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Spin filtering through a metal/organic-ferromagnet/metal structure

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Spin filtering in a metal/organic-ferromagnet/metal sandwich device was investigated based on an extended SSH+Heisenberg (SSH=Su-Schrieffer-Heeger) model and the Landauer-Büttiker formula. A large spin-polarized current was predicted, which is dependent on the property of the organic ferromagnetic interlayer as well as the interfacial interactions between the metal electrode and the organic ferromagnet. The effect of thermal fluctuation of the radical spins on the current polarization is discussed. Our investigation suggests a possible application of an organic ferromagnet in a spin filter device without any external field modulation.

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I. INTRODUCTION

Spintronics is a novel branch of nanoscience which exploits spin properties as a potential basis for a generation of electronic devices with improved performance and enhanced functionality.[1–4] A basic element required by most of the proposed concepts is to generate spin-polarized current through a sandwich structure. Generally speaking, two kinds of interlayers in the sandwich device are adopted. The first uses a nonferromagnetic material as the interlayer, such as a metal, semiconductor, superconductor, or insulator.[5–7] The second less studied method uses a magnetic material as the interlayer. It has been found that in a metal/magnetic semiconductor/metal device, such as a Ag/EuSe/Al or Ag/EuS/Al sandwich structure, a large electron-spin polarization of the tunneling current was reached under a strong applied magnetic field on the order of a tesla.[8,9] Several other schemes of spin-polarized transport in such materials have also been investigated extensively.[10,11] However, in most of the above devices, a strong magnetic field is always crucial to realize the large spin polarization (SP), which is used to generate either the spin-selective barriers or spin splitting of the resonant level. The search for novel materials independent of external magnetic field with advanced performance will be meaningful both for theorists and experimentalists.

Organic semiconductors as an alternative material have been widely used in electronics and photonics, and especially in molecular electronics. Recently, it has been reported both theoretically and experimentally that a conducting polymer can serve as a spin-polarized material. For example, Dediu *et al.* reported a magnetic resistance in an organic device of $\mathrm{Re}_{1-x}\mathrm{Ak}_x\mathrm{MnO}_3$/sexithienyl/$\mathrm{Re}_{1-x}\mathrm{Ak}_x\mathrm{MnO}_3$.[12] Xiong *et al.* built an organic spin valve with small π-conjugated molecular 8-hydroxy-quinoline aluminum (Alq_3).[13] The organic semiconductor is sandwiched between a layer of cobalt and half-metallic manganite $\mathrm{La}_{0.7}\mathrm{Sr}_{0.3}\mathrm{MnO}_3$ [colossal magnetoresistance (CMR)]. It was found that the magnetoresistance could be as high as 40% at low temperature. Xie *et al.* suggested a theoretical model for a CMR/polymer junction and studied the interfacial spin polarization.[14] All of these investigations stressed the importance of the softness of an organic structure, which, for example, may adjust its configuration to form a stable interface with other substance, thus resulting in a low interface potential wall.

In the past decade, organic ferromagnets have been studied to search for a combination of ferromagnet with organic materials. An organic ferromagnet is different from a normal one as there are few transition-metal impurities which can bring the usual magnetic properties from the strong correlations between the itinerant or localized d or f electrons and the itinerant s electrons. Several pure organic ferromagnets have been synthesized from conjugated polymers, such as poly-BIPO,[15,16] m-PDPC,[17] and pyrro-PAN.[18] Korshak *et al.*[15] and Cao *et al.*[16] have separately synthesized an organic ferromagnet called poly-BIPO, which has a simple quasi-one-dimensional structure. Its magnetic properties, such as high Curie temperature (150–190 °C),[15] magnetic hysteresis with residual magnetization (0.025–0.5 emu/g), and coercive force (295–470 Oe),[16] show a promising prospect in the application of organic ferromagnet. A simplified structure scheme is plotted in Fig. 1. The main zigzag chain consists of carbon atoms with π itinerant electrons and R is a type of side radical containing an unpaired electron connecting with the odd sites in the main chain. It is assumed that there are antiferromagnetic interactions between the π-electron spins along the main chain and the residual spins of radical R's. As a result, all of the residual spins of the radical R's will try to form a ferromagnetic order, as shown by arrows in Fig. 1. In this paper, we constitute a metal/organic-ferromagnet/metal (M/OF/M) structure based on poly-BIPO as the interlayer in order to understand its transport properties. As there is a strong correlation between the itinerant electron spin and residual spins of the side radical in the organic interlayer, it is expected that the electric current through the device should be spin polarized. In addition, due to the intrinsic strong electron-lattice (e-la) interaction in the organic layer, the charge transport through the M/OF/M will be different from that of a structure with a normal inorganic ferromagnet as the interlayer. In this paper, the spin-dependent Landauer-

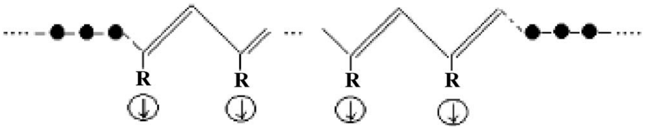

FIG. 1. The simplified structure of a metal/organic-ferromagnet/metal nanojunction.

Büttiker formula[19] is employed to calculate the current-voltage (I-V) curve through an M/OF/M device. An extended SSH+Heisenberg (SSH=Su-Schrieffer-Heeger[20]) model is adopted to describe an organic-ferromagnet poly-BIPO molecule, which contains the e-la coupling and the spin correlation between π electrons and the residual spins of radical R's. The model Hamiltonian is described in the next section. Calculations are carried out, and the results are analyzed in Sec. III. Finally, in Sec. IV, a summary is given.

II. MODEL AND METHOD

We assume an organic-ferromagnet poly-BIPO molecule connected between two one-dimensional semi-infinite metal electrodes. The molecule is modeled with a one-dimensional chain. The Hamiltonian of the system can be written as

$$H = H_{\mathrm{OF}} + H_l + H_r + H_{\mathrm{coup}} + H_E. \tag{1}$$

The first term is the Hamiltonian of the organic-ferromagnet molecule,[21,22]

$$\begin{aligned} H_{\mathrm{OF}} = &-\sum_{n,s}[t_0 - \alpha(u_{n+1}-u_n)](c^+_{n,s}c_{n+1,s} + c^+_{n+1,s}c_{n,s}) \\ &+ \frac{K}{2}\sum_n (u_{n+1}-u_n)^2 + J_f \sum_n \delta_{n,o}\vec{S}_{nR}\cdot\vec{S}_n \\ &+ U\sum_{n,s} c^+_{n,\uparrow}c_{n,\uparrow}c^+_{n,\downarrow}c_{n,\downarrow} + W\sum_{n,s,s'} c^+_{n,s}c_{n,s}c^+_{n+1,s'}c_{n+1,s'}, \end{aligned} \tag{2}$$

where t_0 is the hopping integral of the π electrons along the chain with a uniform lattice, α represents the electron-lattice coupling, u_n is the lattice displacement at site n, K is the elastic constant, and $c^+_{n,s}$ ($c_{n,s}$) denotes the creation (annihilation) operator of an electron at the nth site with spin s. The third term describes the spin correlation between spin $\vec{S}_n$ of the π electron and the residual spin $\vec{S}_{nR}$ of side radical R. It is assumed that radical R's connect with the odd sites in the chain, which is denoted by $\delta_{n,o}$. We select coupling constant J_f positive, which means that the radical and the attached site is an antiferromagnetic coupling. The last two terms describe the electron-electron (e-e) interactions between π electrons, which is considered by the extended Hubbard model. U and W are the on-site and site-site e-e interactions, respectively.

$H_{l(r)}$ is the Hamiltonian of the left (right) electrode, which is a semi-infinite metal chain,

$$H_{l(r)} = \sum_{m,s}\varepsilon_{l(r)}a^+_{m,s}a_{m,s} + \sum_{m,s}t_{l(r)}(a^+_{m,s}a_{m+1,s} + \mathrm{H.c.}), \tag{3}$$

where $\varepsilon_{l(r)}$ is the on-site energy of a metal atom comparing to that of a carbon atom and $t_{l(r)}$ is the nearest-neighbor transfer integral.

The coupling between the electrodes and the molecule is assumed to take place only at the nearest connecting atoms,

$$H_{coup} = \sum_s t_{lM,s}(a^+_{l,s}c_{1,s} + \mathrm{H.c.}) + \sum_s t_{rM,s}(a^+_{N,s}c_{r,s} + \mathrm{H.c.}), \tag{4}$$

where the interfacial coupling $t_{l(r)M,s}$ is spin dependent. N is the number of the sites of the organic ferromagnetic interlayer.

The last term is the contribution of the external field from the bias voltage between the two electrodes,

$$\begin{aligned} H_E = &-\sum_{n,s}|e|E\left[\left(n-\frac{N+1}{2}\right)a + u_n\right]c^+_{n,s}c_{n,s} \\ &+ \sum_n |e|E\left[\left(n-\frac{N+1}{2}\right)a + u_n\right], \end{aligned} \tag{5}$$

with e being the electronic charge and a the lattice constant. The first term is the electric potential energy of the electron at the nth site and the second term is the electric potential energy of lattice sites (ions). It is assumed that each site provides one π electron in the pristine state and so the site is positive single charged. The field is uniform along the whole molecule chain and $E=V/[(N-1)a]$.

The spin-dependent current is calculated through the Landauer-Büttiker formula

$$I_s(V) = \frac{2e}{h}\int_{-\infty}^{+\infty} T_s(E,V)[f(E-\mu_l) - f(E-\mu_r)]dE \tag{6}$$

where $f(E-\mu_{l(r)}) = 1/\{1+\exp[(E-\mu_{l(r)})/k_BT]\}$ is the Fermi function and $\mu_{l(r)}=E_f \pm eV/2$ is the chemical potential of the left (right) metal electrode with the Fermi energy E_f.

The transmission probability $T_s(E,V)$ is determined from the lattice Green's function[23] according to the Fish-Lee relation[19,23] as $T_s(E,V)=\mathrm{Tr}[\Gamma_l G^R_s(E,V)\Gamma_r G^A_s(E,V)]$, where $G^A_s(E,V)=[G^R_s(E,V)]^+$ and $G^R_s(E,V)=1/(E-H_{OF}-H_E-\Sigma_l-\Sigma_r)$, with Σ_l and Σ_r being the self-energy operators due to coupling to the left and right electrodes.

The calculation is performed as follows: using the mean-field approximation to deal with the e-e interaction, we first solve the electronic eigenequation in the Wannier space. By minimizing the total energy, the lattice and electron structures of an isolated poly-BIPO molecule can be obtained self-consistently under an external electric field. Then, the Green's function and the transmission probability are calculated with spin s, and finally the spin-dependent current is obtained from Eq. (6).

In this paper, the parameters are chosen as being generally used,[21,22] $t_0=2.5$ eV, $\alpha=4.10$ eV/Å, $K=21.0$ eV/Å^2, $U=1.0$ eV, and $W=U/3$ for the organic ferromagnet, which appropriately describes the properties of poly-BIPO; $\varepsilon_{l(r)}=0$, $t_l=t_r=2.5$ eV, and $E_f=1.5$ eV for the electrodes. A low temperature of $T=11$ K is taken to avoid the spin flip in the electron-transport progress.

III. RESULTS AND DISCUSSION

In a real M/OF/M device, there is a good interfacial contact due to the self-adjusting capability of the organic mate-

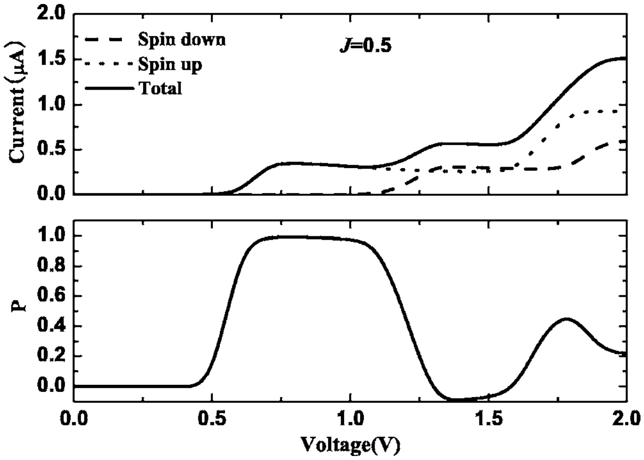

FIG. 2. (a) The calculated current-voltage characteristics of our model device. (b) Spin polarizations of the currents as a function of bias.

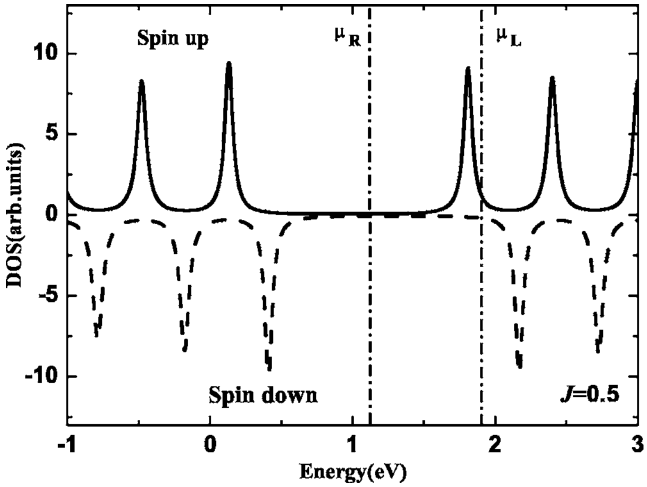

FIG. 3. The density of states (DOS) of the organic-ferromagnet molecule near the Fermi energy at a bias voltage of 0.8 V.

rial. However, the interfacial coupling may be spin dependent, as exemplified by the spin-related density of states in the organic ferromagnet. Here, we first consider the case of spin-independent interfacial coupling and set $t_{lM,s}=t_{rM,s}=0.5$ eV. An organic-ferromagnet interlay consisting of 20 sites is considered here. For the organic molecule, at ground state, all the radical spins orient in the same direction (down, for example) and there is an alternating spin distribution along the main chain of the molecule.[21,22] The total spin of the main chain has a tendency for spin up as there is an antiferromagnetic coupling $J=J_f/t_0>0$ between the radical and the main chain. Defining the spin polarization (SP) of the current as $P=(I_\uparrow-I_\downarrow)/(I_\uparrow+I_\downarrow)$, we calculate the dependence of the SP on the bias at $J=0.5$, with result shown in Fig. 2(b). Figure 2(a) gives the spin-related and the total currents, from which we can obtain the threshold voltage of the device. It is found that the SP oscillates. A SP of nearly 100% first appears when the bias increases to 0.5 V. Then, it drops to nearly zero when the bias is over about 1.2 V for the above parameters. The second peak of the SP appears when the bias reaches about 1.8 V, but in this case, the current becomes only partly spin polarized and the SP reaches about 40%. With the further increasing of the bias, more peaks of the SP will appear (not shown in the figure), while the amplitudes of the SP become smaller and smaller. In order to understand this phenomenon, we calculate the spin-related density of states (DOS) from the Green's function with $\mathrm{DOS}_s(E,V)=-(1/\pi)\mathrm{Im}[G_s^R(E,V)]$. A result at a bias of 0.8 V is shown in Fig. 3, where $\mu_{l(r)}$ represents the chemical potential of the electrodes, and the states falling into the region $\mu_r<E<\mu_l$ mainly contribute to the current. The transmission is provided basically only by the states with spin up, while the spin-down tunnels are not turned on. Therefore, in this case, the current is nearly fully polarized with 100% of the SP. When the bias is increased, the density of states with spins up and down increases alternately in the conducting region, which results in the oscillation of the spin-related transmission and current. It is interesting to note that a negative SP is allowable, which is related to the transmission coefficients of the conducting tunnels under a given bias. As shown in Fig. 2(b), at a bias of around 1.4 V, both the spin-up and spin-down lowest unoccupied molecular orbitals (LUMOs) of the organic interlayer fall into the conducting region. The transmission with spin up may be a little smaller than that with spin down, which results in a negative SP. It should also be mentioned that the present method is only valid when the external bias is not too large, as a uniform distribution of the field along the tunnel is assumed and an equilibrium Green's function is adopted in the calculation.

Then, we calculate the dependence of the SP on the spin-correlation parameter J. The result is shown in Fig. 4, where the bias is fixed at 0.8 V. The SP increases rapidly and then reaches its maximum value at about $J=0.25$, which means that a weak spin correlation of the organic interlayer will result in an apparent spin-polarized current. The effect of J on the spin-dependent transport can be understood as follows: without spin correlation, the electronic states in the molecule are spin degenerate and the SP of current is zero. In the presence of J, the electronic density on each site with different spins is affected by J and a spin-density wave ap-

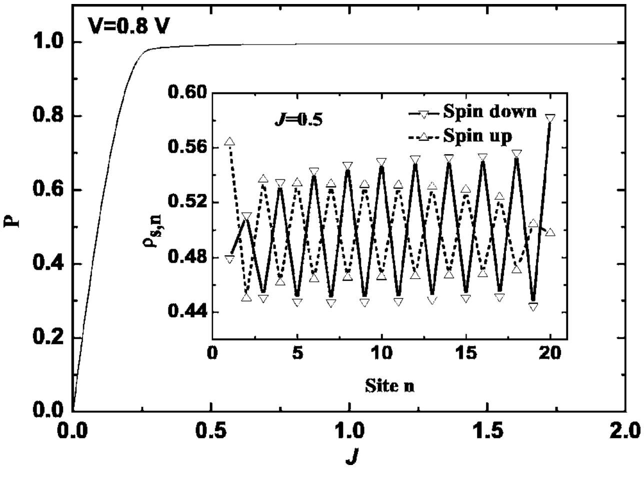

FIG. 4. Spin polarization versus spin-correlation strength J at a bias voltage of 0.8 V. The inset plot is the electronic density with different spins at $J=0.5$.

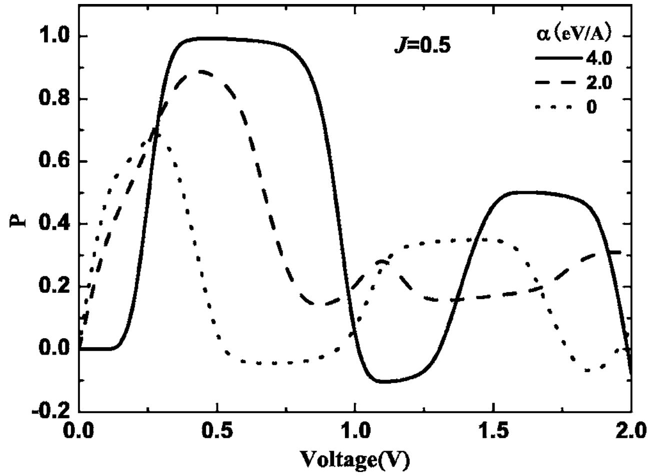

FIG. 5. The current spin polarizations as a function of bias with different e-la coupling constants.

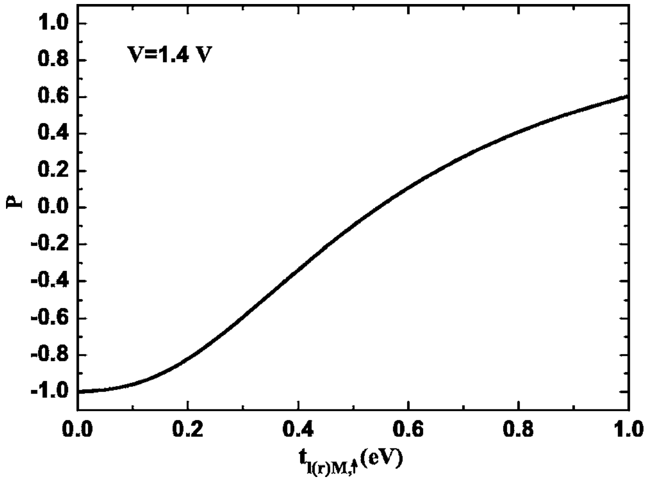

FIG. 6. The dependence of spin polarization on the interfacial coupling parameter $t_{l(r)M,\uparrow}$ at a bias of 0.8 V. Here, $t_{l(r)M,\downarrow}$ is fixed at 0.5 eV. Other parameters are the same as those in Fig. 2.

pears, as displayed in the inset plot of Fig. 4. Here, the electronic density is defined as $\rho_{s,n}=\Sigma'_{\mu}Z_{\mu,n,s}Z_{\mu,n,s}$, where $Z_{\mu,n,s}$ is related to the wave function through $\psi_{\mu,s}=\Sigma_n Z_{\mu,n,s}|n\rangle$, which can be obtained by solving the Schrödinger equation of the electronic Hamiltonian. The resulting spin split of energy levels decreases the number of spin-down states in the conducting window, while increases the spin-up one, which leads to the increase of SP. When the spin-down states are completely ruled out of the conducting window, a nearly 100% SP is achieved. Due to the existence of large Peierls gap, only spin-up states in the conducting window conduct current when J gets larger. The SP will remain unchanged near 100%.

Softness is a key factor for the performance of an organic device. Here, we study the effects of the e-la interaction in the organic material and the interface between organic layer and the metal electrode on the SP. Figure 5 shows the dependence of the SP on the bias with three different values of the e-la interaction at $J=0.5$. $\alpha=0$ means a rigid uniform structure of the organic chain with no gap. It is found that the current polarization appears as soon as the bias is applied in the case of a non-($\alpha=0$) or weak e-la interaction ($\alpha=2.0$ eV/Å). However, when a strong e-la interaction ($\alpha=4.0$ eV/Å, for example) is included, there is a voltage gap of about 0.2 V for the current of the device. It is also found that the maximum SP value and the platform width at the maximum polarization increase with the e-la interaction. The reason is the following: a spin correlation (J) leads to a spin splitting of the energy levels, while the e-la interaction induces a Peierls gap separately for the spin-up and spin-down levels. Because the gaps are nonsymmetrical around the Fermi level of the electrode, the numbers of the spin-up and -down levels falling into the conducting region are different and adjusted by the e-la interaction. Therefore, the SP is changed by the e-la interaction. It is interesting to note that a nearly 100% SP is obtained, as shown by the first platform plotted by solid line in Fig. 5. In our calculation, there is a Peierls gap of about 1.65 eV between the LUMO and highest occupied molecular orbital of spin-up (-down) levels at $\alpha=4.0$ eV/Å. In the presence of spin splitting, the spin-down levels are away from the Fermi energy rather than the spin-up one. When the bias increases to 0.3 V, only the nearest spin-up level (LUMO) falls into the conducting region, which means that only the spin-up tunnel is opened and the spin-down tunnels are blocked by the gap. However, in the case of non- or weak e-la interaction (no gap), both the spin-up and spin-down levels are near the Fermi energy and have contribution to the current. Thus, it is difficult to get a fully polarized current. An analogous conclusion has been reported in a double-bend structure of a quantum wire recently, which utilizes the antiresonance gap generated by weak lateral magnetic modulations to realize a large spin-polarized current.[24] Therefore, a large current polarization is expected in an organic-ferromagnetic device.

Then, we consider the interfacial effect on the SP. It is well known that an organic material can form an adjustable contact with the electrodes due to the softness of the organic material. For the M/OF/M device, the interfacial coupling may be spin dependent. For simplicity, we fix $t_{l(r)M,\downarrow}=0.5$ eV, and calculate the dependence of the SP on $t_{l(r)M,\uparrow}$ at a given bias. If only one kind of spin tunnel is open, the current will be fully spin polarized as the other tunnel is closed. If both spin tunnels are open, it is found that the SP will be dependent upon the interfacial coupling. The result is shown in Fig. 6, where the bias is selected at 1.4 V. Apparently, at $t_{l(r)M,\uparrow}=0$, the spin-up electrons could not be injected into the organic interlayer and only the spin-down electrons are injected. Even assuming the spin-down electrons may reverse their direction during the transport, they cannot be ejected as the spin-up tunnel is closed at the right interface. Therefore, the current is fully polarized in this case. With the opening of the spin-up tunnel at the interface, the current becomes partly spin polarized. When the spin-up interfacial coupling is much larger than the spin-down coupling, again it will be found that the current becomes nearly fully polarized but with a negative SP compared to that at $t_{l(r)M,\uparrow}=0$. The result shows that by adjusting the spin-dependent interfacial coupling (for example, realized by a spin-related assistant interlayer between the electrode and the organic layer, or by

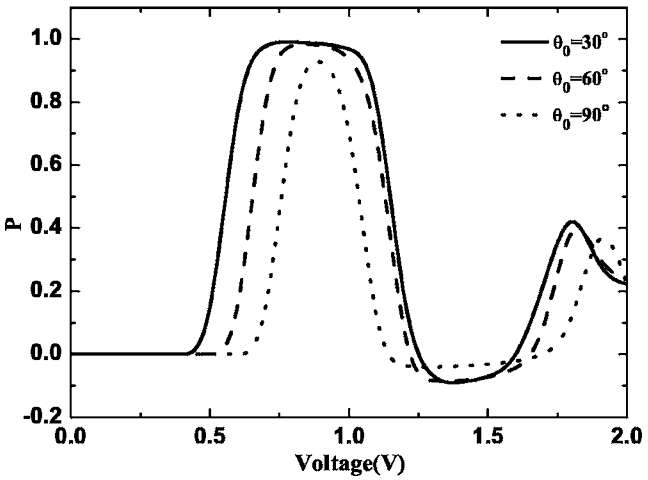

FIG. 7. The effect of orientation fluctuation of radical spins on the current spin polarization.

applying a magnetic field at the interface), we can change the current polarization.

In an actual device, the thermal fluctuation will destroy the ground-state order of the radical spins. Here, we consider the effect of the orientation fluctuation of the radical spins on the current polarization. The fluctuation is described by the angle θ of the radical spin from its ground-state order, where we use a square-random distribution from 0 to θ_0. The value of θ_0 is taken as 30°, 60°, and 90°, respectively. In each case, an average over a hundred computed samples is made and the results are shown in Fig. 7. It is found that the region of the 100% polarization shrinks with the fluctuation. The 100% SP could not be achieved at a large fluctuation ($\theta_0 = 90°$). The reason is that the fluctuation of the residual spin will weaken the spin interaction between the radical and the main chain, thus resulting in a reduction of the current polarization.

IV. SUMMARY

In summary, we have performed a theoretical study of the coherent charge transport through a metal/organic-ferromagnet/metal sandwich structure. Extremely large spin-polarized current is predicted through this device. It is found that the spin correlation between the residual spins of the radical *R*'s and the spins of the π electrons in the main chain are the sources that generate the SP. A detailed study exploits the effects of softness of such organic material on the SP. One effect is that the large Peierls gap induced by the intrinsic strong e-la interaction leads to the appearance of 100% SP. Another is the spin-dependent interfacial coupling, which affects not only the magnitude of SP but also its polarized orientation apparently. The spin filtering effect is still obtained even when a thermal fluctuation of radical spins is included. Our studies suggest that organic ferromagnets may have advantages for the application of a spin filter device for its intrinsic properties, in particular, in molecular circuits. It should be pointed out that the effect of thermal fluctuation of lattice sites is not included in the present work, which will influence the lattice dimerization and the Peierls gap. In addition, there may exist spin flipping on the radical and, in this case, the system is in a spin excited state. A comprehensive consideration about spin flipping should include the mode of flipping, such as the position and the number of flipped spins. All these factors will affect the current polarization of the device and we will have further investigation in our next work.

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