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Spin precession of a polaron induced by a gate voltage in one-dimensional organic polymers

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Abstract

We theoretically investigated the spin dynamics of a polaron under a gate voltage in a one-dimensional organic polymer chain. Spin precession is obtained when a polaron transports along the polymer chain. The spin precession has a certain periodic length, which is in inverse proportion to the strength of the spin-orbital interaction. It was found that spin precession takes place only on the polaron level (intralevel spin precession), while in a rigid semiconductor spin precession takes place among all the conduction levels (interlevel spin precession). In addition, the periodic length of polaron spin precession becomes longer for stronger electron-lattice interaction, indicating long spin relaxation in organic polymers.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In the past few years, the potential applications of electron spin in mesoscopic devices have generated a lot of interest. π -Conjugated organic semiconductors are emerging as an important platform for 'spintronics', which involves the spin freedom of a charge carrier to store, process and communicate information. The underlying idea is to electrically drive a modulated spin-polarized current. One of the simplest solutions is to employ a spin-valve where the output current is governed by switching the spin-polarization in one of the ferromagnetic contacts. With the development of organic spintronics, low-dimensional organic spin-valves have been widely studied, such as thin-film spin-valves [1], molecular spin-valves [2] and nanowire organic spin-valves [3]. Another possibility is the so-called spin field effect transistor [4], in which the spin-polarized current is injected from one ferromagnetic electrode and then collected by another. During the transport process, the spin-orbital interaction induces the spin precession of electrons in the transport layer.

Consequently, by tuning the ejection state of the *spin precession* with respect to the magnetization direction of the ferromagnetic collector, the transmitted spin-polarized current is modulated accordingly. However, most of the previous studies only focused on the rigid semiconductors and spin-orbital interaction is believed to be weak in organic semiconductors. The effect of spin-orbital interaction was often neglected during the investigation of spin transport in organic devices.

The spin-orbital interaction, which induces the spin precession, has attracted much attention in recent years since it plays an important role in the new field of inorganic semiconductor spintronics [5]. Rashba spin-orbital interaction is more interesting due to its capability of electrically controlling the interaction strength. Datta and Das have theoretically analyzed the Rashba spin-orbital interaction in a spin transistor that induced spin precessions in a semiconductor [4]. Tuning of the spin-orbital interaction by an external gate voltage has been realized in an InGaAs/InAlAs quantum well [6]. Recently, it was reported that strong Rashba spin-orbital interaction induces nontrivial subband intermixing

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in quasi-one-dimensional nanowires [7]. In addition, the Rashba interaction in nanostructures gives rise to an interlevel spin-flip term rather than any intralevel spin flips [8].

Different from a rigid semiconductor, the electron–lattice interaction is strong in an organic semiconductor, indicating that charge accumulations will cause a large lattice distortion. The softness of an organic semiconductor induces its carriers to be composite particles with an internal structure characterized by lattice distortion, such as charged spin polarons and charged spinless bipolarons. The formation of polarons leads to hysteretic conductance switching, negative differential resistance effect and giant magnetoresistance in organic spin-valve systems [9, 10]. In view of the important role that polarons play in these spin related devices, it is necessary to study the flipping character of a spin polaron. In this paper we theoretically investigate the transport dynamic process of a spin polaron under a gate voltage in organic polymers. Calculated results show that the spin orientation of a polaron precesses with a certain periodic length during its transport. However, it was found that spin precession takes place only on the polaron level (intralevel spin precession), while in a rigid semiconductor the spin precession takes place among all the conduction levels (interlevel spin precession). In addition, the spin precession periodic length of a polaron under electron–lattice interaction is much larger than that of an electron in a rigid semiconductor. Such results also indicate the long spin relaxation of organic polymers. In section 2 the model is described and the dynamical evolution method is presented. The numerical results and our main conclusions are given in section 3. Finally, in section 4 we summarize our contents.

2. Theoretical model

In this paper we consider a one-dimensional non-degenerate polymer chain. A gate voltage is employed to control the spin–orbital interaction which induces the spin flip and spin precession of moving carriers. By applying the gate voltage along \vec{z} axis, the related Hamiltonian takes the form [11, 12]

$$H_{\text{so}} = -\frac{\beta}{\hbar} \vec{\sigma} \cdot (\vec{p} \times \vec{z}) = i\beta \left(\sigma_x \frac{\partial}{\partial y} - \sigma_y \frac{\partial}{\partial x} \right), \quad (1)$$

where β represents the spin–orbital interaction strength related to the gate voltage and $\vec{\sigma}$ is the Pauli matrix. Although this kind of spin–orbital interaction is an assumption in the absence of direct experimental proof so far, it is also meaningful to study the modification of the spin–orbital interaction by the gate voltage. For a one-dimensional polymer chain along the \vec{x} axis, only the second term in the bracket of equation (1) is left. By the tight-binding approach, the spin–orbital interaction in the following second quantization form is obtained [7, 8]:

$$H_{\text{so}} = -t_{\text{so}} \sum_n \left[C_{n+1,\uparrow}^\dagger C_{n,\downarrow} - C_{n+1,\downarrow}^\dagger C_{n,\uparrow} + \text{h.c.} \right], \quad (2)$$

where only the nearest-neighbor spin-dependent interaction in the perturbation is considered. The isotropic nearest-neighbor transfer integral t_{so} denotes the spin–orbital interaction strength with $t_{\text{so}} = \beta/2a$ [7], in which a is the lattice constant.

$C_{n,s}^\dagger (C_{n,s})$ is the electron creation (annihilation) operator on site n with spin s , where $s = \pm 1$ denotes spin-up and spin-down, respectively. Equation (2) shows that the spin–orbital interaction includes both the flipping and hopping of the spin carrier, which results in the spin precession during the electron transport². Therefore the spin flipping and precession are closely related to the transport of the carriers.

For a one-dimensional non-degenerate polymer, the electronic part of the Hamiltonian

$$H_e = H_0 + H_{\text{so}}, \quad (3)$$

$$H_0 = - \sum_{n,s} t_{n,n+1} \left(e^{-iyA} C_{n+1,s}^\dagger C_{n,s} + \text{h.c.} \right), \quad (4)$$

$$H_{\text{so}} = -t_{\text{so}} \sum_n \left[e^{-iyA} (C_{n+1,\uparrow}^\dagger C_{n,\downarrow} - C_{n+1,\downarrow}^\dagger C_{n,\uparrow}) + \text{h.c.} \right]. \quad (5)$$

Here H_0 is the tight-binding model in the absence of a gate voltage plus a driving electric field. The driving electric field along the chain is added to drive the spin polaron to move along the chain, and is introduced through the vector potential A appearing in a complex phase factor in the transfer integral [13]. During the calculations, periodic boundary conditions are assumed to obtain a long time motion of the polaron. In the presence of this driving electric field, the form of H_{so} is also changed correspondingly. The parameter $t_{n,n+1} = t_0 - \alpha(u_{n+1} - u_n) - (-1)^n t_e$ is the hopping integral of π electrons between atom (site) n and $n + 1$ [14]. t_0 denotes the zero-displacement hopping integral between the n th and $(n + 1)$ th atoms. α is the electron–lattice coupling constant and u_n the displacement of the n th site from its equidistant position. t_e is the Brazovskii–Kirova symmetry breaking term for a non-degenerate polymer [15]. Coefficient γ in the exponent is defined as ea/\hbar , with e and a being electron charge and lattice constant, respectively. The electric field is then given by $E(t) = -\partial_t A(t)$ [13]. Here the internal electron–electron interactions are neglected. It has been predicted that the electron–lattice interaction will dominate the properties of polymers if the electron–electron interactions are not too strong [16, 17]. Due to the soft character of the polymer, the movement of charges will cause the distortion of the lattice. Considering the elastic and kinetic energy of the lattice, the lattice part of the Hamiltonian is described classically by [14]

$$H_{\text{latt}} = \frac{K}{2} \sum_n (u_{n+1} - u_n)^2 + \frac{M}{2} \sum_n \dot{u}_n^2, \quad (6)$$

where K is the elastic constant and M the mass of a CH group.

From the Hamiltonian, we can obtain the dynamics of the electronic state of the π -electrons from the quantum Schrödinger equation and the corresponding lattice distortion from the classical Newton equation [13]. The dynamics is described within the mean-field approximation where transitions between instantaneous eigenstates are allowed [18] in contrast to so-called adiabatic dynamics with fixed level occupation [19]. The evolution of the μ th electronic state

² Different from this, Larmor precession caused by a magnetic field only contains onsite spin flip which is independent of the carrier movement.

$|\Psi_\mu(t)\rangle$ with initial value $|\Psi_\mu(t=0)\rangle$ is obtained by solving the time-dependent Schrödinger equation

$$i\hbar\dot{\psi}_{\mu,s}(n,t) = -t_{n-1,n}e^{-iyA}\psi_{\mu,s}(n-1,t) - t_{n,n+1}e^{iyA}\psi_{\mu,s}(n+1,t) - s \cdot t_{so} \times e^{-iyA}\psi_{\mu,-s}(n-1,t) + s \cdot t_{so}e^{iyA}\psi_{\mu,-s}(n+1,t), \quad (7)$$

with

$$|\Psi_\mu(t)\rangle = \begin{pmatrix} |\Psi_{\mu,\uparrow}(t)\rangle \\ |\Psi_{\mu,\downarrow}(t)\rangle \end{pmatrix} = \begin{pmatrix} \sum_n \psi_{\mu,\uparrow}(n,t)|n\rangle \\ \sum_n \psi_{\mu,\downarrow}(n,t)|n\rangle \end{pmatrix}, \quad (8)$$

and $|n\rangle$ is the Wannier state. The Newton equation related to the lattice displacement is

$$M\ddot{u}_n(t) = \alpha\{e^{iyA}[\rho_{n,n+1}(t) - \rho_{n-1,n}(t)] + \text{c.c.}\} - K[2u_n(t) - u_{n+1}(t) - u_{n-1}(t)]. \quad (9)$$

The charge density matrix $\rho_{n,n'}$ and the density matrix of spin $s (s = \pm 1)\rho_{n,n'}^s$ are defined as

$$\rho_{n,n'}^s(t) = \sum_\mu \psi_{\mu,s}^*(n,t) f_\mu \psi_{\mu,s}(n',t), \quad (10)$$

$$\rho_{n,n'}(t) = \rho_{n,n'}^s(t) + \rho_{n,n'}^{-s}(t), \quad (11)$$

where f_μ is the time-independent distribution function determined by initial occupation (being 0, 1 and 2). The coupled differential equations (7) and (9) can be solved numerically with a Runge–Kutta method of order eight with step-size control [20], which has been proven to be an effective approach to study the electronic state evolution [13, 21].

We choose the initial state as the eigenstate of a polaron, which is given by iteratively solving the static Schrödinger equation and self-consistent equilibrium condition in the absence of driving electric field. The initial electronic state given by the static Schrödinger equation is

$$\varepsilon_\mu \psi_{\mu,s}(n,t=0) = -t_{n-1,n}\psi_{\mu,s}(n-1,t=0) - t_{n,n+1}\psi_{\mu,s}(n+1,t=0) - s \cdot t_{so} \times \psi_{\mu,-s}(n-1,t=0) + s \cdot t_{so}\psi_{\mu,-s}(n+1,t=0). \quad (12)$$

The initial site displacements given by the self-consistent equilibrium condition are

$$u_n(t=0) = \frac{\alpha}{K}[\rho_{n+1,n}(t=0) - \rho_{n,n-1}(t=0)] + [u_{n-1}(t=0) + u_{n+1}(t=0)]/2. \quad (13)$$

Selecting parameters referring to those of *cis*-polyacetylene with no loss of universality [22], $t_0 = 2.5$ eV, $\alpha = 4.1$ eV Å⁻¹, $t_e = 0.05$ eV, $K = 21$ eV Å⁻², $a = 1.22$ Å, $M = 1349.14$ eV fs² Å⁻². Since a polaron will dissociate as the driving electric field is higher than 6×10^{-3} V Å⁻¹ [23], we set $E_0 = 5 \times 10^{-5}$ V Å⁻¹ for the numerical simulation, which is small enough to maintain the polaron state during its movement. It has been pointed out that the behavior of a polaron is mode-independent when the electric field is lower than 1.4×10^{-4} V Å⁻¹ [24]. In calculations, the driving electric field $E(t)$ is linearly applied, $E(t) = E_0/t_c \cdot t$ for $0 < t < t_c$ and $E(t) = E_0$ for $t > t_c$, respectively, with $t_c = 30$ fs being a turn-on period.

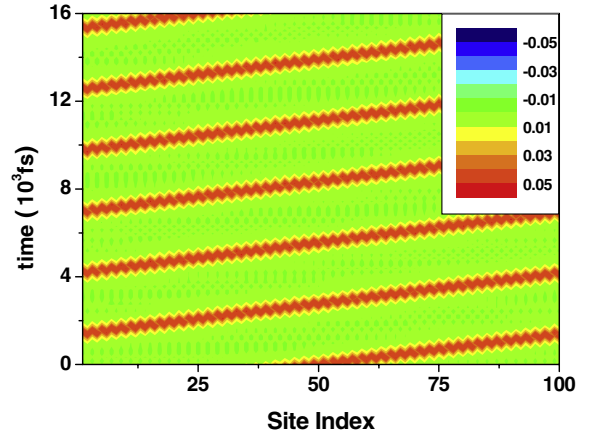


Figure 1. Spin evolutions of the polymer without spin–orbital interaction. The positive points denote spin-up states, negative points denote spin-down, and zero denotes spinless.

3. Results and discussion

Here we briefly describe the object to be studied. A spin-up electron (+1/2 along the \vec{z} axis) is assumed to be injected into the polymer chain with a period of $N = 100$ sites forming a spin polaron. Then the polaron is the self-trapping state of one electron and it carries one electronic charge unit with spin 1/2. An electric field to drive the polaron is applied along the $-\vec{x}$ direction. First, the spin evolution of the polaron without the gate voltage is represented by

$$S_n^Z(t) = \frac{1}{2}[|\psi_{p,\uparrow}(n,t)|^2 - |\psi_{p,\downarrow}(n,t)|^2] \quad \text{or} \quad \frac{1}{2}[\rho_{n,n}^\uparrow(t) - \rho_{n,n}^\downarrow(t)]. \quad (14)$$

The polaron transports along the polymer chain with a width full wave at half maximum (FWHM) of about 9.4 sites and a velocity of 0.036 site fs⁻¹. Since there is no spin related interaction, the spin keeps its orientation unchanged while the position of polaron changes during its movement, as shown in figure 1.

Then the gate voltage is applied to build the spin–orbital interaction with $t_{so} = 0.02$ eV, which is treated as a perturbation compared to the other interactions. It was found that the trivial spin–orbital interaction can scarcely change the energy band structure of the system. Therefore, the polaron transports along the polymer chain with the same width and velocity as the case without the gate voltage. Interestingly, a spin precession is induced which is similar to the phenomena in rigid semiconductors [7], and has never been reported before in organic polymers. The spin evolution with the gate voltage is shown in figure 2(a). The spin of the polaron $S^Z(t) = \sum_n S_n^Z(t)$ in the uppermost line decreases as the polaron transports along the polymer chain. And the trace of polaron is continually shown in the second line under the periodic boundary condition. With the evolution, the spin of the polaron became zero and even spin-down. The spin of the polaron in the third trace reached the maximum of spin-down orientation at about 5000 fs, which means that the projection of the spin is completely flipped. Then the spin is reversed again and the process restarts.

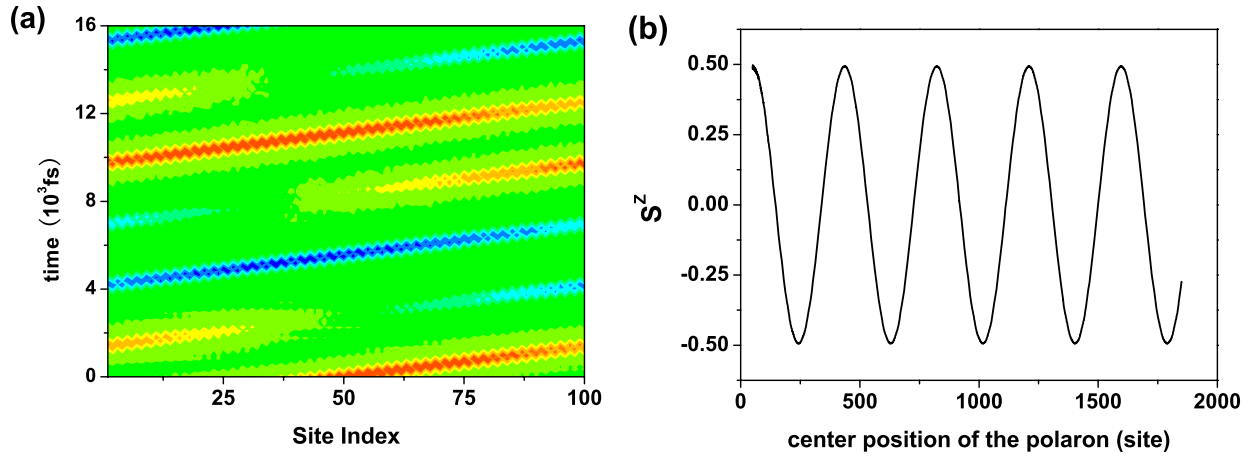


Figure 2. (a) Spin evolutions of the polymer with the spin–orbital interaction ($t_{so} = 0.02$ eV). The color scale is the same as in figure 1. (b) Dependence of spin precession on the center position of the moving polaron.

In addition, a polaron has a particle-like characteristic with a well-defined position. Therefore it is convenient to display the periodic length of the precession in organic semiconductors rather than rigid semiconductors, in which traces of the electrons are extended. For a clear view of the spin precession, we define the center of the polaron state x_c by

$$x_c = \begin{cases} N\theta/2\pi, & \text{if } \langle \cos\theta_n \rangle \geq 0 \text{ and } \langle \sin\theta_n \rangle \geq 0; \\ N(\pi + \theta)/2\pi, & \text{if } \langle \cos\theta_n \rangle < 0; \\ N(2\pi + \theta)/2\pi, & \text{otherwise.} \end{cases} \quad (15)$$

Here

$$\theta = \arctan \frac{\sin \theta_n}{\cos \theta_n}, \quad (16)$$

the averages of $\sin \theta_n$ and $\cos \theta_n$ are defined as

$$\begin{aligned} \langle \sin \theta_n \rangle &= \sum_{n,s} |\psi_{p,s}(n, t)|^2 \sin \theta_n, \\ \langle \cos \theta_n \rangle &= \sum_{n,s} |\psi_{p,s}(n, t)|^2 \cos \theta_n, \end{aligned} \quad (17)$$

and $\psi_{p,s}(n, t)$ is the electronic state of the polaron. This center is in superposition with that of the charge density of the system [13], defined by

$$\begin{aligned} \langle \sin \theta_n \rangle &= \sum_{n,s} (\rho_{n,n}^s - 0.5) \sin \theta_n, \\ \langle \cos \theta_n \rangle &= \sum_{n,s} (\rho_{n,n}^s - 0.5) \cos \theta_n. \end{aligned} \quad (18)$$

The instantaneous projection of $S^Z(t)$ of the polaron during its transport is shown in figure 2(b). It was shown that the spin evolution behaves as a cosinoidal rule with a precession periodic length of about 385 sites in the present parameters.

By adjusting the gate voltage, the strength of the spin–orbital interaction can be tuned. Although the polaron transports with the same velocity and width, its spin precesses faster for a stronger spin–orbital interaction. It was shown in figure 3 that the corresponding periodic length of the

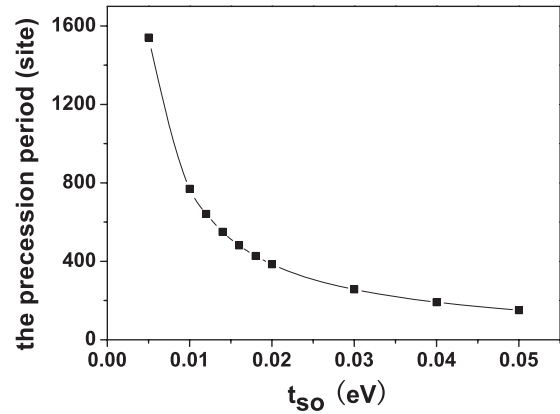


Figure 3. The periodic length of the precession for different strength of spin–orbital interaction in the organic semiconductor.

precession is in inverse proportion to the strength of the spin–orbit interaction. That is the spin detected at the same place is different, and can be controlled by the gate voltage. This can also be used in organic spin field effect transistors.

To explicitly understand the spin precession of polaron, we expand $|\Psi_\mu(t)\rangle$ in a basis of the instantaneous eigenstates $|\Phi_\nu(t)\rangle$,

$$|\Psi_\mu(t)\rangle = \sum_\nu C_{\mu,\nu} |\Phi_\nu(t)\rangle, \quad (19)$$

where

$$C_{\mu,\nu} = \begin{pmatrix} C_{\mu,\nu}^\uparrow & 0 \\ 0 & C_{\mu,\nu}^\downarrow \end{pmatrix}. \quad (20)$$

The eigenwavefunction of the ν th instantaneous eigenstate

$$|\Phi_\nu(t)\rangle = \begin{pmatrix} |\Phi_{\nu,\uparrow}(t)\rangle \\ |\Phi_{\nu,\downarrow}(t)\rangle \end{pmatrix} = \begin{pmatrix} \sum_n \phi_{\nu,\uparrow}(n, t) |n\rangle \\ \sum_n \phi_{\nu,\downarrow}(n, t) |n\rangle \end{pmatrix} \quad (21)$$

is determined by the instantaneous eigen Schrödinger equation at that moment

$$H_e(t) |\Phi_\nu(t)\rangle = E_\nu |\Phi_\nu(t)\rangle. \quad (22)$$

The occupation probability on the ν th instantaneous eigenstate with spin s can be given by $\sum_\mu f_\mu |C_{\mu,\nu}^s|^2$. The self-trapping

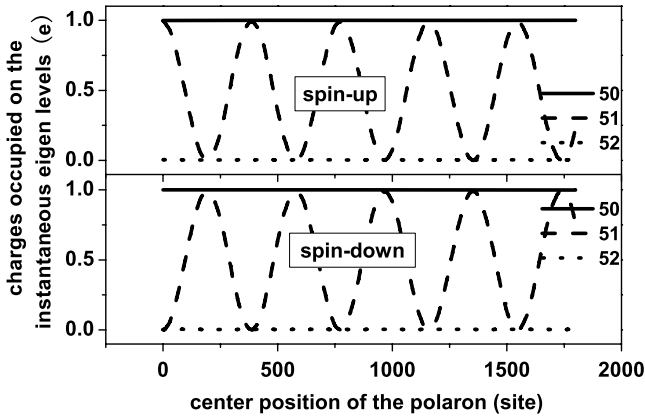


Figure 4. The charge with spin-up and spin-down occupying on the instantaneous eigenlevels during movement of the polaron. $\nu = 50$ (solid lines) and $\nu = 51$ (dashed lines) denote the lower polaron level and the higher polaron level, respectively. And $\nu = 52$ (dotted lines) denotes the lowest unoccupied one.

polaron state results in two deep levels appearing in the energy gap. The upper level of the two ($\Psi_u(t)$ or $\Psi_p(t)$) is singly occupied and the lower one ($\Psi_d(t)$) is doubly occupied. Figure 4 shows the occupation probability of some instantaneous eigenstates, which correspond to the lower polaron level ($\nu = 50$, doubly occupied), the higher polaron level ($\nu = 51$, singly occupied) and the lowest unoccupied one ($\nu = 52$), respectively. It was found that the electronic states on the lower polaron level make no contribution to the spin precession since it is doubly occupied. Although the higher polaron level is always singly occupied, the occupation probability of spin-up and spin-down alternately changes during the evolution, which induces the spin precession of the polaron. Spin precession in the polymer layer takes place only within the polaron level (intralevel). This is different from the case of a rigid semiconductor, where the spin precession takes place among all the conduction levels (interlevel) [8].

In the following we briefly discuss the dependence of polaron spin precession on the system parameters. It was found that the driving electric field shows a small impact on the periodic length of the spin precession. This can be interpreted that in organic polymers the polaron moves as an entity with its charge and lattice configuration coupled. The driving electric field only affects the velocity of the polaron. For a perpendicular gate voltage applied on a carrier, the Hamiltonian is [8]

$$H_{so} \propto \frac{1}{m^*} \frac{dV_G}{dz} \vec{\sigma} \cdot (\vec{v} \times \vec{z}) = \frac{1}{m^{*2}} \frac{dV_G}{dz} \vec{\sigma} \cdot (\vec{p} \times \vec{z}), \quad (23)$$

where v is the velocity of the polaron. With a definite gate voltage applied, the spin precession period time $T \sim m^*/B_{\text{eff}} \sim m^*/v$. The spin precession period T and length λ have a relation $\lambda = vT$, so that λ remains unchanged for the same polaron. However, the spin precession of a polaron and the interaction strength β or t_{so} are also sensitively related to the electron–lattice interaction, which induces a heavy effective mass of the carriers, besides the gate voltage. A polaron is naturally a localized spin carrier

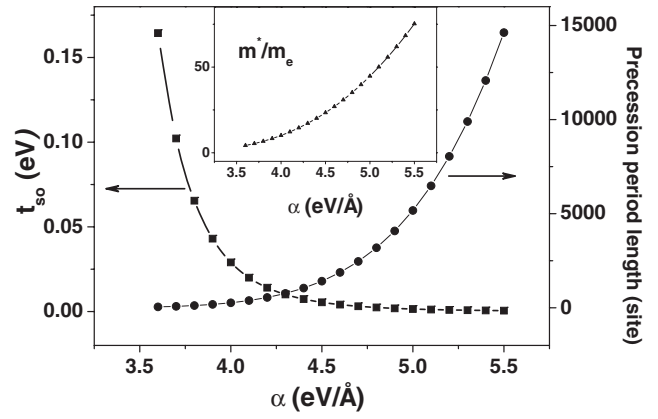


Figure 5. The dependence of the spin precession periodic length (solid circle) and corresponding spin–orbital interaction (solid square) on electron–lattice coupling. The inset shows the dependence of polaron effective mass on electron–lattice interaction.

due to the inherent strong electron–lattice interaction. The electron–lattice interaction couples the charge and the lattice together, resulting in a heavy effective mass of the carriers in organic polymers. For instance, the polaron effective mass in polyacetylene is $m^* \approx 11.7m_e$ [25]. In addition, the polaron effective mass varies with different electron–lattice interaction strengths. According to [25], the dependence of the polaron effective mass m^* on electron–lattice interaction α is shown in the inset of figure 5. The polaron effective mass becomes larger for stronger electron–lattice interaction, due to the strong coupling between lattice and charge. Furthermore, the dependence of λ and spin–orbital interaction t_{so} on electron–lattice interaction α is given in figure 5. It was obtained from equation (23) that $\beta \sim 1/m^{*2}$, and from figure 3 that the spin precession periodic length $\lambda \sim 1/\beta$ or ($\lambda \sim 1/t_{so}$), therefore $t_{so} \sim 1/m^{*2}$ and $\lambda \sim m^{*2}$ when the gate voltage remains a constant. The spin precession periodic length λ rapidly becomes large when electron–lattice interaction is strengthened. Therefore, the spin precession in a polymer with strong electron–lattice interaction decreases correspondingly. It should be mentioned that for a polaron under electron–lattice interaction with effective mass $m^* \approx 11.7m_e$, the spin precession periodic length is about 10 000 times larger than that of InGaAs semiconductor with $m^* \approx 0.042m_e$. Such results indicate the long spin relaxation of organic polymers. Recently, Wu and Hu [26] reported that the deposited metal atoms enhance the spin–orbital interaction at the polymer/metal interface. This experiment hints at the possibility of controlling spin–orbital interaction by an external gate voltage which is beneficial for detecting the spin precession in an organic polymer.

4. Conclusion

In summary, the spin dynamics under a gate voltage in a one-dimensional organic semiconductor is investigated. It was found that the spin of a polaron will appear to precess periodically when it moves along the chain. The localization of a polaron (particle-like) makes it easy to probe the dependence

of spin precession on the position of a polaron. By analyzing the electronic states, we conclude that spin precession takes place only within the highest occupied state of the polaron, which is intralevel spin flips compared to the interlevel spin flips in the case of a rigid semiconductor. For a localized spin carrier in a polymer, the periodic length of the spin precession of a polaron is closely related to the strength of the spin–orbital interaction. Therefore, it is possible to control spin transport in an organic spin electric device by the external gate voltage.

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