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Influence of optical interference and carrier lifetime on the short circuit current density of organic bulk heterojunction solar cells

You Hai-Long(游海龙)[a)b)] and Zhang Chun-Fu(张春福)[c)†]

[a)] *School of Microelectronics, Xidian University, Xi'an* 710071, *China*
[b)] *Key Laboratory of Ministry of Education for Wide Band-Gap Semiconductor Materials and Devices, Xi'an* 710071, *China*
[c)] *Department of Electrical and Computer Engineering, National University of Singapore, Singapore* 117576

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Based on simple analytical equations, short circuit current density (J_{SC}) of the organic bulk heterojunction solar cells has been calculated. It is found that the optical interference effect plays a very important role in the determination of J_{SC}; and obvious oscillatory behaviour of J_{SC} was observed as a function of thickness. At the same time, the influence of the carrier lifetime on J_{SC} also cannot be neglected. When the carrier lifetime is relatively short, J_{SC} only increases at the initial stage and then decreases rapidly with the increase of active layer thickness. However, for a relatively long carrier lifetime, the exciton dissociation probability must be considered, and J_{SC} behaves wave-like with the increase of active layer thickness. The validity of this model is confirmed by the experimental results.

Keywords: organic solar cells, short circuit current density, optical interference, carrier lifetime
PACC: 8630J, 7360F, 7220J

1. Introduction

Solar cells based on organic materials are of tremendous interest[1−7] because of their attractive properties such as flexibility, ease of fabrication, low materials and energy budget. The efficiency has improved steadily in the past decade, especially for bulk heterojunciton (BHJ) solar cells, reaching nowadays power conversion efficiency (PCE) of more than 6%. Usually, BHJ organic solar cells consist of a blend layer of conjugated polymer as electron donor (D) and fullerene as electron accepter (A). For a well blended layer, the length scale of D and A phases is smaller than exciton diffusion length (typically less than 10 nm), so that most of the generated excitons can be dissociated into free carriers. The dissociation probability is not a constant, depending on many factors such as electric field and temperature.[2] After dissociation, the generated free electrons and holes are transported in fullerene towards the cathode and in polymer towards the anode, respectively. And then the photocurrent is generated.

In order to achieve higher PCE, a lot of works have been done, such as the optimization of D/A weight ratio[4] and nanoscale morphology.[5] It is believed that increasing the light harvesting ability of the blend layer is an effective method to increase PCE. Some optical models[8−10] have been built to optimize the blend layer thickness. However, only optimizing the thickness for better light absorption is difficult to improve PCE. This is because the PCE depends on not only the light absorption, but also the exciton dissociation and charge collection. When the thickness is increased to optimize the light absorption, the electric field in the blend layer will decrease, which will lower the exciton dissociation probability[2] and charge collection[10] simultaneously. As a result, J_{SC} may become low although the thickness has been optimized for light absorption. Then, to obtain a higher J_{SC}, both the optical and the electric properties should be considered at the same time. In previous works,[11,12] Lacic and Inganas[11] and Monestier *et al*[12] studied the characteristic of J_{SC}. However, they neglected the influence of exciton dissociation probability which is very important for organic solar cells.[13] Another researchers, Koster *et al*[14] considered this parameter, but they neglected the optical interference effect in organic solar cell, which is a ba-

†Corresponding author. E-mail: zhangchunfu@nus.edu.sg

sic property for the very thin organic film. All the above works are based on the numerical method, and it is not easy to solve the equations and understand the direct influence of various parameters on the J_{SC}.

In this work, a model for predicting J_{SC} is proposed only based on very simple analytical equations. It is found that the optical interference effect has a very important influence on J_{SC} and the carrier lifetime also cannot be neglected. Generally, when the carrier lifetime is long enough, dissociation probability will play a very important role for the thicker active layer. And J_{SC} will behave wave-like with the variation of active layer thickness. When the carrier lifetime is too short, the accumulation of charges will appear near the electrode and J_{SC} will increase at the initial stage and then decrease rapidly with the increase of active layer thickness. The validity of our model is confirmed by the experimental results.

2. Theory

2.1. Exciton generation

The active layer in organic solar cell will absorb the light energy when it is propagating through this layer. How much energy can be absorbed depends on the complex index of refraction $\bar{n} = n + \mathrm{i}\kappa$ of the materials. At position z in the organic film (Fig.1(a)), the time average of the energy dissipated per second for a given wavelength λ of incident light can be calculated by[9]

$$Q(z,\lambda) = \frac{1}{2} c\varepsilon_0 \alpha_j n \left|\bar{E}(z)\right|^2, \tag{1}$$

where c is the vacuum speed of light, ε_0 is the permittivity of vacuum, n is the real index of refraction, α is absorption coefficient, $\alpha = \dfrac{4\pi\kappa}{\lambda}$, $E(z)$ is the electrical optical field at point z. $Q(z,\lambda)$ has the unit of $W/\left(\mathrm{m}^2 \cdot \mathrm{m}\right)$. Assuming that every photon will generate one exciton, the exciton generation rate at position z in the material is given by

$$G(z,\lambda) = \frac{Q(z,\lambda)}{h\gamma} = \frac{\lambda}{hc} Q(z,\lambda), \tag{2}$$

where h is Planck's constant, γ is the frequency of incident light. The total excitons generated in the material at position z in solar spectrum is calculated by using

$$G(z) = \int_{300}^{800} G(z,\lambda)\mathrm{d}\lambda, \tag{3}$$

Here the integration is performed from 300 to 800 nm, because beyond this range, only very weak light can be absorbed by P3HT: PCBM blend. In inorganic solar cells,[15] $Q(z,\lambda)$ is usually modeled by

$$Q(z,\lambda) = \alpha I_0 \mathrm{e}^{-\alpha z}, \tag{4}$$

where I_0 is the incident light intensity. Here, the optical interference of the materials is neglected. But in organic solar cells, the active layers are so thin compared to the wavelength that the interference effects cannot be neglected, which will be discussed in the next section.

2.2. Optical model

In order to obtain the distribution of electromagnetic field in a multilayer structure, the optical transfer-matrix theory is one of the most elegant methods, which was introduced by Heavens,[16] and was first applied in organic solar cells by Pettersson *et al.*[8] In this method, the light is treated as a propagating plane wave, which will be transmitted to and reflected on the interface. As shown in Fig.1, organic solar cells usually consist of a stack of layers. Every layer can be treated as a smooth, homogeneous layer and described by the same complex index of refraction $\bar{n} = n + \mathrm{i}\kappa$. The optical electrical field at any position in the multilayer can be decomposed into two parts, the upstream E^+ and a downstream component E^- as shown in Fig.1.

According to Fresnel theory, the complex reflection and transmission coefficients for a propagating plane wave along the surface normal between two adjacent layers i and j are

$$r_{jk} = \frac{\bar{n}_j - \bar{n}_k}{\bar{n}_j + \bar{n}_k}, \tag{5a}$$

$$t_{jk} = \frac{2\bar{n}_j}{\bar{n}_j + \bar{n}_k}, \tag{5b}$$

where r_{jk} and t_{jk} are reflection coefficient and transmission coefficient, $\bar{n}_j$ and $\bar{n}_k$ are complex index of refraction for layer j and layer k respectively. So the interface matrix between the two adjacent layers is simply described as

$$\boldsymbol{I}_{jk} = \frac{1}{t_{jk}} \begin{bmatrix} 1 & r_{jk} \\ r_{jk} & 1 \end{bmatrix} = \begin{bmatrix} \dfrac{\bar{n}_j + \bar{n}_k}{2\bar{n}_j} & \dfrac{\bar{n}_j - \bar{n}_k}{2\bar{n}_j} \\ \dfrac{\bar{n}_j - \bar{n}_k}{2\bar{n}_j} & \dfrac{\bar{n}_j + \bar{n}_k}{2\bar{n}_j} \end{bmatrix}, \tag{6}$$

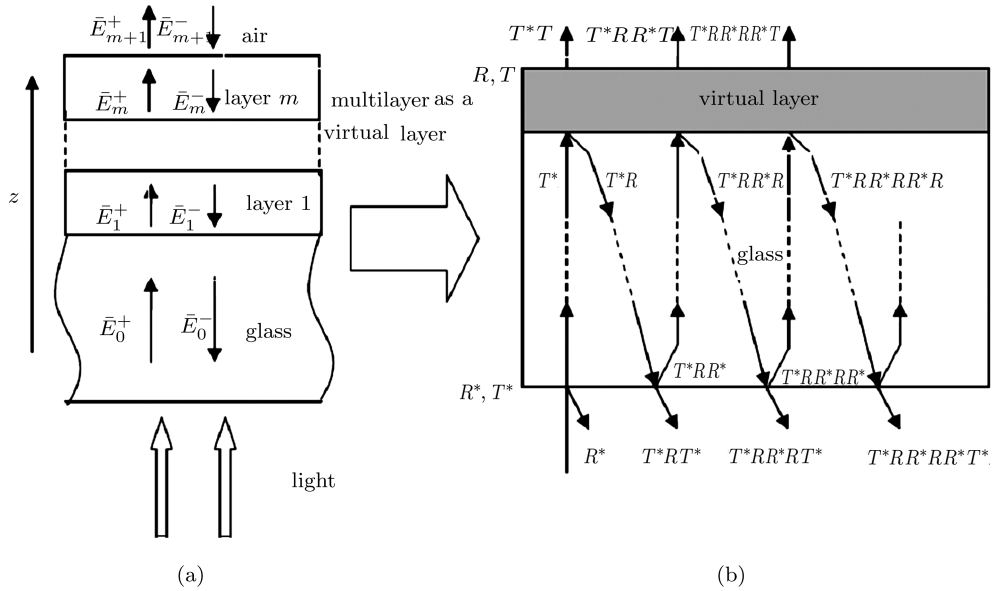


Fig.1. BHJ organic solar cells consisting of multilayer structure between air and a semi-infinite substrate. The light is incident on the cells from the downside; (a) the optical electric field in every layer can be denoted as an upstream optical electric field E^+ and a downstream optical electric field E^-, (b) the multilayer is treated as a virtual layer.

When light is travelling in layer j with a thickness d, the phase changing can be described by the layer matrix (phase matrix)

$$L_j = \begin{bmatrix} \mathrm{e}^{-\mathrm{i}\beta_j} & 0 \\ 0 & \mathrm{e}^{\mathrm{i}\beta_j} \end{bmatrix}, \tag{7}$$

where $\beta_j = \dfrac{2\pi\bar{n}_j}{\lambda}$ is phase change the wave experiences as it traverses in layer j. For optical electric field in the final layer and the substrate (subscript 0), there is a relation

$$\begin{bmatrix} \bar{E}_0^+ \\ \bar{E}_0^- \end{bmatrix} = S \begin{bmatrix} \bar{E}_{m+1}^+ \\ \bar{E}_{m+1}^- \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{12} & S_{22} \end{bmatrix} \begin{bmatrix} \bar{E}_{m+1}^+ \\ \bar{E}_{m+1}^- \end{bmatrix} = \left(\prod_{v=1}^{m} I_{(v-1)v} L_v \right) \bullet I_{m(m+1)}. \tag{8}$$

Because in the final layer $\bar{E}_{m+1}^-$ is 0, it can be derived that the complex reflection and transmission coefficients for the whole multilayer are

$$r = \frac{\bar{E}_0^-}{\bar{E}_0^+} = \frac{S_{21}}{S_{11}}, \tag{9a}$$

$$t = \frac{\bar{E}_{m+1}^+}{\bar{E}_0^+} = \frac{1}{S_{11}}. \tag{9b}$$

In order to gain the electric field $E_j(z)$ in layer j, S is divided into two parts,

$$S = S'_j L_j S''_j \tag{10}$$

with

$$S'_j = \left(\prod_{v=1}^{j-1} I_{(v-1)v} L_v \right) \bullet I_{j(j-1)}, \tag{11a}$$

and

$$S''_j = \left(\prod_{v=j+1}^{m} I_{(v-1)v} L_v \right) \bullet I_{m(m+1)}, \tag{11b}$$

at the lower interface in layer j, the upstream optical electric field is denoted as

$$\bar{E}_j^+ = t_j^+ \bullet \bar{E}_0^+ = \frac{S''_{j11}}{S'_{j11} S''_{j11} + S'_{j12} S''_{j21} \mathrm{e}^{\mathrm{i}2\beta_j}} \bar{E}_0^+. \tag{12}$$

Similarly, at the upper interface in layer j, the downstream optical electric field is

$$\bar{E}_j^- = t_j^- \bullet \bar{E}_0^+ = \frac{S''_{j21}}{S''_{j11}} \mathrm{e}^{\mathrm{i}2\beta_j} \bar{E}_j^+. \tag{13}$$

The optical electric field $\bar{E}_j(z)$ at any position z in layer j is the sum of upstream part $\bar{E}_j^+(z)$ and downstream part $\bar{E}_j^-(z)$:

$$\bar{E}_j(z) = \bar{E}_j^+(z) + \bar{E}_j^-(z) = (t_j^+ \mathrm{e}^{\mathrm{i}\beta_j} + t_j^- \mathrm{e}^{-\mathrm{i}\beta_j}) \bar{E}_0^+. \tag{14}$$

2.3. Light loss due to the substrate

Because the glass substrate is very thick compared to wavelength (usually 1 mm $\gg$ wavelength), then the interference effect of substrate can be neglected. Here only the correction of the light intensity

at the air/substrate and substrate /multilayer interfaces is made. As shown in Fig.1(b), the multilayer can be treated as a virtual layer whose complex reflection and transmission coefficients can be calculated using above equations. Then the irradiance to the multilayer is

$$I_{\mathrm{g}} = T\left(\sum_{i=0}^{\infty}(R^{*}R)^{i}\right)I_{0} = \frac{1-R^{*}}{1-RR^{*}}I_{0}, \tag{15}$$

I_{g} is described as

$$I_{\mathrm{g}} = \frac{1}{2}c\varepsilon_{0}n_{\mathrm{g}}\left|E_{0}^{+}\right|^{2}. \tag{16}$$

It can be derived that

$$\left|E_{0}^{+}\right| = \sqrt{\frac{2(1-R^{*})*I_{0}}{\varepsilon_{0}cn_{\mathrm{g}}(1-RR^{*})}}. \tag{17}$$

2.4. Free carrier generation

When the excitons are generated, not all of them can be dissociated into free carriers. The dissociation probability depends on the field and temperature. Recently, the dissociation probability has been taken into consideration in organic solar cells.[2,14] The geminate recombination theory, first introduced by Onsager and refined by Braun later, gives the probability of electron–hole pair dissociation,

$$P(F,T) = \frac{k_{\mathrm{D}}(F)}{k_{\mathrm{D}}(F)+k_{\mathrm{F}}}, \tag{18}$$

where T is the temperature and F is the electric field, k_{F} is the decay rate to the ground state and k_{D} is the dissociation rate of a bound pair. Braun gave the simplified form for dissociation rate

$$k_{\mathrm{D}}(E) = k_{\mathrm{R}}\mathrm{e}^{-U_{\mathrm{B}}/k_{\mathrm{B}}T}\left[1+b+\frac{b^{2}}{3}+\ldots\right], \tag{19}$$

where a is the initial distance of a given electron–hole pair, U_{B} is the exciton binding energy described as

$$U_{\mathrm{B}} = \frac{q^{2}}{4\pi\varepsilon a}, \tag{19a}$$

and

$$b = \frac{q^{3}F}{8\pi\varepsilon k_{\mathrm{B}}T^{2}}; \tag{19b}$$

k_{B} is Boltzmann constant, ε is the dielectric constant for exciton in the material. And k_{R} is a function of the carrier recombination and the initial distance of a bounded exciton. For simplicity, we treat k_{R} as a constant. So the dissociation probability of a bound exciton only depends on the electric field F.

2.5. J_{SC} and the blend layer thickness

J_{SC} is determined by the number of carriers collected by the electrode in the period of their lifetime τ under short circuit condition. If the active layer thickness L is shorter than the electron and hole drift lengths (which is the product of carrier mobility μ, the electric field F and the carrier lifetime τ) or in other word, the lifetime of both types of carriers exceeds their transit time (case I in Fig.2(a)), all generated free carriers can be collected by the electrodes.

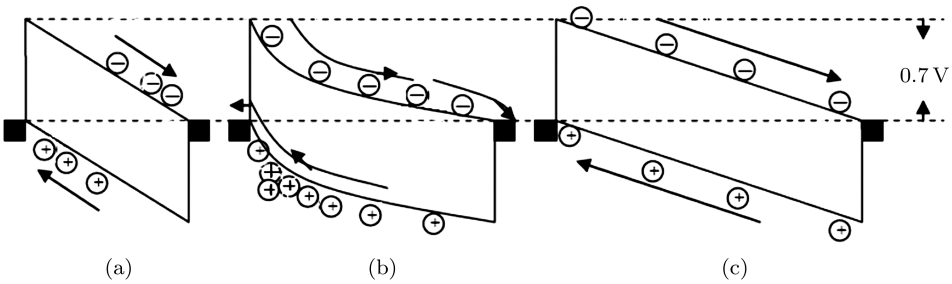


Fig.2. Energy band diagrams of an organic solar cell subject to irradiation under short circuit condition. The built-in voltage is 0.7 V determined by the electrodes. (a) Device thickness is shorter than both hole and electron drift lengths. (b) Device thickness is longer than hole drift length but shorter than electron drift length. (c) Device thickness is longer than both hole and electron drift lengths.

Considering the dissociation probability, we can write J_{SC} as

$$J_{\mathrm{SC}} = qP(F,T)GL, \tag{20}$$

where G is the average exciton generation rate in the active layer.

If L is longer than drift lengths of electrons and holes, that is to say, the lifetime (τ) of both types of carriers is smaller than the transit time, the carrier will be accumulated in the active layer. In a

steady state, the short circuit current density will follow Ohm's law,[17] considering the dissociation rate

$$J_{\mathrm{SC}} = qP(F,T)G(\mu_{\mathrm{e}}\tau_{\mathrm{e}} + \mu_{\mathrm{h}}\tau_{\mathrm{h}})F = qP(F,T)G(\mu_{\mathrm{e}}\tau_{\mathrm{e}} + \mu_{\mathrm{h}}\tau_{\mathrm{h}})V_{\mathrm{bi}}/L, \quad (21)$$

where V_{bi} is the built-in potential which is usually determined by the difference between cathode and anode work functions. This is case III described in Fig.2(c).

Between case I and case III, it is more complicated. Under this condition, L is only longer than the drift length of one type of carrier. For P3HT: PCBM based organic solar cells, the mobility of holes and electrons in P3HT: PCBM (1:1 by weight) layer is $2 \times 10^{-8}\mathrm{m}^2 \cdot \mathrm{V}^{-1} \cdot \mathrm{s}^{-1}$ and $3 \times 10^{-7}\mathrm{m}^2 \cdot \mathrm{V}^{-1} \cdot \mathrm{s}^{-1}$, respectively.[16] Because the hole mobility is one order lower than the electron mobility, the holes are easy to accumulate in the active layer, thus making the electric field nonuniform. In order to enhance the extraction of holes, the electric field increases near the anode. And on the other hand, in order to diminish the extraction of electrons, the electric field is decreased near the cathode. The electric field is modified until the extraction of holes equal to the extraction of electrons. Goodman and Rose[18] studied this case and gave an equation for the photocurrent. Consider the dissociation rate,

$$J_{\mathrm{sc}} = qP(F,T)GL(1+c) \times \frac{-c + (c^2 + 4(1-c)V\mu_{\mathrm{h}}\tau_{\mathrm{h}}/L^2)^{\frac{1}{2}}}{2(1-c)}, \quad (22)$$

where $c = \dfrac{\mu_{\mathrm{h}}\tau_{\mathrm{h}}}{\mu_{\mathrm{e}}\tau_{\mathrm{e}}}$ is the drift length ratio of holes and electrons. When $c \ll 1$, the equation is simplified to

$$J_{\mathrm{sc}} = qP(F,T)G(\mu_{\mathrm{h}}\tau_{\mathrm{h}})^{1/2}V^{1/2}. \quad (23)$$

3. Results and discussion

3.1. Exciton generation profile in the active layer

All the optical constants (n,k) of ITO, PEDOT:PSS, active layer and Al electrode[12,19] are input into our program and then the exciton generation rate in organic solar cells can be calculated.

If the interference effect is neglected, the exciton generation rate will decrease with increasing thickness of the active layer as described in Eq.(4), which will make the corresponding average exciton generation rate (total exciton generation rate divided by the thickness) become smaller. However, when the optical interference effect is considered, the modulation effect of average exciton generation rate with the variation of thickness is very clear as shown in Fig.3.

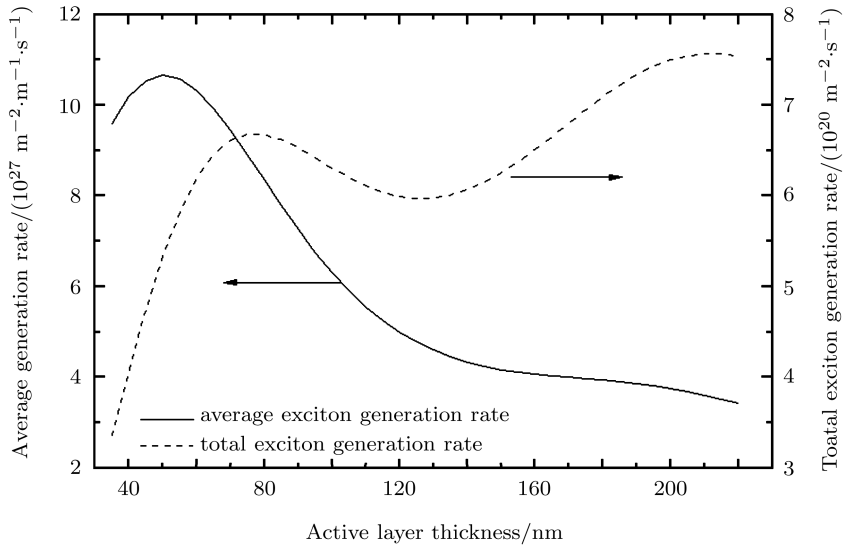


Fig.3. The calculated exciton generation rate in the active layer when considering the optical interference effect.

At the initial stage, the average exciton generation rate increases with increasing thickness of the active layer. This is because the first light peak will not appear in the active layer due to the interference effect when the film is very thin. With the increase of the active layer thickness, the first light peak approaches and enters the active layer so that the average generation rate becomes larger and larger. With further increase of the active layer thickness, the average generation rate begins to decrease although other light peaks enter the active layer. This is because for a thicker film, the thickness of the active layer will dominate the generation rate. This evolution of exciton generation is plotted in Fig.4 for the 500 nm light wavelength.

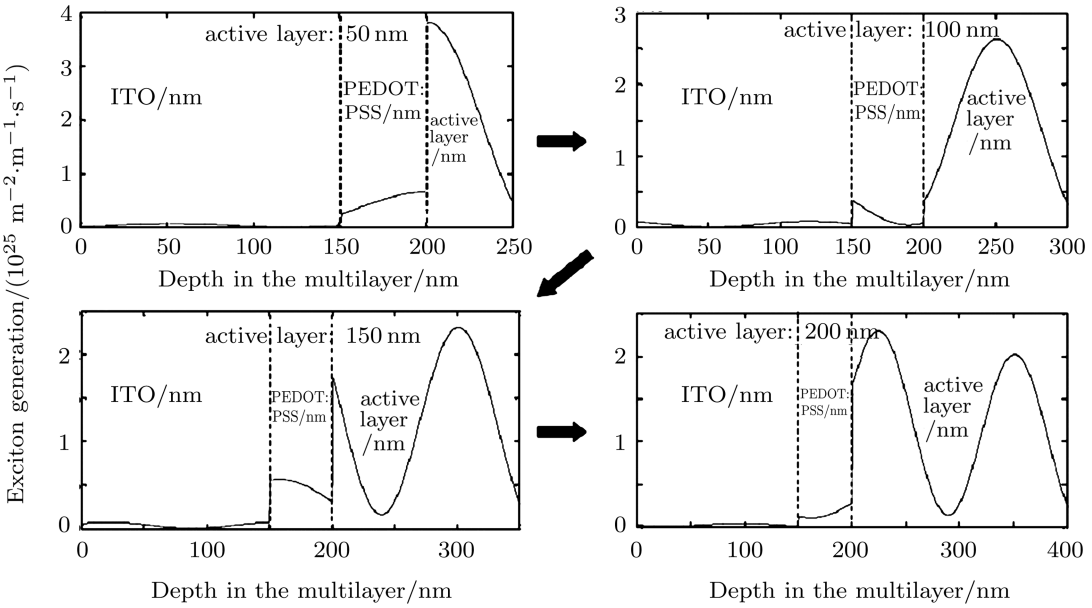


Fig.4. Evolution of exciton generation in the active layer. The wavelength is 500 nm. It can be seen that with the increase of thickness, the first peak enters the active layer, making the average exciton generation rate become large. For very thick film, although other peaks can enter the active layer, the absolute value for the peaks will become small, which will lead to the decrease of average exciton generation rate.

3.2. J_{SC} and the active layer thickness

Based on the calculated exciton generation rate, it is very easy to predict J_{SC} when the drift lengths of both carriers are larger than the blend layer thickness. If all the generated excitons can be dissociated into free carriers, and then collected by the electrodes, J_{SC} should be proportional to the total exciton generation rate and behaves wave-like as shown in Fig.5 (solid line).

Monestier *et al*[12] have found this trend based on P3HT:PCBM systems. In the experiment, the active layer thickness is varied from a few tens of nanometres to 215 nm. When the thickness is 70 nm, J_{SC} reaches the maximum value, and then followed by a little decrease until 140 nm. When the thickness is increased further, J_{SC} increases again. Unfortunately, there is an obvious deviation between the prediction and the experimental results, especially in the thick film as shown in Fig.5 (solid line). Obviously, the assump-

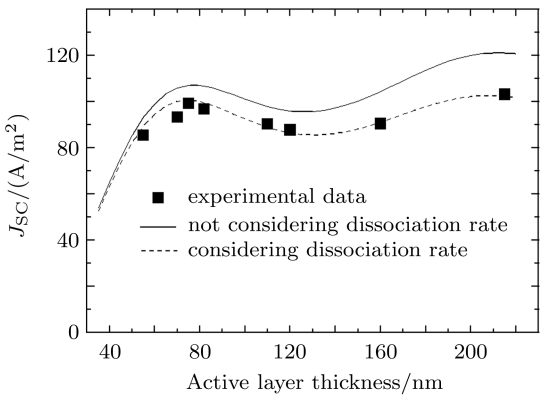


Fig.5. Long carrier lifetime condition: both carrier lifetimes are always longer than transient time. Short circuit current densities are predicted with and without considering dissociation probability; experimental data are taken from Ref.[12].

tion that the dissociation probability is unity is not correct. The influence of dissociation probability on J_{SC} must be considered.

In the previous work, Mihailetchi *et al*[20] exactly predicted photocurrent of P3HT: PCBM solar cells by assuming the same e–h separation distance a and decay frequency (k_F). By fitting the experimental data, they obtained the e–h separation distance of $a = 1.8\,\text{nm}$, room temperature bound pair decay frequency of $k_F \approx 2 \times 10^4 \text{s}^{-1}$ for a 120 nm active layer. We use the same data and derive the parameter $k_R = 3.9662 \times 10^8 \text{S}^{-1}$ (Eq.(19)). Then the dissociation rate can be calculated according to Section 2.4. The results are shown in Fig.6. Obviously, the exciton dissociation probability becomes lower with the increase of the active layer thickness. Using the results to correct J_{SC}, another J_{SC} curve is obtained and shown in Fig.5 (dashed line). It can be seen that the predicted J_{SC} is exactly in accordance with the experimental results. This confirms the validity of our model. In the previous work, Monestier *et al*[12] modelled J_{SC}, and found that the predicted J_{SC} is larger than the experimental data, especially for the thickness larger than 180 nm. They attributed this to the thickness dependence of optical constants. Here, according to our model, it is found that the deviation should come from the low exciton dissociation probability for a thick active layer.

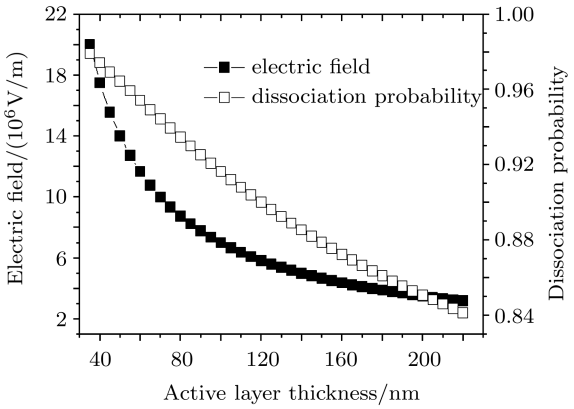


Fig.6. Variations of electric field and exciton dissociation probability with the active layer thickness.

We have predicted J_{SC} precisely for the long enough carrier lifetime case. However, for organic solar cells, the performance is very sensitive to the process and experimental conditions. This may make the carrier lifetime relative short. For P3HT:PCBM system, the hole mobility is one order lower than the electron mobility, then holes are easy to accumulate in the active layer and limit the photocurrent. By tuning the parameters to fit the experimental results,[21] the best fitting curve is obtained when the average hole lifetime τ is 6.2×10^{-7}s and dissociation probability is unity, as shown in Fig.7. Here, we notice that the dissociation probability is unity. This may be due to the defects. A short lifetime τ may imply that there are more defects. These defects can increase the exciton dissociation probability.[22] At the same time, the transport process becomes the dominated limit for J_{SC}, and the exciton dissociation process becomes relative unimportant. Then it seems that the assumption of exciton dissociation rate being unity can satisfy the requirement of the prediction. In Fig.7, we can see that there are two parts in the fitting curve. The left part is determined by Eq.(20). In this region, both carrier lifetimes are longer than transient time. The right part is determined by Eq.(22). In this region, hole lifetime is shorter than transit time and electron lifetime is longer than it.

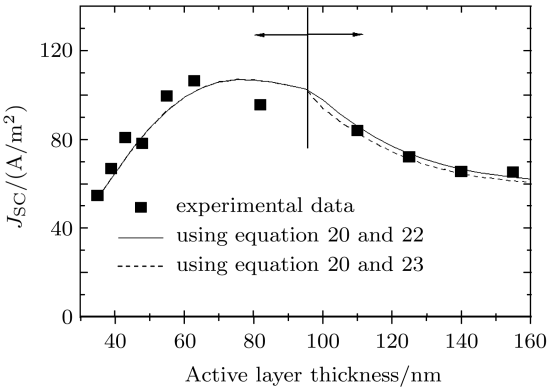


Fig.7. J_{SC} as a function of active layer thickness ,for short hole carrier lifetime condition. Left arrow: carrier lifetime is longer than transient time; right arrow: carrier lifetime is shorter than transient time, and hole lifetime is 6.2×10^{-7} s, and electron lifetime is 1×10^{-6} s. Experimental data are taken from Ref.[21].

If we assume that the drift length ratio of hole and electron $\left(c = \dfrac{\mu_h \tau_h}{\mu_e \tau_e}\right)$ is very small, then Eq.(23) can be used to predict J_{SC}. As shown in Fig.7 (dotted line), it can predict J_{SC} very well.

4. Conclusion

In summary, in this work the exciton generation rate is calculated by taking into account the optical interference. Based on the calculated exciton genera-

tion, the dependence of J_{SC} on the device thickness is analysed and compared with experimental data. Briefly, because of the optical interference effect, the total exciton generation rate does not increase monotonically with the increase of the active layer thickness, but behaves wave-like, which induces the corresponding variation of J_{SC}. The carrier lifetime also influence J_{SC} greatly. When the carrier lifetime is long enough, dissociation probability will play a very important role for a thicker active layer. J_{SC} will behave wave-like with the variation of active layer thickness. When the carrier lifetime is too short (drift length is smaller than device thickness), accumulation of charges will appear near the electrode and J_{SC} will increase at the initial stage and then decrease rapidly with the increase of active layer thickness. The agreement between the predictions and the experimental results confirms the validity of the proposed model.

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