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Theoretical investigation of voltage effect on magnetoresistance in an organic small molecule device

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ABSTRACT

We theoretically study the voltage effect on organic magnetoresistance (OMAR) in a weak disordered small molecule device on the basis of the quantum dynamics. It is found that with the increase of the voltage, the OMAR effect is reduced. The results show a good agreement with the experimental data. In addition, the carrier density effect on OMAR has also been discussed.

1. Introduction

Recently, there has been a growing interest in the magnetic field effect (MFE) on photocurrent, electroluminescence and electrical-injection current, etc. opto-electronic properties of the non-magnetic organic semiconductor devices [1–3]. Among these researches, the MFE on electrical-injection current is most studied, which is also known as organic magnetoresistance (OMAR) [4–17]. Up to now, the OMAR effect has been observed in various organic semiconductor materials, from polymers like PFO to small molecules like Alq_3. The observed OMAR effect seems to have different performances from small molecules, oligomers to polymers, and from weak to strong magnetic fields [7]. By analyzing the experimental data from different groups, it is summarized that the OMAR curve can be well fitted with the empirical Lorentzian function $B^2/(B^2+B_0^2)$ or the non-Lorentzian function $B^2/(|B|+B_0)^2$ [6,13] or their combination [8,14]. Besides these, other functions such as power law B^n [9] and the polynomial expansion $d_1B^2+d_2B^4$ or $f_1/B^2+f_2/B^4$ [7] can also be used to simulate the OMAR curve. Thus it can be seen that the magnetoresistance (*MR*) response to magnetic field is complex.

During the past decade, several mechanisms have been proposed and some of the above fitting functions are obtained theoretically [13–15]. For example, Sheng et al. succeeded in deducing the empirical law $B^2/(B^2+B_0^2)$ by using the Hamiltonian that includes both the Zeeman interaction between the applied magnetic field and the electronic spin, and the hyperfine interaction between the effective hyperfine field (produced by the hydrogen nuclear spin) and the electronic spin. In their derivation, it is supposed that the current density is proportional to the density of the free charges, and then the Lorentzian fitting is obtained qualitatively. However, from the definition of current density $j=env$, we know that the current density is proportional to not only the carrier density n, but also the carrier velocity v. Therefore, in our previous paper, we investigated the carrier (polaron) velocity response to the magnetic field and understood the OMAR well [16].

Up to now, the mechanism of OMAR effect is still in debate although three generally accepted models have been put forward: (1) Electron–hole pair model, which is based on the singlet/triplet exciton formation from electron–hole pairs [10]. (2) Bipolaron model, which is based on the intercrossing between polarons and bipolarons [11]. (3)

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Exciton-charge interaction model, which is based on the quenching action between triplet excitons and polarons [12]. From these models, some experimental results about OMAR could be well understood qualitatively. During the theoretical investigations, it seems to show that the hyperfine interaction between the injected carrier spin and the hydrogen nuclear spin is vital for the appearance of the OMAR effect [10,11,13].

Recently, we employed a band-like model to explore the polaron mobility response to the applied magnetic field in a weak disordered molecule crystal [16]. Our study gives a quantitative understanding on OMAR effect in a small molecule device, and the calculated results are well consistent with some experimental data. However, in our previous work we did not consider the voltage effect on *MR*, which is usually an important part in the experimental researches. For example, Bloom et al. studied the voltage effect on *MR* in Alq_3, and found that with the increase of the voltage, the *MR* traces show a sign change from positive to negative [17]. Thus they concluded that the voltage can tune the sign of *MR*. However, in Mermer et al.'s investigation *MR* is negative in all voltage range [5]. In addition, they also measured *MR* in different temperatures. The results show that, at 100 K and 300 K, the value of *MR* increases initially and then decreases with the voltage. While at 10 K and 200 K, its value increases and decreases respectively with the voltage. Thus they suggested that the response behavior of *MR* to voltage is temperature-dependent. Martin et al. explored the voltage effect on *MR* with different thicknesses of organic layer [7]. Their results show that, for the device with the organic layer thickness of 200 nm, the value of *MR* decreases with the voltage. While for 100 nm thickness device, the value of *MR* initially decreases and then increases. Obviously, the relation between *MR* and voltage is also thickness-dependent.

All these investigations show that the voltage effect on *MR* is diverse and is dependent on extrinsic parameters, such as temperature and organic layer thickness. In this paper, we try to give a theoretical understanding on the voltage effect on *MR*. We consider an actual organic device and propose the voltage distribution along the whole device. The model is presented in the following section. The main results and discussions are given in Section 3 and finally a summary is concluded in Section 4.

2. Model and method

As we all know, due to the strong electron–phonon interaction, an injected electron (or hole) is trapped by a molecule to form a polaron. Driving by an external electric field, the polaron moves from one molecule to another. It transports through the whole organic layer and acts as carrier. The current density is given by $j = en\mu E$, where e denotes the electronic charge, n the carrier density, μ the carrier mobility and E the driving electric field. *MR* is defined as the change of the current density j caused by the applied magnetic field B,

$$MR = \frac{j(0) - j(B)}{j(B)}. \tag{1a}$$

For a certain value of the driving electric field E, *MR* is related to two factors: the carrier density n and the carrier mobility μ, as is described by

$$MR \approx MR(n) + MR(\mu). \tag{1b}$$

In the bipolar organic semiconductor devices, it is difficult to determine whether the magnetic field affects the carrier density or the carrier mobility. With this consideration, Veeraraghavan et al. performed a *MR* measurement in the unipolar hole-only PFO device, and revealed that *MR* is most likely related to the magnetic field effect on the carrier mobility μ but not the carrier density n [1]. Therefore, in this paper we consider that the carrier mobility variation is mainly responsible to *MR*, then it is given by

$$MR = \frac{\mu(0) - \mu(B)}{\mu(B)}. \tag{1c}$$

To obtain the carrier mobility in the organic layer, we consider a molecule chain along the electric field direction. For the band-like transport, the Hamiltonian reads

$$H = H_0 + H_E + H_B. \tag{2}$$

The first part of Eq. (2) is the one-dimensional tight-binding model [18],

$$H_0 = -\sum_{j,s}[\tau - \alpha(u_{j+1} - u_j) - (-1)^j\tau_e](C^+_{j+1,s}C_{j,s} + C^+_{j,s}C_{j+1,s}) + \sum_j \tfrac{1}{2}M\dot{u}_j^2 + \sum_j \tfrac{1}{2}K(u_{j+1} - u_j)^2, \tag{3}$$

where τ denotes the transfer integral between the neighbor molecules. α is the electron–phonon coupling constant, u_j the displacement of jth molecule from its equilibrium position, and τ_e the symmetry breaking term. $C^+_{j,s}(C_{j,s})$ is the creation (annihilation) operator of an electron at jth molecule with spin s. M is the mass of the small molecule and K the elastic constant.

The second part of Eq. (2) corresponds to the effect of an external electric field applied on the molecule chain, so as to drive the movement of the polaron,

$$H_E = |e|E\sum_{j,s}(ja + u_j)\left(C^+_{j,s}C_{j,s} - \frac{1}{2}\right), \tag{4}$$

where a is the lattice constant.

The third part of Eq. (2) denotes the effect of the magnetic field, which includes the external magnetic field $\vec{B}$ and the effective hyperfine field $\vec{B}_{hyp,j}$ of the hydrogen nuclei at jth molecule [11,19,20]

$$H_B = g\mu_B\sum_j(\vec{B} + \vec{B}_{hyp,j})\cdot\hat{\vec{S}}_j. \tag{5}$$

Here, g is the Lande factor, μ_B the Bohr magneton, and $\hat{\vec{S}}_j$ the operator at jth molecule. Supposing that $\vec{B}$ is along the z direction, we rewrite Eq. (5) as

$$H_B = \frac{1}{2}g\mu_B\sum_j(B + B_{hyp}\cos\theta_j)(C^+_{j,\uparrow}C_{j,\uparrow} - C^+_{j,\downarrow}C_{j,\downarrow}). \tag{5'}$$

Here, θ_j denotes the angle between $\vec{B}_{hyp,j}$ and z direction, which is random and is governed by the Boltzmann