiency as we do. Other experimental reported works also show a good correlation with our study such as Zheng *et al.*¹⁰ with two-fold efficiency increase in 20 nm diameter and 40 nm long nanopillars, and Wiedemann *et al.*¹¹ with an improvement of about 80% in 40 nm-width and 80 nm-periodicity structures.

IV. CONCLUSIONS

In this work, we analyze the influence of the nanostructured dissociation interface on the behavior of interdigitated heterojunction full organic solar cells. By means of finiteelement numerical modeling, we perform a systematic and complete analysis of light absorption, exciton diffusion and carrier transport, all in the same numerical framework, and for a 2D model of such cells. With this, we obtain their dependence on the cell geometrical parameters: pillar diameter and height, and nanostructure period. The donor and the acceptor materials of our cells are P3HT and PCBM, respectively. The aim of this study is to develop a model for structures of this kind that can be achieved via nanoporous anodic alumina templates, which can be used for its optimal design.

From the point of view of light absorption, results show that the maximum amount of light is absorbed only for two specific values of the nanopillar height, T = 80 nm and T = 230 nm, independently of the nanopillar diameter. This suggests that it is due to light interference effects. For these nanopillar heights, the maximum absorption is achieved by nanostructures with a period of $\beta = 25$ nm. By simulating the exciton diffusion process the maximum attainable photocurrent density (J_{PHOTO}) can be determined. Results show that to obtain a higher J_{PHOTO} than that of the better planar bilayer structure, small nanostructure periods, up to 100 nm, should be used. By examining the exciton diffusion maps, we can conclude that small nanopillars favor the excitons to reach the donor-acceptor interface and that the upper limit of the nanopillar height is directly related to the limited exciton diffusion length.

Finally, from the J–V characteristics, we obtain the overall efficiency of the cells. We observe that there is a direct correlation between the J_{PHOTO} and the cell efficiency, which indicates that the charge transport has a lower influence on the overall charge collection efficiency. Thus, in order to increase the efficiency, the exciton diffusion process is the key limiting factor and this process is better for the smaller nanopillar diameters. For $\beta = 25$ nm, the efficiency depends strongly on the ratio of the nanopillar diameter to the nanopillar period (γ) being bigger for higher values of it. Instead, if β values above 100 nm are considered, then the efficiency is essentially independent of the γ since the increase in light absorption is almost compensated by the exciton recombination losses. Best results have been achieved for $\beta = 25$ nm, T = 80 nm, and $\gamma = 0.75$, with an efficiency that is 3.6 times higher than the best planar bilayer reference device.

An optimal interdigitated structure should find a compromise between a high proportion of electron donor to increase light absorption and a small pillar diameter to ensure an extended D-A interface for exciton dissociation. Due to the short exciton diffusion length of organic materials, big nanopillars diameters will result in cells with an efficiency even lower than the one of planar bilayer equivalents. The method developed in this work can be useful to design optimal full organic solar cells, taking into account technological parameters and constraints.

ACKNOWLEDGMENTS

This work was supported by the Spanish Ministry of Science and Innovation (MICINN) under Grant Nos. TEC2009-09551 and TEC2012-34397, CONSOLIDER HOPE Project No. CSD2007-00007, and by the Catalan Authority under Project No. 2009SGR549.

- ¹M. A. Green, K. Emery, Y. Hishikawa, W. Warta, and E. D. Dunlop, Prog. Photovoltaics **20**, 12 (2012).
- ²J. R. Sheats, J. Mater. Res. **19**, 1974 (2004).
- ³P. Würfel, *Physics of Solar Cells: From Basic Principles to Advanced Concepts*, 2nd ed. (Wiley, Weinheim, 2009).
- ⁴Semiconducting Polymers: Chemistry, Physics and Engineering, edited by G. Hadziioannou and G. G. Malliaras (Wiley, Weinheim, 2007).
- ⁵H. Hoppe and N. S. Sariciftci, J. Mater. Res. **19**, 1924 (2004).
- ⁶J. S. Kim, Y. Park, D. Y. Lee, J. H. Lee, J. H. Park, J. K. Kim, and K. Cho, Adv. Funct. Mater. **20**, 540 (2010).
- ⁷F. Yang, M. Shtein, and S. R. Forrest, Nature Mater. 4, 37 (2005).
- ⁸S. Yu, C. Klimm, P. Schäfer, J. P. Rabe, B. Rech, and N. Koch, Org. Electron. **12**, 2180 (2011).

- ⁹X. M. He, F. Gao, G. L. Tu, D. G. Hasko, S. Hüttner, N. C. Greenham, U. Steiner, R. H. Friend, and W. T. S. Huck, Adv. Funct. Mater. **21**, 139 (2011).
- ¹⁰Y. Zheng, R. Bekele, J. Ouyang, and J. Xue, Org. Electron. **10**, 1621 (2009).
- ¹¹W. Wiedemann, L. Sims, A. Abdellah, A. Exner, R. Meier, K. P. Musselman, J. L. MacManus-Driscoll, P. Müller-Buschbaum, G. Scarpa, P. Lugli, and L. Schmidt-Mende, Appl. Phys. Lett. 96, 263109 (2010).
- ¹²S. Baek, J. B. Park, W. Lee, S. H. Han, J. Lee, and S. H. Lee, New J. Chem. **33**, 986 (2009).
- ¹³T. Kim, D. Kurunthu, J. J. Burdett, and C. J. Bardeen, J. Appl. Phys. 108, 033114 (2010).
- ¹⁴A. Santos, P. Formentín, J. Pallarés, J. Ferré-Borrull, and L. F. Marsal, Sol. Energy Mater. Sol. Cells 94, 1247 (2010).
- ¹⁵R. Palacios, P. Formentín, T. Trifonov, M. Estrada, R. Alcubilla, J. Pallarés, and L. F. Marsal, Phys. Status Solidi (RRL) 2, 206 (2008).
- ¹⁶H. Masuda, H. Yamada, M. Satoh, H. Asoh, M. Nakao, and T. Tamamura, Appl. Phys. Lett. **71**, 2770 (1997).
- ¹⁷F. Yang and S. R. Forrest, ACS Nano 2, 1022 (2008).
- ¹⁸L. Meng, Y. Shang, Q. Li, Y. Li, X. Zhan, Z. Shuai, R. G. E. Kimber, and A. B. Walker, J. Phys. Chem. B **114**, 36 (2010).
- ¹⁹J. Kim, K. Kim, S. H. Ko, and W. Kim, Sol. Energy Mater. Sol. Cells 95, 3021 (2011).
- ²⁰See http://www.comsol.com for COMSOL Inc.), MA, USA.
- ²¹C. Defranoux, "Spectroscopic ellipsometry applied to solar cell characterization," SOPRA Application note, see http://www.sopra-sa.com/.
- ²²R. A. Synowicki, Thin Solid Films **313**, 394 (1998).
- ²³L. A. A. Pettersson, S. Ghosh, and O. Inganäs, Org. Electron. 3, 143 (2002).
- ²⁴A. D. Rakic, Appl. Opt. **34**, 4755 (1995).
- ²⁵V. Andersson, K. Tvingstedt, and O. Inganäs, J. Appl. Phys. **103**, 094520 (2008).
- ²⁶M. Born and E. Wolf, *Principles of Optics: Electromagnetic Theory of Propagation, Interference and Diffraction of Light*, 7th ed. (Cambridge University Press, Cambridge, 1999) p. 10.
- ²⁷S. Cook, A. Furube, R. Katoh, and L. Han, Chem. Phys. Lett. **478**, 33 (2009).
- ²⁸G. F. Burkhard, E. T. Hoke, S. R. Scully, and M. D. McGehee, Nano Lett. 9, 4037 (2009).
- ²⁹C. J. Brabec, Sol. Energy Mater. Sol. Cells 83, 273 (2004).
- ³⁰G. Dennler, K. Forberich, M. C. Scharber, C. J. Brabec, I. Tomiš, K. Hingerl, and T. Fromherz, J. Appl. Phys. **102**, 054516 (2007).
- ³¹L. A. A. Pettersson, L. S. Roman, and O. Inganäs, J. Appl. Phys. 86, 487 (1999).
- ³²T. Kirchartz, B. E. Pieters, K. Taretto, and U. Rau, J. Appl. Phys. 104, 094513 (2008).
- ³³J. A. Barker, C. M. Ramsdale, and N. C. Greenham, Phys. Rev. B 67, 075205 (2003).
- ³⁴Y. Shang, Q. Li, L. Meng, D. Wang, and Z. Shuai, Theor. Chem. Acc. 129, 291 (2011).
- ³⁵H. Wang, H. Y. Wang, B. R. Gao, L. Wang, Z. Y. Yang, X. B. Du, Q. D. Chen, J. F. Song, and H. B. Sun, Nanoscale 3, 2280 (2011).
- ³⁶F. Monestier, J. J. Simon, P. Torchio, L. Escoubas, F. Flory, S. Bailly, R. Bettignies, S. Guillerez, and C. Defranoux, Sol. Energy Mater. Sol. Cells **91**, 405 (2007).