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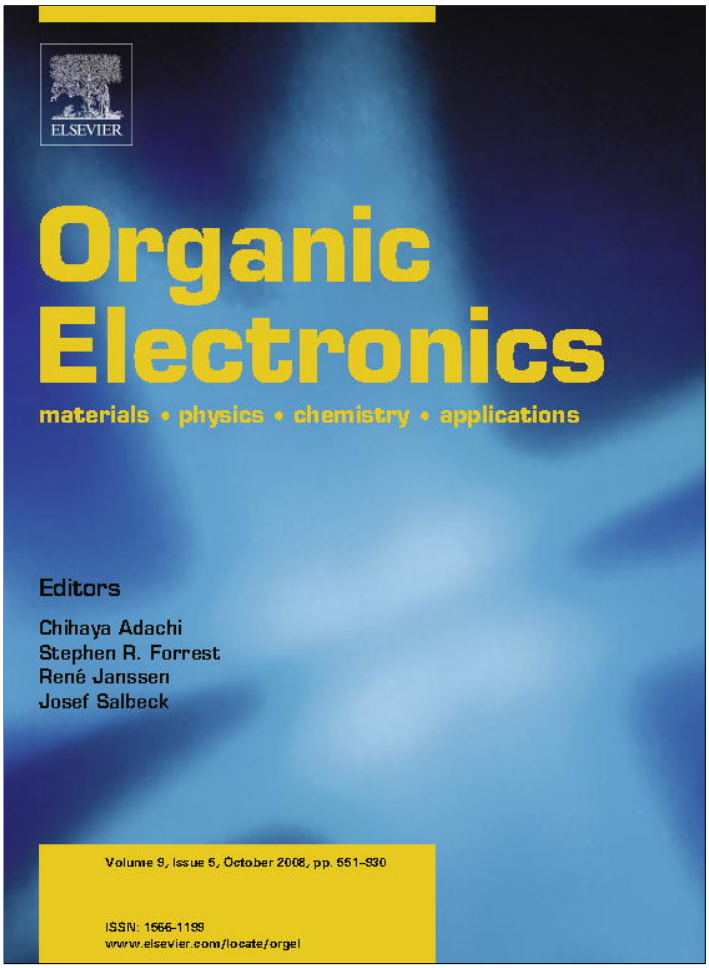

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Effects of polarons and bipolarons on spin polarized transport in an organic device

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ABSTRACT

A polaron–polaron interaction model is suggested to study the spin injection and transport in an organic semiconductor (OSC) device. The evolutions of spin polarons and spinless bipolarons are calculated from the drift-diffusion equations, in which both the polaron–bipolaron transition and the spin-flipping of a spin polaron are included. Then the spin polarized current is obtained. It is found that the polarons are responsible for the spin polarized transport in an OSC. Different from the case in a normal inorganic semiconductor, spinless bipolarons will affect the spin polarization of the OSC device. Finally, effects of the spin-flip time and the mobility of the carriers on the spin polarization in an organic device are discussed.

1. Introduction

Organic semiconductors (OSCs) are a class of electronic materials that have attracted considerable attention in the last decades. They have revolutionized important technological applications including large-area electrics, owing to their processing and performance advantages compared with conventional semiconductors for low-cost or large-area device applications [1–4]. Recently, OSCs find their another application as spin transport materials. Their soft lattice structure and capability of reconstructing the structure also provide an opportunity to form a good interface with ferromagnetic metal (FM) or half-metal contacts, reducing the probability of spin scattering at the interface. In addition, the spin–orbit coupling and hyperfine interaction are very weak in the OSCs, so that the electron spin diffusion length of an OSC is expected much longer than that of a usual inorganic material [5]. A new field called organic spintronics is emerging out and attracting the attention of both physical and chemical scientists [6–16]. In 2002, Dediu's group has observed spin injection into thin films of the conjugated organic material sexithienyl (T_6) on nanostructured planar hybrid junction $La_{0.7}Sr_{0.3}MnO_3$/T_6/$La_{0.7}Sr_{0.3}MnO_3$ at room temperature [6]. The spin diffusion length in T_6 is about 200 nm at room temperature. Xiong et al. have also observed spin injection and transport in a $La_{0.7}Sr_{0.3}MnO_3$/Alq_3/Co organic spin valve. The measured magnetoresistance can be as high as 40% at low temperature [7]. Majumdar et al. have observed as much as 80% magnetoresistance (MR) at 5 K and 1.5% MR at room temperature in the structure of $La_{0.67}Sr_{0.33}MnO_3$/poly 3-hexylthiophene/Co organic spin valve [8]. They also found

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that there is a thin spin-selective tunneling interface between LSMO and the polymer, which improves the spin injection. On the theoretical side, Yu et al. studied the influence of weak magnetic field for spin transport process in magnet/polymer/magnet structures [9]. Xie et al. suggested a model for $Re_{1-x}Sr_xMnO_3$/polymer structure and studied the spin density distribution of the system from the Su–Schrieffer–Heeger-like Hamiltonian [10]. They indicated that the carriers in OSCs contain spin polarons and spinless bipolarons. We used the spin-dependent diffusion model [9,11–13] of the electrochemical potential to study the spin polarized injection and transport in the structure of FM/OSC hetero junction and FM/OSC/FM trilayers [14,15]. By supposing that polarons and bipolarons coexist in OSCs with a given proportion r defined as $r = n_p/(n_p + n_{bp})$, we found that spin polarons are the effective carriers of the spin polarized current. A large spin polarization can be realized even that the spin polarons are minority of carriers in the OSCs.

However, in present investigations polarons and bipolarons were supposed to exist independently. In fact, due to temperature, pressure, impurity or external fields and so on, polarons and bipolarons in OSCs can transit each other. Two polarons with opposite spins can annihilate into one spinless bipolaron, while one spinless bipolaron can be decomposed into two polarons [17]. The stability and transition of polarons and bipolarons have been widely studied both from theoretical and experimental aspects [18,19]. In this paper, we do not research the transition origin of polarons and bipolarons. Instead we focus on the spin polarized transport in an OSC by considering the transition of spin polarons and spinless bipolarons. We will suggest a new mechanism about the polaron and bipolaron transition in a spin non-equivalent OSC and then build the evolution equations for polarons and bipolarons separately. Section 2 will provide a description of the model and the derivation of the relevant formulae, Numerical results are presented and discussed in Section 3, and conclusions are drawn in Section 4.

2. Model and method

We consider an OSC with a FM as contact. The spin polarized current injects from the semi-infinite FM contact ($x < 0$) into the semi-infinite organic layer ($x > 0$) as shown in Fig. 1, which will result in a non-equilibrium distribution of spins. We suppose that the spin-up electrons are the majority carriers and the spin-down electrons the minority ones. When they are injected into the organic layer, both the spin-up and spin-down electrons will evolve into either spin polarons or spinless bipolarons. Our theoretical investigation is based on the drift-diffusion model [20,23]. We assume that: (1) spin-up and spin-down polarons may transfer each other by reversing the spin; (2) a spin-up polaron may combine with a spin-down one to form a spinless bipolaron and vice versa. Therefore, the evolution equations may be written as,

$$\frac{\partial n_{\uparrow(\downarrow)}}{\partial t} = \frac{1}{e}\mathrm{div}\vec{j}_{\uparrow(\downarrow)} \pm \left(\frac{n_\downarrow}{\tau_{\downarrow\uparrow}} - \frac{n_\uparrow}{\tau_{\uparrow\downarrow}}\right) + s_{\uparrow(\downarrow)}(\vec{r},t), \tag{1a}$$

$$\frac{\partial N}{\partial t} = \frac{1}{2e}\mathrm{div}\vec{j}_N + s_N(\vec{r},t), \tag{1b}$$

where e is the electron charge, $n_{\uparrow(\downarrow)}$ the density of spin-up (spin-down) polarons, and N the density of bipolarons. The spin-flip time $\tau_{\uparrow\downarrow}$ indicates the average time of a spin-up polaron flipping its spin and $\tau_{\downarrow\uparrow}$ that of a spin-down polaron. $s_{\uparrow(\downarrow)}(\vec{r},t)$ is the source of spin polarons and $s_N(\vec{r},t)$ that of spinless bipolarons, which result from the transition between polarons and bipolarons within the OSC. $\vec{j}_{\uparrow(\downarrow)}$ is the current density of spin-up (-down) polarons and $\vec{j}_N$ that of bipolarons, which can be described by

$$\vec{j}_{\uparrow(\downarrow)} = \sigma_{\uparrow(\downarrow)}\vec{E} + eD_n\nabla n_{\uparrow(\downarrow)}, \tag{2a}$$

$$\vec{j}_N = \sigma_N\vec{E} + 2eD_N\nabla N, \tag{2b}$$

where $\sigma_{\uparrow(\downarrow)} = en_{\uparrow(\downarrow)}\mu_n$ and $\sigma_N = 2eN\mu_N$ are the conductivities of polarons and bipolarons respectively. $\vec{E}$ is the applied external electric field. D_n, μ_n, D_N and μ_N are the diffusion coefficients and mobilities of polarons and bipolarons which obey Einstein relations $D_n/\mu_n = k_0T/e$ and $D_N/\mu_N = k_0T/2e$ respectively, where k_0 is Boltzmann constant and T the temperature. We neglect the spin dependence of the diffusion coefficients and mobilities of polarons in present investigation.

As stated above, polarons and bipolarons are not independent but transit each other during their transport, which provide the sources of the carriers in the organic layer. Considering the spin non-degeneracy in present system, we propose the rate equation for polarons and bipolarons as,

$$s_{\uparrow(\downarrow)} = -kn_\uparrow n_\downarrow + bN, \tag{3a}$$

$$s_N = kn_\uparrow n_\downarrow - bN. \tag{3b}$$

The first term on the right of Eq. (3a) describes the probability that a spin-up (-down) polaron encounters a spin-down (-up) polaron to annihilate into a spinless bipolaron. The second term describes the reversed process that a bipolaron decomposes into a spin-up and a spin-down polaron. Parameter k and b express the transition strength, which are dependent on the temperature. For a spin-degenerate system, we have $n_\uparrow = n_\downarrow$ and Eq. (3) gives the result of Refs. [21,22].

Substituting Eqs. (2) and (3) into Eq. (1), we finally obtain the evolution equations for spin polarons and spinless bipolarons along the x-direction,

$$\frac{\partial n_{\uparrow(\downarrow)}}{\partial t} = D_n\frac{\partial^2 n_{\uparrow(\downarrow)}}{\partial x^2} + \mu_n E\frac{\partial n_{\uparrow(\downarrow)}}{\partial x} + n_{\uparrow(\downarrow)}\mu_n\frac{\partial E}{\partial x} \pm \left(\frac{n_\downarrow}{\tau_{\downarrow\uparrow}} - \frac{n_\uparrow}{\tau_{\uparrow\downarrow}}\right) - kn_\uparrow n_\downarrow + bN, \tag{4a}$$

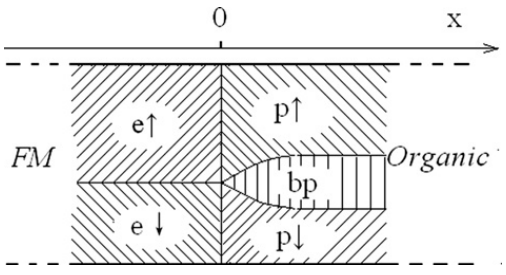

Fig. 1. A schematic diagram of the "FM/OSC" hetero structure.

$$\frac{\partial N}{\partial t} = D_N \frac{\partial^2 N}{\partial x^2} + \mu_N E \frac{\partial N}{\partial x} + N\mu_N \frac{\partial E}{\partial x} + k n_\uparrow n_\downarrow - bN. \quad (4b)$$

3. Results and discussion

First let us consider the polaron and bipolaron transition in an isolated OSC system. The sources mean the creation rates of the carriers and Eq. (3) can be solved analytically with initial condition $n_\uparrow(t=0) = n_1$, $n_\downarrow(t=0) = n_2$ and $N(t=0) = n_3$

$$n_\uparrow = \frac{\left(B + \sqrt{\frac{A}{k}}\right)e^{-Ct+C_0} + \sqrt{\frac{A}{k}} - B}{1 - e^{-Ct+C_0}}, \quad (5a)$$

$$n_\downarrow = \frac{\left(B + \sqrt{\frac{A}{k}}\right)e^{-Ct+C_0} + \sqrt{\frac{A}{k}} - B}{1 - e^{-Ct+C_0}} - n_1 + n_2, \quad (5b)$$

$$N = \frac{1}{2}(n_1 + n_2) + n_3 - \frac{1}{2}(n_\uparrow + n_\downarrow), \quad (5c)$$

where $A = \frac{k}{4}\left(n_2 - n_1 + \frac{b}{k}\right)^2 + b(n_1 + n_3)$, $B = \frac{1}{2}\left(n_2 - n_1 + \frac{b}{k}\right)$, $C = 2\sqrt{Ak}$ and $C_0 = \ln\frac{n_1 + B - \sqrt{\frac{A}{k}}}{n_1 + B + \sqrt{\frac{A}{k}}}$. There is a conservation condition during the transition, i.e., $n_1 - n_\uparrow = n_2 - n_\downarrow = N - n_3$.

It can be found from Eq. (5) that, after a long time evolution, the system will approach a equilibrium distribution of polarons and bipolarons. From experimental measurements [24,25], it has been known that polarons and bipolarons may coexist in most of conducting polymers. In theory [19], it was indicated that a bipolaron is energetically favorable to two isolated polarons in most polymers, which suggests a positive value of parameter k and a small positive value of b. Here we take $k = 5.6 \times 10^{-10}$ cm^3/s and $b = 1.7 \times 10^7$ s^{-1}. The evolution of carriers density with time is shown in Fig. 2. We supposed that the majority carriers are spin-up polarons with a density of $n_1 = 3.0 \times 10^{16}$ cm^{-3}, the minority ones are spin-down polarons with $n_2 = 2.0 \times 10^{16}$ cm^{-3} and there are no bipolarons with $n_3 = 0$ at the beginning. It is found that, with time evolution, the densities of both spin-up polarons and spin-down polarons decrease and, at the same time, the density of bipolaron increases, which means that the transition between polarons and bipolarons takes place. The transition approaches a dynamic equilibrium or saturation of the carriers density after a duration of $T_{p-bp} = 50$ ns in present parameters. The duration time T_{p-bp} is sensitive to the value of k. It is obtained that T_{p-bp} will be as long as seconds to reach a transition equilibrium if parameter k is smaller than 10^{-18} cm^3/s as taken in Refs. [21,22]. Parameter b mainly affects the saturation densities of polarons and bipolarons. A small value of b means that most of the spin minorities (spin-down polarons) will couple with spin majorities (spin-up polarons) to composite spinless bipolarons and at the same time the decomposition of bipolarons is small.

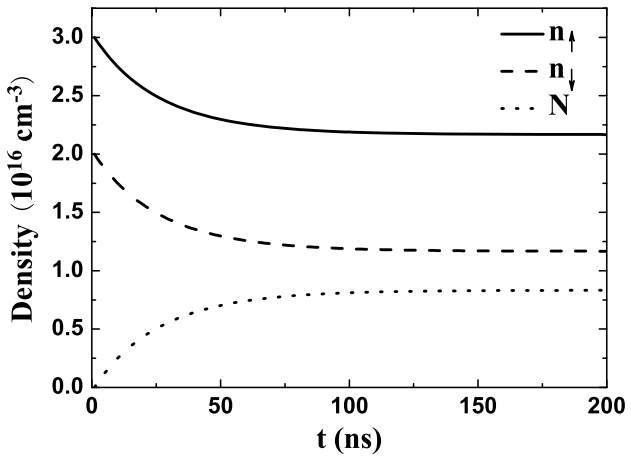

Fig. 2. The evolution of carriers density with time. The parameter is $k = 5.6 \times 10^{-10}$ cm^3/s, $b = 1.7 \times 10^7$ s^{-1}.

Now let us consider the injection and transport of spin carriers from a ferromagnetic contact to an organic semiconductor in an FM/OSC system based on Eq. (4), which could not be solved analytically. We take a numerical calculation by applying a constant driving electric field $E = 5.0 \times 10^4$ v/cm within the organic region and so $\partial E/\partial x = 0$. In addition, it is assumed that the injected electrons are completely spin polarized and are transited into spin-up polarons as soon as they are injected into the organic layer at the interface. So that we set the initial and boundary conditions as $n_\uparrow(x=0,\ t=0) = 5.0 \times 10^{16}$ cm^{-3}, $n_\downarrow(x=0,\ t=0) = 0$ and $N(x=0,\ t=0) = 0$. This assumption may be rational for a half-metal (CMR, Colossal Magnetic Resistance, such as $La_{1-x}Sr_xMnO_3$) contact if we neglect the spin-flip at the interface. All the other parameter values are listed in the captions of the corresponding figures.

To analyze the spin characteristic of the injected charges in the OSC, we define the spin polarization $P = (n_\uparrow - n_\downarrow)/(n_\uparrow + n_\downarrow)$, which is similar to that in a normal inorganic semiconductor, but with a different meaning due to the existence of bipolarons in the OSCs. As the bipolarons have not any contribution to the spin polarization, they are not included in the denominator of the definition. For example, if all the spin minorities (spin-down polarons) compose with the spin majorities (spin-up polarons) into bipolarons, there will no free spin-down carriers left in the channel. So we should have the spin polarization $P = 100\%$ which is consistent to the definition.

The evolutions of spin-up, spin-down polarons and spinless bipolarons in the OSC layer are shown in Fig. 3a. Due to the spin-flip effect and the polaron–polaron interactions, although there is only the injection of spin-up polarons, spin-down polarons and spinless bipolarons will be created during the transport. The densities of these carriers will reach a dynamic balance at a distance of about 250 nm from the interface, which reflects the spin relaxation in an organic semiconductor. It should be mentioned that the spin relaxation in an OSC is different from that in a normal inorganic semiconductor. In an OSC, the spin relaxation contains two factors: one is the spin-flipping of spin polarons, and the other is the transition between spin polarons and spinless bipolarons. Due to the spin-flip effect, the minorities (spin-down polarons) will be created in the transport along with the annihilating of majorities (spin-up polarons), which will affect the spin polarization of the injected current. In addition, due to the polaron–polaron interaction, some spin opposite polarons will annihilate into spinless bipolarons (or its inversed process), which will further affect the spin polarization. The spin polarization is shown in Fig. 3b. Apparently, it decays into the OSC.

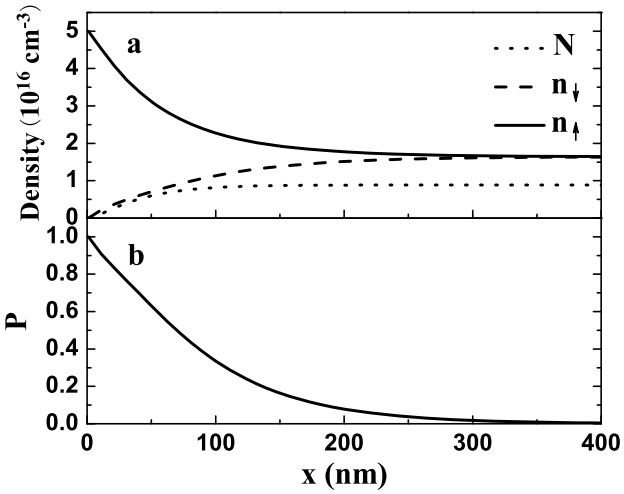

Fig. 3. The distribution of carrier densities (a) and spin polarization P (b) in the OSC layer. The solid, dashed, dot lines in fig. (a) indicate the spin-up polaron, spin-down polaron and bipolaron, respectively. The parameters are $\mu_n = \mu_N = 5.0 \times 10^{-4}$ cm^2/V s, $k = 5.6 \times 10^{-10}$ cm^3/s, $b = 1.7 \times 10^7$ s^{-1}, $\tau_{\uparrow\downarrow} = \tau_{\downarrow\uparrow} = 5.0 \times 10^{-7}$ s.

It is interesting to note that, from the present definition about the spin polarization, the appearance of bipolarons will increase the spin polarization. Effect of bipolaron density on the spin polarization P in OSC layer is shown in Fig. 4. If there are not any bipolarons during the transport, the spin polarization decays exponentially, which is consistent with that in a normal inorganic semiconductor injection if we substitute electrons in an inorganic semiconductor with spin polarons in an OSC. As the creation of bipolarons will change the distribution of spin polarons and therefore affect the spin polarization, it is found that the spin polarization will deviate from the exponential. There is an inflexion at x_0 in spin polarization curve $P(x)$, which means that at this point, the transition between polarons and bipolarons reach a dynamical equilibrium. The density of bipolarons keeps unchanged after x_0, as showed in the inset of Fig. 4. It also means that after x_0, bipolarons has no

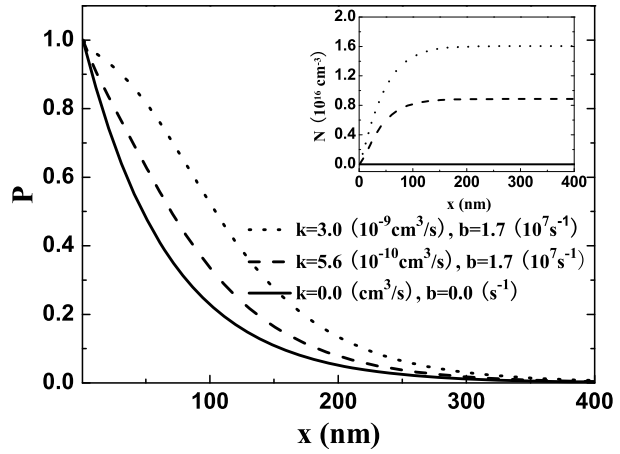

Fig. 4. The distribution of spin polarization P with the different bipolaron density. The different bipolarons density which is shown in the inset is obtained by adjusting the value of the parameter k and b, the solid line for $k = 0.0$ cm^3/s and $b = 0.0$ s^{-1}, the dash line for $k = 5.6 \times 10^{-10}$ cm^3/s and $b = 1.7 \times 10^7$ s^{-1}, and the dot line for $k = 3.0 \times 10^{-9}$ cm^3/s and $b = 1.7$ - 10^7 s^{-1}, respectively. The other parameters are same as those in Fig. 3.

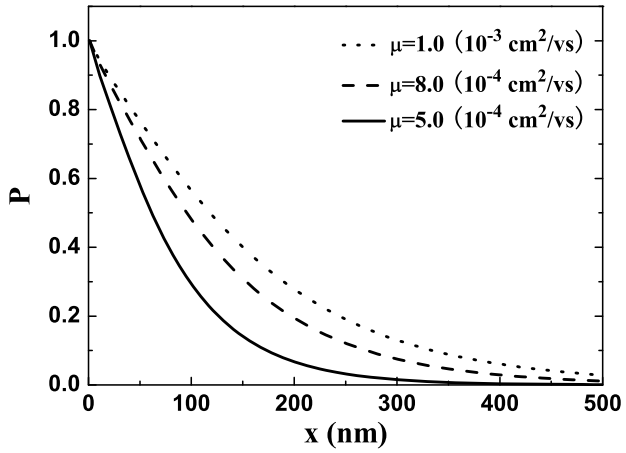

Fig. 5. The distribution of spin polarization P in the OSC layer for the different mobility. The solid, dash and dot lines indicate the different mobilities, i.e., $\mu = 5.0 \times 10^{-4}$ cm^2/V s, $\mu = 8.0 \times 10^{-4}$ cm^2/V s and $\mu = 1.0 \times 10^{-3}$ cm^2/V s, respectively. The other parameters are same as those in Fig. 3.

apparent effect on the spin polarization. Therefore, the enrollment of bipolarons in OSCs makes the spin transport more complex than that in normal semiconductors. In a normal inorganic semiconductor, there are only spin carriers, electrons or holes. At any point of the channel, one may recognise the spin-up and spin-down carriers with a spin detector. Then the spin polarization can be calculated. But in an OSC, one only could recognise the spin-up and spin-down polarons, and the spinless bipolarons could not be measured with a spin detector.

It has been indicated that a polaron moves faster than a bipolaron. We take different mobilities of polarons from bipolarons as $\mu_n = 2\mu_N = \mu$ and investigate the effect of mobility on spin polarization. The results are shown in Fig. 5. It is found that a large mobility result in a long spin diffusion in an OSC, which is consistent to that in a normal inorganic semiconductor. It should be mentioned that, due to bipolarons, the inflexion of spin polarization curve will be apart from the interface, which means that the distance

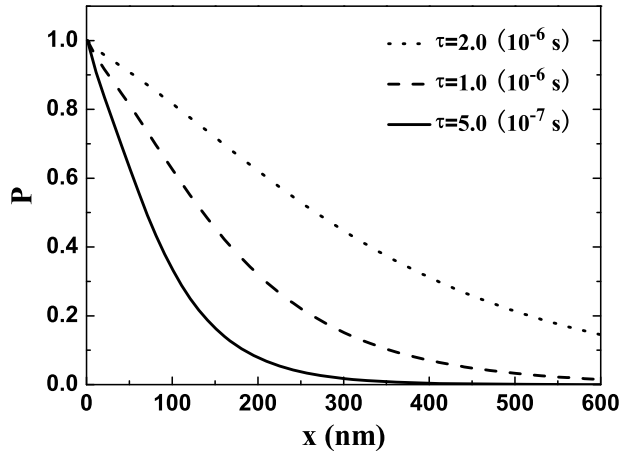

Fig. 6. The distribution of spin polarization P in the OSC layer for the different spin-flip time of the polarons when $\tau_{\uparrow\downarrow} = \tau_{\downarrow\uparrow} = \tau$. The solid, dash and dot lines indicate the different spin-flip times, i.e., $\tau = 5.0 \times 10^{-7}$ s, $\tau = 1.0 \times 10^{-6}$ s and $\tau = 2.0 \times 10^{-6}$ s respectively. The other parameters are same as those in Fig. 3.

of the effect of bipolarons on the spin polarization will increase with its mobility during the transport. The influence of the spin-flip time on the spin polarization is shown in Fig. 6. It is found that, consistent with the case of an inorganic semiconductor, a long spin-flip time will be helpful to the spin polarization in OSC. And the distance of the effect of bipolarons on the spin polarization will increase with its spin-flip time during the transport.

4. Summary

In summary, we investigated spin injection and transport from ferromagnetic contact to an OSC based on the drift-diffusion equations, where all the possible spin factors are included. Especially, a polaron–polaron interaction model is suggested to understand the polaron–bipolaron transition. This model considers the spin dependence in an organic spin device. Different from the case in an inorganic semiconductors, the relaxation of spin in the OSCs contains two factors: one is the spin-flip effect, and the other is the transition between polarons and bipolarons. By supposing that the injected electrons from the ferromagnetic contact are transited into fully spin polarized polarons when they enter an OSC, we obtain the evolutions of polarons and bipolarons during their transport in the OSC. It is found that the appearance of bipolarons decreases the density of the spin carriers (polarons) and so affects the spin polarization of the system. Polarons and bipolarons will reach a dynamical equilibrium in density during their transport. And then the spin polarization is dominated mainly by the spin-flipping of polarons. We also found that a large mobility or a long spin-flip time of spin polarons will result in a large spin diffusion length in the OSC.

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