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Quantum efficiency of exciton-to-charge generation in organic photovoltaic devices

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We present an analysis of the internal monochromatic quantum efficiency of photovoltaic devices based on polymer and polymer/fullerene thin films. A quantum efficiency of exciton-to-charge generation is defined as the external monochromatic quantum efficiency normalized to the absorption in the active materials of the device. An upper limit of the efficiency can be determined, and results show that much of the light is absorbed in photoactive layers of the device, whereas only a fraction of the generated excitons is converted to charge carriers and can be collected as photocurrent. © 2001 American Institute of Physics. [DOI: 10.1063/1.1359425]

I. INTRODUCTION

One of the most important parameters when studying the performance of photodiodes or solar cells is the quantum efficiency. The quantum efficiency is generally defined as the ratio of the number of collected charge carriers to the number of incident photons at the device, and is named external quantum efficiency (EQE) or sometimes incident photon to current efficiency (IPCE). One advantage of analyzing the EQE rather than photocurrent is that effects due to the spectral shape of the incident light, due to light source or measurement equipment, are removed and the true response of the device is obtained. Here it must also be understood that a possible intensity dependence of the photocurrent is properly accounted for. Understanding the reasons for obtaining a specific quantum efficiency of a device is essential to obtain fundamental understanding and thus to improve the device performance.

Much work has been directed towards the intrinsic optical and electronic properties of the active materials, which are tacitly assumed to be the most obvious and important factors to affect the efficiency of the photodiode.^{1–3} Nevertheless, the configuration and geometry of the device are also important for the efficiency;^{4,5} an improper combination of materials in the device, or geometries where light is absorbed at less favorable position in the photodiode, will ruin performance. For instance, a very thick active layer can, due to internal filter effects, reduce the efficiency of the device; photons can be absorbed in or reflected at the electrode on the illumination side of the device, and thereby decrease the efficiency of the device. Inversely, the layer might not be thick enough to absorb impinging light and we therefore lose efficiency since most light is reflected and/or transmitted. This will result in that few of the incident photons are absorbed in the active part of the device. However, in all these cases it is still possible that the internal generation process in the active part of the device is very efficient. Thus the main objective in the design of photodiodes and solar cells is to

build devices in which the absorption of the incident light will be maximized in the active parts of the device and minimized in the nonactive parts. In this way we may increase the efficiency of the device. Accordingly, information about the internal operation of the devices is required. One way to express this is through an internal quantum efficiency (IQE), which commonly has been used for inorganic semiconductor photodiodes or solar cells.⁶ In the IQE external optical effects, such as the reflectance of the cell, should be compensated for. In contrast to the conventional devices from inorganic semiconductors, active layers in organic photodiodes are usually thin compared to the wavelength of the incident light. Moreover, a highly reflecting (metal) interface is part of the device, resulting in a much more complex distribution of the optical electric field inside the device. Thus not only the total absorptance of the device must be considered, but also the absorptance of the individual layers. For this reason reflections and interference inside the device must be accounted for in an appropriate expression for an IQE.

In contrast to most inorganic semiconductors the generation of charge carriers in molecular semiconductors is a secondary process.^{7,8} In the molecular semiconductor bound electron-hole pairs, excitons are created by absorption of light. Charge collection requires dissociation of these excitons, which typically occur as a result of interaction of the excitons with acceptors, interfaces, impurities or defects, or in high electrical fields. One of the most important limiting factors for the efficiency of these materials at this time is the short exciton diffusion length.^{4,9} Conjugated molecules usually have a high absorption coefficient ($\approx 10^5 \text{ cm}^{-1}$) in the visible at the $\pi-\pi^*$ transition and thereby a short penetration depth of light. Hence it is possible to have a large absorptance using thin layers of the materials (typically 10–2000 nm), which is important because of the low charge mobility of the materials. Photodiodes based on organic molecules also have the advantage of being easy to process and give the opportunity to produce large area devices, and can be anticipated to result in low-cost manufacturing.

In this article we derive an explicit relationship for absorptance of the individual layers in a multilayer configuration at oblique incidence. An expression for the quantum

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efficiency of exciton-to-charge generation based on the experimental efficiency of a device is given. The so-defined layer internal efficiency takes the optical absorption in the photoactive layers into account and is used in the study of some devices. We have determined the optical constants within the active spectrum range, for all materials of the devices, by using spectroscopic ellipsometry.

II. THEORY

A. Internal quantum efficiency of a thin film photovoltaic device

The monochromatic quantum efficiency (EQE) of a photodiode or photovoltaic device is defined as the ratio of the number of generated and collected charge carriers to the number of incident photons as

$$\eta_{\text{EQE}} = \frac{J_{\text{SC}}/q}{I_0/h\nu} = \frac{J_{\text{SC}}/q}{N}, \quad (1)$$

where J_{SC} is the photogenerated short-circuit current density (A/m^2), q is the elementary charge (C), I_0 is the incident intensity (W/m^2), and $h\nu$ is the energy of the incident light (J), which means that $N = I_0/h\nu$ is the incident photon flux density (No. of photons/ m^2s)—No. of photons per unit area per unit time).

The conservation of energy holds that the sum of the total absorbance (A), transmittance (T), and reflectance (R) must be unity, which means that the total absorbance in the case of a multilayer system can be written as

$$A = 1 - T - R. \quad (2)$$

The total absorbance for the complete multilayer stack is simply the sum of the absorbance of the individual layers. However, only those photons that are absorbed inside the device will have the possibility to contribute to the photocurrent. In effect, only a fraction $AI_0/h\nu = AN$ of the incident photons can contribute to the photocurrent. For inorganic semiconductor devices the transmittance is presumably zero ($T=0$) and an internal quantum efficiency (IQE) is calculated as

$$\eta_{\text{IQE}} = \frac{\eta_{\text{EQE}}}{1 - R} = \frac{\eta_{\text{EQE}}}{A}. \quad (3)$$

If T is not zero, as often is the case for organic thin film photovoltaic devices due to thin metal electrodes, the denominator of Eq. (3) should be replaced by Eq. (2). Nevertheless, of those photons that are absorbed in the device, only a fraction will be absorbed in photoactive regions or layers and contribute to the generation of photocurrent. The useful absorbance in a device is therefore typically restricted to one or a few active layers, i , as given by

$$A_{\text{active}} = \sum_i A_i. \quad (4)$$

Having this in mind we can define a quantum efficiency of exciton-to-charge generation (QEC) of an organic photodiode or photovoltaic device as

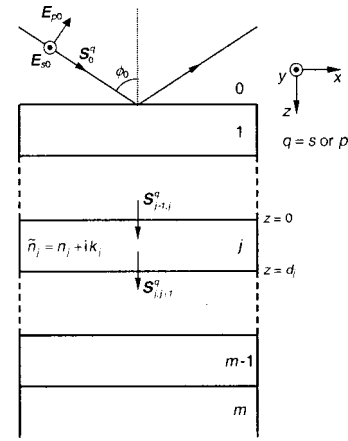


FIG. 1. A schematic multilayer structure of m layers. Each layer has a thickness d_j and its linear optical response is described by a complex index of refraction, $\tilde{n}_j = n_j + ik_j$.

$$\eta_{\text{QEC}} = \frac{\eta_{\text{EQE}}}{A_{\text{active}}} \quad (5)$$

describing the true efficiency of the material in the photoactive layers of the device. In this way, the reflected and transmitted energy of the incident light, and light that is absorbed in electrodes and other layers which do not generate photocurrent, will not be accounted for. Only those photons which are absorbed within the active layers and generate a charge carrier contributing to the photocurrent contribute to the η_{QEC} . This layer collection efficiency is a number, which is unity or less than unity. In the case of η_{QEC} equal to unity we have an internal quantum efficiency of 100% and all the absorbed photons in the active region of the device contribute to the photocurrent generation.

B. Absorbance in a layer

By reference to Fig. 1, consider a plane wave of polarization q ($q = s$ or p) incident at an angle of incidence ϕ_0 from the transparent ambient at a multilayer stack having m layers (or phases). The polarization is described by two mutually perpendicular polarizations; s polarization and p polarization that correspond to light incident with its electric field vector perpendicular to or parallel to the xz plane (the plane of incidence). Each phase j ($j = 1, 2, \dots, m$) in the structure has a thickness d_j and is homogeneous and isotropic with its linear optical response described by a complex index of refraction $\tilde{n}_j = n_j + ik_j$, where n_j and k_j are the refractive index and extinction coefficient. The complex angle of refraction $\tilde{\phi}_j$ in layer j is related to the angle of incidence through $n_0 \sin \phi_0 = \tilde{n}_j \sin \tilde{\phi}_j$.

The absorbance in layer j can be described as the difference in energy flux of the optical field at the internal surfaces of the layer compared to the incident energy flux (intensity) S_0^q as given by

$$A_{qj} = \frac{S_{j-1,j}^q - S_{j,j+1}^q}{S_0^q}, \quad (6)$$

where $S_{j-1,j}^q$ and $S_{j,j+1}^q$ are the normal components of the energy flux at the first internal boundary of layer j ($z_j=0$) and the second internal boundary of layer j ($z_j=d_j$), respectively.

In layer j the absorptance between z_1 and z_2 ($z_1 \leq z_2$, $0 \leq z_1, z_2 \leq d_j$) for a plane wave of polarization q at an incidence angle ϕ_0 is given as

$$A_{qj} = T_{qj} \left[(e^{-A_j z_1} - e^{-A_j z_2}) (1 + |r_{qj}''|^2 e^{-A_j (2d_j - z_1 - z_2)}) + 4\Gamma_q |r_{qj}''| e^{-A_j d_j} \sin(B_j(z_2 - z_1)) \cos(B_j(2d_j - z_1 - z_2) + \delta_{qj}'') \right]. \quad (7)$$

The total absorptance of layer j is given by writing $z_1=0$ and $z_2=d_j$. In Eq. (7) an absorption coefficient dependent on the angle of incidence and the refractive index and extinction coefficient of the layer, equivalent to the customary absorption coefficient [Eq. (17)] is written as

$$A_j = \frac{4\pi K_j}{\lambda} \quad (8)$$

(λ is the wavelength of the incident light) and the corresponding extinction coefficient as

$$K_j = \text{Im}\{\tilde{n}_j \cos \tilde{\phi}_j\} = \left[\frac{1}{2} (-(n_j^2 - k_j^2 - n_0^2 \sin^2 \phi_0) + \sqrt{(n_j^2 - k_j^2 - n_0^2 \sin^2 \phi_0)^2 + 4n_j^2 k_j^2}) \right]^{1/2}. \quad (9)$$

With equivalence to Eq. (18) we also obtain

$$B_j = \frac{2\pi N_j}{\lambda}, \quad (10)$$

where

$$N_j = \text{Re}\{\tilde{n}_j \cos \tilde{\phi}_j\} = \left[\frac{1}{2} ((n_j^2 - k_j^2 - n_0^2 \sin^2 \phi_0) + \sqrt{(n_j^2 - k_j^2 - n_0^2 \sin^2 \phi_0)^2 + 4n_j^2 k_j^2}) \right]^{1/2}. \quad (11)$$

For s-polarized light we obtain

$$\Gamma_s = \frac{\text{Im}\{\tilde{n}_j \cos \tilde{\phi}_j\}}{\text{Re}\{\tilde{n}_j \cos \tilde{\phi}_j\}} = \frac{K_j}{N_j} \quad (12)$$

and

$$T_{sj} = \frac{\text{Re}\{\tilde{n}_j \cos \tilde{\phi}_j\}}{n_0 \cos \phi_0} |t_{sj}^+|^2. \quad (13)$$

For p-polarized light corresponding results are

$$\Gamma_p = \frac{\text{Im}\{\tilde{n}_j / \cos \tilde{\phi}_j\}}{\text{Re}\{\tilde{n}_j / \cos \tilde{\phi}_j\}} \quad (14)$$

and

$$T_{pj} = \text{Re} \left\{ \frac{\tilde{n}}{\cos \tilde{\phi}_j} \right\} \frac{|\cos \tilde{\phi}_j|^2}{n_0 \cos \phi_0} |t_{pj}^+|^2. \quad (15)$$

In Eqs. (13) and (14) $|t_{qj}^+|^2$ is the internal intensity transmittance coefficient from the incident medium to layer j of the actual device and $r_{qj}'' = |r_{qj}''| e^{i\delta_{qj}''}$ is the complex reflection

coefficient for the second subsystem of the multilayer stack from the boundary between interface $j(j+1)$ to phase $m+1$.⁴

At normal incidence the total absorptance in layer j according to Eqs. (7)–(15) reduces to

$$A_j = T_j \left[(1 - e^{-\alpha_j d_j}) (1 + |r_j''|^2 e^{-\alpha_j d_j}) + 2 \frac{\alpha_j}{\beta_j} |r_j''| e^{-\alpha_j d_j} \sin(\beta_j d_j) \cos(\beta_j d_j + \delta_j'') \right]. \quad (16)$$

In this expression

$$\alpha_j = \frac{4\pi k_j}{\lambda} \quad (17)$$

is the absorption coefficient,

$$\beta_j = \frac{2\pi n_j}{\lambda} \quad (18)$$

is the phase coefficient, and

$$T_j = \frac{n_j}{n_0} |t_j^+|^2 \quad (19)$$

is the internal intensity transmittance.

For devices to be used as solar cells the total integrated absorptance of layer j can be calculated by using the energy flux of the solar spectral distribution $F(\lambda)$ as

$$A_{j,\text{solar}} = \frac{\int A_j F(\lambda) d\lambda}{\int F(\lambda) d\lambda}. \quad (20)$$

From the point of view of device design Eqs. (5), (7), (16), and (20) provide the opportunity to optimize layer thickness (device geometry) and thus control where the absorption occurs in the cells in order to enhance the efficiency.

The monochromatic power conversion efficiency is given as

$$\eta_{\text{Power}} = \frac{J_{\text{sc}} V_{\text{oc}} FF}{I_0},$$

where V_{oc} is the open circuit voltage and FF is the fill factor of the device.

III. EXPERIMENTAL DETAILS

Photodiodes were fabricated as thin film multilayer structures. The devices consisted of a poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT-PSS) (Baytron-Bayer AG) layer spin coated on an indium tin oxide (ITO) glass substrate as the hole collecting electrode and an evaporated Al layer as the electron collecting elec-

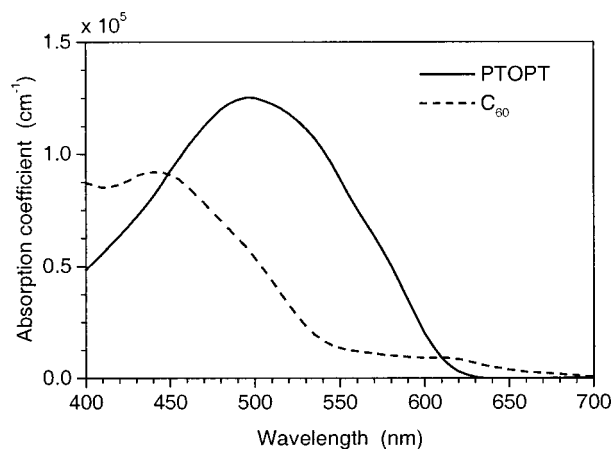


FIG. 2. Absorption coefficients of poly(3-(4-octylphenyl)-2,2'-bithiophene) (PTOPT) and fullerene (C_{60}).

trode. In the single active layer devices the polymer poly(3-(4-octylphenyl)-2,2'-bithiophene) (PTOPT) was used and in the bilayer PTOPT was used together with fullerene (C_{60}). For both types of devices the polymer layer was spin coated from a chloroform solution, and in the case of the bilayer device the C_{60} layer was sublimed on top of the polymer.

Photocurrents were measured with a Keithley 485 picoammeter during illumination of the device through the glass/ITO side with monochromatic light from an MS 257 Oriel monochromator and a tungsten-halogen lamp. The intensity of the incident light was measured using a calibrated silicon photodiode at the same position as the samples prior to measurements of the photocurrent. For the devices used in this study, we have linearity in the relation between photocurrent-illumination intensity over the range of illumination from 1–100 $\mu\text{W}/\text{cm}^2$, and photocurrents may then be found in the range of (typically) 0.1–10 $\mu\text{A}/\text{cm}^2$. The linearity of photocurrent and light intensity is a necessary condition for obtaining the quantum efficiency. At high polychromatic illumination levels series resistance may be a problem; this is not the case at the low intensities used when measuring the EQE.

The complex index of refraction, $\tilde{n} = n + ik$, of each material and the thickness of each layer in the devices are needed in the calculations and were determined by analysis of spectroscopic ellipsometry.^{10,11} Ellipsometric data were measured at multiple angles of incidence on separate layers of the materials with a rotating analyzer NIR-VIS-UV variable-angle spectroscopic ellipsometer (J. A. Woollam Co., Inc.).

IV. RESULTS AND DISCUSSION

Absorption coefficient spectra (400–700 nm) of PTOPT and C_{60} as determined by spectroscopic ellipsometry are shown in Fig. 2. The maximum value of the absorption coefficient for PTOPT and C_{60} were $1.25 \times 10^5 \text{ cm}^{-1}$ (at 500 nm) and $0.92 \times 10^5 \text{ cm}^{-1}$ (440 nm), respectively. Corresponding values for the onset of absorption were 640 and 725 nm. The C_{60} has a much stronger optical transition between 300 and 400 nm,⁴ not shown here.

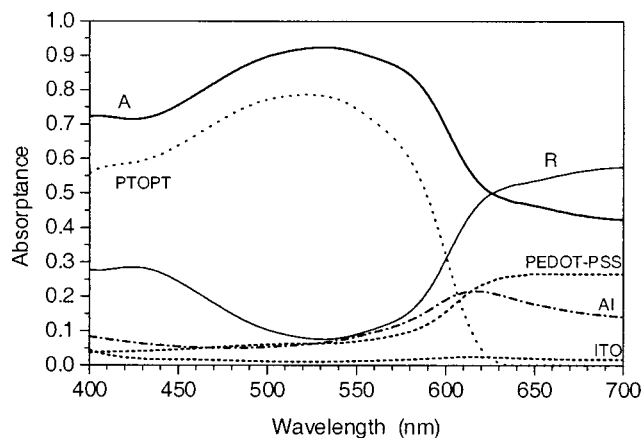


FIG. 3. Total absorbance (A) and reflectance (R) together with the absorbance of the individual layers in a glass (1 mm)/ITO (120 nm)/PEDOT-PSS (80 nm)/PTOPT (60 nm)/Al device.

The approach in Sec. II has been applied to a single layer device and a bilayer (heterojunction) device. However, special care must be given to the transparent glass substrate in the absorbance calculations for our device structures. The transmission of light through the glass must be treated as being incoherent with respect to other beams, due to the large thickness (1 mm), nonuniformity in glass thickness, and finite bandwidth of the light source. This was accomplished by calculating the resultant transmission through the glass substrate by summation of transmitted energies (intensities) instead of complex amplitudes.

In Fig. 3 the reflectance and absorbance of a glass (1 mm)/ITO (120 nm)/PEDOT-PSS (80 nm)/PTOPT (60 nm)/Al device are shown, together with the absorbance of the individual layers. The transmittance (T) of the device was zero since the thickness of the Al was sufficient to obtain zero transmittance through the device. In the wavelength range 400–600 nm there is a high total absorbance which is mainly due to the absorption in the PTOPT layer. The absorbance peaks at 525 nm where the total absorption of the device reaches 0.8, i.e., 80% of the incident photons are absorbed in the device. Of those photons 90% are absorbed in the PTOPT layer where the absorbance is 0.72. Above 600 nm most light is reflected since the onset of absorption of PTOPT is at 640 nm. Observe that a significant amount of photons is absorbed in ITO, PEDOT-PSS and Al, an absorption which is not contributing to the photocurrent. The absorbance of the polymer layer determines the upper limit to the efficiency of this device.

The monochromatic quantum efficiency of this device is very low, $<0.20\%$. Considering the fact that some of the photons are reflected as in the calculated IQE value (η_{IQE}) the internal quantum efficiency is 1.29 times (at 400 nm) and 1.12 (at 500 nm) larger than the EQE (see Fig. 4). If only those photons absorbed in the PTOPT layer are taken into account in the QEC value (η_{QEC}) the corresponding values are 1.79 at the short wavelength side and 1.3 at 500 nm. Hence this device is inefficient since excitons are generated while only a small fraction of these contribute to the generation of photocurrent. It therefore can be concluded that it is not the optical part of the device that limits the efficiency in

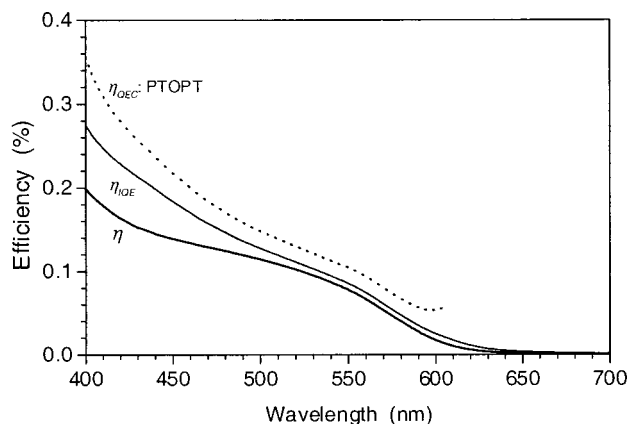


FIG. 4. Monochromatic quantum efficiency (η_{QE}), internal quantum efficiency (η_{IQE}), and quantum efficiency of exciton-to-charge generation (η_{QEC}) of a glass (1 nm)/ITO (120 nm)/PEDOT-PSS (80 nm)/PTOPT (60 nm)/Al device.

this system, but rather exciton dissociation, internal traps, and charge transport that are the limiting factors. In a system with a much lower absorbance in the polymer layer and with the same efficiency the value of the QEC would become higher. It should also be noted that the calculated absorbance in the PTOPT layer is low above 620 nm and hence the QEC is only calculated to 600 nm.

The heterojunction bilayer photovoltaic devices typically have a better performance than the single layer devices. In Fig. 5 the total absorbance, total reflectance, and absorbance of the individual layers of a glass (1 nm)/ITO (120 nm)/PEDOT-PSS (90 nm)/PTOPT (60 nm)/C₆₀ (35 nm)/Al device are shown. Observe that the thickness of the PTOPT layer is the same as for the single layer device. The Al thickness was also in this case large enough to obtain zero transmittance of the device. In this device a significant amount of photons is absorbed in the electrode materials; ITO, PEDOT-PSS, and Al. The PTOPT has a peak in the absorbance at 500 nm where it reaches 0.64 and corresponding values for the C₆₀ layer are at the short wavelength side 0.40 (400 nm). Considering the absorbance for both the PTOPT and the C₆₀ (the heterojunction) we obtain at 464 nm a value of 0.81, which determines the upper limit of the efficiency consider-

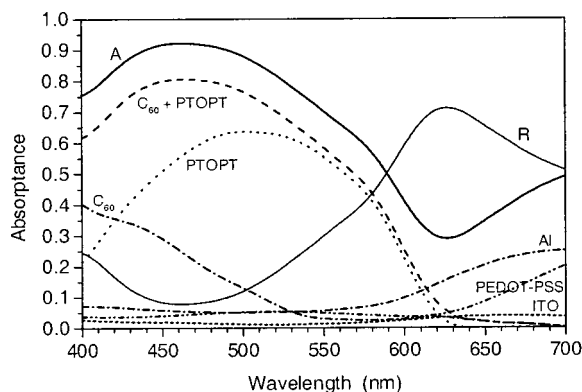


FIG. 5. Total absorbance (A) and reflectance (R) together with the absorbance of the individual layers and PEOPT in a glass (1 nm)/ITO (120 nm)/PEDOT-PSS (90 nm)/PEOPT (60 nm)/C₆₀ (31 nm)/Al device.

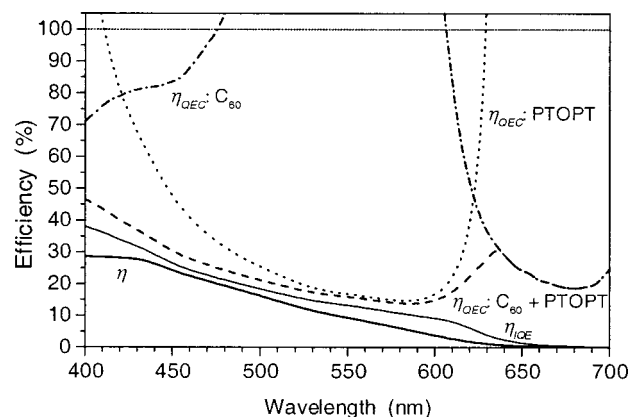


FIG. 6. Monochromatic quantum efficiency (η_{QE}), internal quantum efficiency (η_{IQE}), and quantum efficiency of exciton-to-charge generation (η_{QEC}) of a glass (1 nm)/ITO (120 nm)/PEDOT-PSS (90 nm)/PEOPT (60 nm)/C₆₀ (31 nm)/Al device.

ing the absorption of light. The absorbance values as shown in Fig. 5 were used to calculate the IQE and QEC of the device as shown in Fig. 6. The EQE reaches 28.7% at the short wavelength side and an efficiency of 16.2% at 500 nm in comparison to the IQE values of 38.0% and 18.4%, respectively. Thus the IQE is 1.32 (400 nm) and 1.13 (500 nm) larger than the EQE at this wavelength. It has previously been shown that in the case of a polythiophene/fullerene device, contributions to the photocurrent must come from absorption in both the polymer and the C₆₀ layer.⁴ Using this assumption the QEC (PTOPT+C₆₀) reaches a value of 21.2% at 500 nm, which is 1.3 times larger than the EQE. When considering QEC values of absorption only in the PTOPT or in the C₆₀ the values becomes very large and at some wavelengths larger than 100% (especially the C₆₀). This would imply that more charge carriers are generated than the number of photons absorbed, which is not a realistic presumption; we regard 100% as an upper limit for the validity of the model. Hence also from this type of consideration it must be concluded that the photocurrent generation is due to absorption in both the polymer and the fullerene.

Moreover, the QEC was employed to determine the width of the charge generation regions in the PTOPT/C₆₀ device. A QEC that reaches 100% can be calculated by assuming that only a fraction of the total absorption in the PTOPT and C₆₀ layers contributes to the generation of photocurrent. The width of the active regions in the PTOPT layer and the C₆₀ layer can then be determined by adjusting them to obtain a QEC of 100% for the total action spectrum. However, this method to determine the widths of the active regions is in principle the same as the box model proposed by Rostalski and Meissner,¹² which is based on the assumption of a space charge region of charge generation, rather than of exciton diffusion as described by Ghosh and Feng.¹³ Assuming that the active absorption occurs at regions at the heterojunction in the PTOPT layer and the C₆₀ layer as well as at the C₆₀/Al interface, widths of the active regions were determined to be 7 nm in the PTOPT layer and 11 nm in the C₆₀ layer. If the active region is positioned only at the PTOPT/C₆₀ interface, the result became 7 and 15 nm in the

PTOPT layer and the C₆₀ layer, respectively. Thus only a small part of the total thickness of the layers is contributing to the photocurrent. In addition, much light is absorbed in regions that do not give any contribution. These regions also increase the internal resistance of the device and therefore negative effects on the device efficiency are expected.

Much better performance can be obtained in organic and polymeric photodiodes than the data used in this analysis; recent results claim EQEs up to 50%¹⁴ and others report solar energy efficiencies of single organic crystals reaching 4.5%.¹⁵ There is therefore encouraging examples of high performance organic photodiodes. The use of the layer efficiency concept can help in the analysis and optimization of thin film photovoltaic multilayered devices, in combination with models of photocurrent generation.

V. CONCLUSIONS

The internal quantum efficiency of photovoltaic devices based on organic thin films in terms of a quantum efficiency of exciton-to-charge generation, QEC, is presented. The QEC is based on the monochromatic experimental value of the efficiency, which means that all sources contributing or not contributing to the photocurrent are included in the QEC value. The layer efficiency QEC gives the possibility to optimize layer thicknesses and to extract the true photoelectric capability of the materials.

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