Organic bulk heterojunction photodetectors based on polymer blends with unbalanced mobilities

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Abstract

We investigate bulk heterojunction organic photodetectors with unbalanced charge carrier mobilities in the active layer. We present an experimental example of an organic photodiode with very unbalanced mobilities but achieving state-of-the-art parameters in terms of external quantum efficiency exceeding 75\% (at illumination 638 nm), and of bandwidth exceeding 1 MHz. The active layer of the photodiode is a 50 nm thick bulk heterojunction made of a blend of PCBM and copolymer of 5,6-difluoro2,1,3-benzothiadiazole, with mobility of slower charge carriers of order of $5 \times 10^{-7} \text{cm}^2 (\text{Vs})^{-1}$, estimated using space-charge-limited photocurrent technique. To understand why such a low mobility does not have deteriorating effects on the photodetecting performance of the photodiode, we performed drift-diffusion simulations of bulk heterojunction photodiodes with Langevin recombination. According to the simulation, the bandwidth of the photodiode is approximately independent of the mobility of slower charge carriers in the blend, and the negative effect of low mobility on the responsivity can be compensated by increasing the reverse bias. Our study shows that well-performing organic photodetectors can be fabricated using organic semiconductors having too low mobility for photovoltaic applications.

Keywords: organic semiconductor, charge carrier mobility, drift-diffusion simulation

(Some figures may appear in colour only in the online journal)

1. Introduction

Organic electronics is a promising way to realize flexible, large-area light sensors \cite{1, 2}. Organic photodiodes with responsivity and dark-current competing with state-of-the-art inorganic devices had already been demonstrated \cite{3, 4}. Recently, organic photodetectors with advanced functionality, such as photomultiplication \cite{5, 6}, have been developed. Further research is necessary to improve the bandwidth, lifetime and manufacturing cost \cite{2, 7, 8}.

A particularly simple realization of the organic photodetector utilizes the organic bulk-heterojunction architecture, in which a blend of donor and acceptor organic semiconductors is used as the active layer. Such a single layer structure is desirable for manufacturing from solution, preferably by printing \cite{9, 10}. Organic photodetectors fabricated in this way have the same device structure as the organic bulk heterojunction solar cells.

Despite of the same structure, the figure of merits of bulk heterojunction solar cells and photodiodes are different \cite{11}. The performance of solar cells is expressed by the energy conversion efficiency, normally evaluated for solar...
irradiance spectra AM1.5 at fixed illumination of one Sun (100 mW cm\(^{-2}\)). On the other hand, photodetectors must satisfy requirements which depend on the application. The main figures of merit of photodetectors are the dark current, the noise density, the specific detectivity, the responsivity, and the bandwidth [12, 13]. In contrast to the solar cells, the photodetectors are not optimized for solar irradiance, and normally work under lower illumination.

The influence of charge carrier mobilities and contact properties on the energy conversion efficiency of bulk heterojunction solar cells is a well studied topic [14–17]. Significantly less attention has been given to the study of the influence of the same factors on the performance of photodetectors. Popescu et al [18] studied the influence of incoming light intensity on the bandwidth of the photodetectors. In reference [19], bandwidth and transient response was studied only in the bilayer case. Fallahpour et al [20] studied the influence of material properties on the dark-current of organic photodiodes. Christ et al [21] considered the influence of faster mobility on nanosecond response of photodiodes illuminated by laser pulse.

In this contribution, we investigate the achievable performance of bulk heterojunction photodetectors with unbalanced mobilities. The question is of practical importance, because such blends have low photovoltaic performance [15, 17, 22], but could have superior photodetector performance because of improved absorption, easier manufacturing, reduced density of gap and interfacial states, or improved stability [12, 23].

The presence of unbalanced mobilities in the active layer of a photodiode can be proven by observing the space-charge-limited (SCL) photocurrent [24]. However, unlike in the case of photovoltaic devices [15], the photodetector performance of photodiodes exhibiting SCL photocurrent is less studied. Therefore, we start by characterizing a model photodiode with unbalanced mobilities in section 2. We follow it by presenting simulations of photodiodes with unbalanced mobilities in section 3. The simulation allows us to systematically investigate the effect of charge carrier mobility imbalance on the most important steady-state and transient responses of photodetectors.

2. Example photodiode

We fabricate a a photodiode with unbalanced charge carrier mobilities using a blend of PCBM and a copolymer of 5,6-difluoro2,1,3-benzothiadiazole as the active layer [25]. This material was selected because of high stability and almost trap-free transport, which simplifies comparing with the simulation. The manufactured bulk heterojunction photodiodes have the standard device structure: ITO glass substrate (20 \(\Omega\) sq\(^{-1}\)), 30 nm thick layer of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS), 50 nm thick layer of the organic blend and vacuum evaporated calcium (20 nm)/aluminium top electrode (100 nm). The organic blend was prepared by dissolving the hole transporting polymer together with electron transporting fullerene derivative [60]PCBM (1:3 weight ratio) in mixture of solvents: 1,2-dichlorobenzene (DCB) (350 \(\mu\)L) and chloroform (CHCl3) (800 \(\mu\)L). The active layer was deposited by spin-coat 80 \(^\circ\)C for 10 min before top electrode evaporation. The device was encapsulated using epoxy and glass slide. Except for the PEDOT:PSS layer, the layers were fabricated in an inert, glovebox atmosphere. The pixel area was 4.5 mm\(^2\).

Figure 1. Dependence of the SCL photocurrent on the effective voltage \(V_0 - V\), measured for light intensities varied from 40 W m\(^{-2}\) to 700 W m\(^{-2}\). The regions satisfying equation (1) are indicated with lines. \(V_{\text{sat}}\) denotes the transition voltage to saturation region [24].
Figure 2. Analysis of SCL photocurrent from figure 1. In each case, $S$ denotes logarithmic slope, to be compared with powers in equation (1). (a) Dependence of the photocurrent density on the light intensity, $J_{ph} \propto G^S$, in the SCL photocurrent regime ($V - V_0 = 0.5\, \text{V}$), and in the saturation regime ($V - V_0 = 10\, \text{V}$). (b) Dependence of the saturation voltage on the light intensity, $V_{sat} \propto G^S$. The slopes $S$ are determined using linear fit to experimental data.

Below we briefly review the key properties of space-charge-limited (SCL) photocurrent [15, 26]. The presence of SCL photocurrent simultaneously proves unbalanced mobilities in the blend and allows to estimate the mobility of slower charge carriers [24]. When applicable, the analysis of SCL photocurrent allows to avoid the well known difficulties in the determination of mobility in thin films. In particular, SCL photocurrent does not depend critically on the quality of the injecting contact, which is a major problem in conventional mobility measurements on unipolar samples [27, 28]. The difficulties are even more significant in thin samples, where the thickness of the electrode contact area cannot be considered negligible compared to the thickness of the sample [29].

In the majority of known PCBM blends, the electron mobility is known to be in range $10^{-4}$–$10^{-3}\, \text{cm}^2\, (\text{Vs})^{-1}$ [30–32].
Hole mobility is typically much lower in devices with SCL photocurrent [15, 33]. Thus, in the formulas below, we will follow standard convention by denoting the mobility of slower charge carriers as $\mu_h$.

The SCL photocurrent obeys the formula [24]:

$$J_{ph} = q \left( \frac{9 \epsilon \mu_h}{8q} \right)^{1/4} G^{3/4} (V_0 - V)^{1/2}$$  \hspace{1cm} (1)

where $q$ is the elementary charge and $\epsilon = \epsilon_0 \epsilon_r$ is the dielectric permittivity. $V_0 - V$ denotes the effective voltage, with $V_0$ denoting the open circuit voltage and $V$ denoting the externally applied voltage. The effective e-h generation rate $G$ is proportional to the incoming light intensity, and is assumed to be uniform in volume of the active layer.

In the SCL regime, the current is limited by the accumulation of slower charge carriers in the region with thickness $L_1$. The photocurrent is given by SCL current formula [24]:

$$J_{ph} = \frac{9}{8} \epsilon \mu_h \frac{(V_0 - V)^2}{L_1^3}.$$  \hspace{1cm} (2)

The saturation voltage $V_{sat}$ is defined as the voltage of transition from the SCL photocurrent regime described by equation (1) to the saturation regime. At saturation voltage, the thickness of charge carrier accumulation region is approximately equal to the device thickness $L$ [24]. Thus, the mobility of slower charge carriers can be estimated from formula (2) by setting $L_1 = L$ and $V_0 - V = V_{sat}$.

The photocurrent responses of our diodes, measured under varying light illumination, are plotted in figure 1. The regions of square-root scaling, in which equation (1) is satisfied and SCL photocurrent is observed, are indicated by lines in figure 1. Further analysis necessary to confirm the observation of SCL photocurrent is presented in figure 2. In the SCL region, for $V_0 - V = 0.5$ V, estimated logarithmic slope of photocurrent density as a function of light intensity is 0.74 (figure 2(a)), which is in agreement with term $G^{3/4}$ in equation (1) within experimental uncertainty. As expected, in the saturation region, for $V_0 - V = 10.0$ V, a distinct value of slope 0.93 is obtained. The slope of logarithmic plot of saturation voltage as a function of light intensity is 0.47. This is close, within experimental uncertainty, to value 1/2 resulting from combining equations (1) and (2) with $L_1 = L$ and $V_0 - V = V_{sat}$. Assuming typical value of electric permittivity $\epsilon_r = 3$, this allows to estimate the slower mobility using equation (2) as $\mu_h \approx \frac{5}{10^{-7} \text{cm}^2 \text{V}^{-1}}$.

The results of standard photovoltaic characterization of our device are shown in figure 3. The current-voltage characteristics were measured using AM1.5 illumination with LOT Oriel solar light simulator. The device exhibits low energy conversion efficiency 2.7% and low fill factor 37.8%. These results are expected for a device with highly unbalanced mobilities [14, 22].

The results of photodetector characterization of the same device are shown in figures 4 and 5. In figure 4(a), current-voltage-light characteristics are plotted. They were measured under calibrated monochromatic light source emitting red light with wavelength $\lambda = 634$ nm. Comparing with photovoltaic characterisation in figure 3, the influence of reverse voltage on photocurrent is slightly stronger in figure 4 because of lower illumination intensity, limited to roughly one fifth of AM1.5. In figure 4(b), the responsivity as a function of bias voltage is plotted. The responsivity at each voltage is calculated as a linear fit of the photocurrent to the light intensity. An example line fit for the case of reverse voltage of $-4.5$ V is shown in
Figure 4. Measured steady-state responses of a photodiode sample illuminated by a calibrated red light source (\(\lambda = 634\) nm). a) Current-voltage-light characteristics measured for varying illumination intensity. b) Responsivity (quantum efficiency) as a function of applied voltage for reverse voltages. Inset: linearity at \(V = -4.5\) V.

In figure 5, transient response of the photodiode is plotted. The cut-off of the frequency response of the photodiode, defined as a frequency at which the gain drops by 3 dB, is approximately 1 MHz. Inset of figure 5 shows a response to a square wave illumination signal with frequency of 100 kHz. The square wave is reasonably well reconstructed. Slightly higher distortion of the falling edge is a signature of trap states [16]. Overall, the device exhibits quantum efficiency up to 75\% (figure 4(b)), linearity over 6 orders of magnitude of incoming light, and cutoff-frequency around 1 MHz. (figure 5). These are state-of-the-art parameters [34, 35]. The dark current is typical for organic photodiode without blocking layers [4].

3. Device simulation

In order to generalize our measurements and to further investigate the influence of unbalanced mobilities on performance of organic photodiodes, we run drift-diffusion simulations of photodiode devices using oedes open-source simulator [36, 37]. We use a bulk heterojunction model with Langevin recombination. The device model is thoroughly investigated in reference [17], and is used as included in oedes distribution. Exciton dissociation is not included because it is believed to occur at a much faster timescale than considered here [19, 20]. Simulated device thickness is set to the thickness of our measured devices (50 nm). Illumination with red light with...
Figure 5. Measured frequency response of a photodiode sample. dB unit is defined as $10\log_{10}{A/A_0}$. Inset: transient response to a rectangular illumination signal with frequency of 100 kHz.

$\lambda = 634\text{ nm}$, barriers at contacts 0.3 eV, and non-selective contacts are assumed. Values of other parameters, except for the charge carrier mobilities, are taken from reference [17].

For plotting the results, the responsivity normalization $R_0$ constant is introduced. It is defined as the simulated responsivity of a photodiode with the highest considered values of symmetric mobilities $\mu_h = \mu_e = 10^{-4}\text{ cm}^2\text{ (Vs)}^{-1}$, calculated for illumination intensity in photovoltaic range. In the calculation of $R_0$, generation rate typical for photovoltaic conditions is assumed $G = 1.5 \times 10^{28}\text{ m}^{-3}$ following reference [17]. With assumed wavelength of illuminating light, numeric value of responsivity normalization was $R_0 = 0.43\text{ A W}^{-1}$, which corresponds to EQE of 84%.

Frequency responses of simulated photodiodes are shown in figure 6. The results are calculated for zero bias voltage, for mobilities varied in range $10^{-8} - 10^{-4}\text{ cm}^2\text{ (Vs)}^{-1}$, which are typically observed values in thin film organic optoelectronic devices [15, 30, 31]. The responses were calculated using AC small signal analysis, and therefore correspond to the limit of weak illumination. In order to verify the physical validity of the calculations, we also performed large signal calculations with illuminating intensity of 1 mW cm$^{-2}$, and we obtained the same results.

The results of simulations of frequency responses of diodes with balanced mobilities are shown in figure 6(a). With electron and hole mobilities set equal, the responses show typical low-pass behavior, with the cut-off frequency determined by the mobility. As expected, the cut-off frequency decreases with decreasing mobility, from over 1 MHz calculated for a blend with symmetric mobility of $10^{-3}\text{ cm}^2\text{ (Vs)}^{-1}$ to under 1 kHz calculated for blend with symmetric mobility of $10^{-8}\text{ cm}^2\text{ (Vs)}^{-1}$. At the same time, the calculated maximum responsivity, achievable in the low frequency limit, also decreases, from 1 to 0.025$R_0$ for the smallest value of symmetric mobility.

The simulated frequency responses of diodes with unbalanced mobilities are shown in figure 6(b). The electron mobility $\mu_e$ is set to $10^{-4}\text{ cm}^2\text{ (Vs)}^{-1}$, which is typical for popular organic acceptor PCBM. The hole mobility is varied in the range $10^{-8} - 10^{-4}\text{ cm}^2\text{ (Vs)}^{-1}$. In this case, the cut-off frequency does not change significantly with hole mobility and is close to 1 MHz in all cases, which coincides with the experimental cut-off frequency. The responsivity, in the low frequency limit, drops by one order of magnitude, from $R_0$ to 0.1$R_0$.

To investigate the influence of the applied bias voltage on the responsivity, we calculated responsivity in the low frequency limit as a function of applied bias voltage. The results of calculations for asymmetric blend are shown figure 7. The results of calculations for symmetric blend are not shown because they were similar. The simulation shows that the reduction of mobility of slower charge carriers in the blend unavoidably reduces the responsivity at zero bias voltage. However, this can be effectively counteracted by applying the reverse bias voltage. According to the model, with increasing reverse bias voltage, the responsivity eventually saturates at the maximum value of 100% EQE. This is because the model assumes that only one e-h pair can generated per photon, and that the contact is always blocking regardless of applied electric field. This is an approximation, which holds in typical bulk heterojunction devices, but is true only for a limited range of applied voltage.
Comparing the responsivity calculated with $\mu_h = 10^{-6}\text{cm}^2\text{(Vs)}^{-1}$ with our experimental data, the agreement of responsivity as a function of bias voltage is qualitative. The steep initial part and the saturation of responsivity with increasing reverse bias is reproduced by the simulation. The slope and numeric values of responsivity are less accurate. This is expected because the parameters of the simulation were taken from the literature and were not optimized for agreement with our measurements.

With parameters assumed in our simulation, a photodiode with slower mobility of $10^{-7}\text{cm}^2\text{(Vs)}^{-1}$ has approximately 70% of responsivity of the best photodiode at applied bias voltage of $-5\text{ V}$. Moreover, the results obtained suggest that an existence of a significant dependence of the responsivity...
on applied voltage over wide range of the bias voltage could indicate asymmetric transport in the active layer.

4. Conclusions

In conclusion, we present experimental and theoretical results demonstrating that state-of-the-art organic bulk-heterojunction photodiodes can be manufactured using polymer blends with highly unbalanced charge carrier mobilities. We give an example of a photodiode utilizing a low-mobility donor-acceptor co-polymer with mobility of slower charge carriers of order of $5 \times 10^{-7}$ cm$^2$/(Vs)$^{-1}$. The photodiode achieves external quantum efficiency of 75% and bandwidth of 1 MHz.

Further investigation by the means of drift-diffusion simulation indicate that, in the considered range of mobilities, the bandwidth of photodetector is almost independent of the mobility of slower charge carriers in the blend. On the other hand, in a photodiode with very unbalanced mobilities, the responsivity of photodetector at zero bias voltage is largely reduced. However, if sufficiently high reverse bias voltage can be applied, quantum efficiency close to that of devices utilizing blends with high mobility can be achieved.

Our contribution suggests that organic semiconductors for applications in state-of-the-art organic photodiodes are not limited to semiconductors with state-of-the-art photovoltaic performance. On the other hand, a significantly wider group of organic semiconducting materials can be successfully used in organic photodiodes. Such semiconductors can trade charge carrier mobility for other advantages, such as the possibility of solution processing, enhanced stability, or enhanced absorption at a specific wavelength. The results of simulations also suggest that when optimizing the transient response of bulk heterojunction photodetectors, attention should be focused on the semiconductor with higher mobility.

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