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# The effect of interchain coupling on energy bands and ferromagnetic order in quasi-one-dimensional organic polymer ferromagnets

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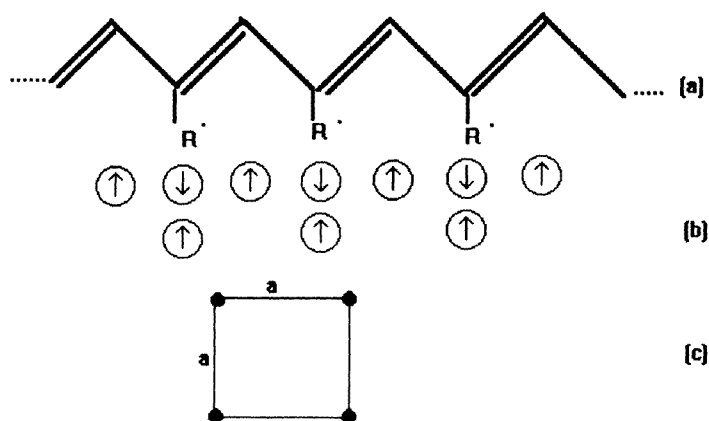
**Abstract.** Considering the interchain interaction as an interchain electron transfer, a theoretical model is developed to describe quasi-one-dimensional organic ferromagnets. We also take into account the interaction between the itinerant  $\pi$  electrons and the distortion of the lattice, the Hubbard electron–electron repulsion, and the antiferromagnetic correlation between the  $\pi$  electrons and the nonbonding electrons at the site radicals. By employing the mean-field approximation, four energy bands which exhibit a three-dimensional anisotropic aspect are obtained. When the interchain coupling increases, the energy gap decreases. In the case of small dimerization order parameter, due to the interchain coupling, the band gap may disappear and the conjugated  $\pi$  electrons in the main chain show ferromagnetic order in the ground state. The interchain coupling destabilizes the ferromagnetic ground state of the system.

## 1. Introduction

Recently, there has been great interest in exploring organic ferromagnets, such as poly-BIPO [1], *m*-PDPC [2] and *p*-NPNN [3]. However, only a little is known about ferromagnetism in organic materials. As is well known, the magnetism in transition metals originates from the strong correlation between the itinerant or localized d or f electrons and s electrons. In organic polymer molecular ferromagnets there are no magnetic ions as in common ferromagnetic materials (such as iron and nickel). The search for the origin of ferromagnetism in organic ferromagnets has become a challenge that has attracted considerable attention.

Ovchinnikov and Spector [4] proposed a simplified structure schematically shown in figure 1(a), which contains the structural features of a quasi-one-dimensional organic polymer ferromagnet. The main chain consists of carbon atoms with  $\pi$  electrons and R is a kind of side radical containing an unpaired electron. By treating the  $\pi$  electrons along the main carbon chain as an antiferromagnetic spin chain, they assumed that there are antiferromagnetic interactions between the  $\pi$ -electron spin and the residual spin of radical R. Then all of the residual spins at the side radicals can form a ferromagnetic spin chain as shown in figure 1(b). Recently, Fang *et al* [5, 6] proposed a theoretical model to describe the quasi-one-dimensional organic ferromagnet. They considered the itineracy of  $\pi$  electrons

and the strong electron–electron correlation in a one-dimensional system. Within the mean-field theory, they calculated the energy levels which split off with respect to different spins. Owing to the Peierls distortion of the lattice, there exists an energy gap at the Fermi level. In the ferromagnetic ground state, there is the same number of  $\pi$  electrons with up spin and down spin. The  $\pi$  electrons show no ferromagnetic order. However, in previous work, the system was described in terms of the behaviour of isolated chains. Using the SSH result [7], a typical intrachain hopping energy is 2–3 eV, while the interchain hopping is an order of magnitude smaller. Though, at first sight, a single-chain description would appear to be a reasonable approximation, there are certain situations in which interchain coupling can have a significant effect as will be demonstrated in this paper. It is shown in this paper that, due to the interchain interaction between two neighbouring chains in the quasi-one-dimensional organic ferromagnet, the electrons energy spectra exhibit a three-dimensional aspect which is obviously anisotropic, and the band gap is affected dramatically by the competition between the dimerization along the main chain and the interchain coupling. The ferromagnetic order and the state which is occupied by  $\pi$  electrons with different spins are different from those of [5] and [6].



**Figure 1.** (a) A chain structure of an organic polymer ferromagnet; (b) the arrangement of spin; and (c) the projection of the simplified structure proposed by us on the  $x$ – $y$  plane.

In this paper, to describe the organic polymer ferromagnet, we propose a structure with interchain coupling shown in figure 1. Figure 1(a) is the simplified chain structure along the  $z$  axis proposed by Ovchinnikov and Spector. Figure 1(c) is the projection of the structure on the  $x$ – $y$  plane, where the black dots forming a square lattice denote chains along the  $z$  axis. The interchain interaction is considered as an interchain electron transfer between the  $\pi$  electrons of nearest chains. We also take into account the itinerant  $\pi$  electrons, the Peierls distortion of the lattice, the antiferromagnetic spin correlations between the  $\pi$  electrons and the unpaired electrons at radical R and the Hubbard electron–electron interactions. By employing the mean-field approximation, we diagonalize the Hamiltonian in the Bloch representation. In section 2, we give the theoretical model and the formalisms of the energy spectra. In section 3, the results and discussion using the formalisms will be presented.

## 2. The Hamiltonians and the formalisms of the energy spectra

On the basis of the consideration in section 1, a Hamiltonian different from that of Fang *et al* [5] can be written as follows:

$$H = H_1 + H_2 + H_3 \quad (1)$$

$$H_1 = \sum_{l\sigma} (t_{l+1,l} c_{l+1,\sigma}^+ c_{l,\sigma} + \text{HC}) + \frac{1}{2} \sum_l \kappa (u_{l+1} - u_l)^2 \quad (2)$$

$$H_2 = V \sum_l n_{l\alpha} n_{l\beta} \quad (3)$$

$$H_3 = J \sum_l \delta_l \mathbf{S}_{lR} \cdot \mathbf{S}_l. \quad (4)$$

The first term  $H_1$  (2) describes the  $\pi$ -electron hopping, the electron–phonon interaction and the distortion of the lattice. Where  $l$  denotes the  $l$ th lattice site in the simplified structure with interchain coupling,  $c_{l\sigma}^+$  ( $c_{l\sigma}$ ) denotes the creation (annihilation) operator of a  $\pi$  electron with spin  $\sigma$  on the  $l$ th site,  $\kappa$  is the elastic constant of the lattice, we assume perfect dimerization on each chain and the displacement of carbon along the main chain is

$$u_l = (-1)^l u_0. \quad (5)$$

The hopping integral  $t_{l+1,l}$  of  $\pi$  electrons can be expanded for small displacement as

$$t_{l+1,l} = t_0 - \gamma (u_{l+1} - u_l) \quad \text{along the } z \text{ axis} \quad (6a)$$

$$t_{l+1,l} = \eta t_0 \quad \text{along the } x \text{ or } y \text{ axis} \quad (6b)$$

where  $t_0$  is the hopping integral when there is no dimerization,  $\gamma$  is the electron–phonon coupling constant along the  $z$  axis and  $\eta$  is the anisotropic parameter which is assumed to be much less than one.

The second term  $H_2$  in  $H$  (see (3)) describes the Hubbard repulsion between two  $\pi$  electrons when they are on the same carbon atom, and  $n_{l\sigma} = c_{l\sigma}^+ c_{l\sigma}$  ( $\sigma = \alpha, \beta$ ) where  $\alpha$  and  $\beta$  denote up spin and down spin respectively.

The third term  $H_3$  in  $H$  (see (4)) describes the antiferromagnetic correlations between the spin  $\mathbf{S}_l$  of  $\pi$  electrons and the residual spin  $\mathbf{S}_{lR}$  of R. We assume the coupling  $J > 0$  between the main chain and side radical. The radical R is connected to an even carbon atom, so

$$\begin{aligned} \delta_l &= 1 & l \text{ is even} \\ \delta_l &= 0 & l \text{ is odd.} \end{aligned} \quad (7)$$

The term  $\mathbf{S}_{lR} \cdot \mathbf{S}_l$  can be rewritten as

$$\mathbf{S}_{lR} \cdot \mathbf{S}_l = S_{lR}^z S_l^z + \frac{1}{2} [S_{lR}^+ S_l^- + S_{lR}^- S_l^+] \quad (8)$$

where  $S_l^z$  and  $S_l^+$  denote the Pauli spin matrix [8]

$$\begin{aligned} S_l^z &= \frac{1}{2} (n_{l\alpha} - n_{l\beta}) \\ S_l^+ &= c_{l\alpha}^+ c_{l\beta}. \end{aligned} \quad (9)$$

Then we use the mean-field approximation to divide  $n_{l\sigma}$  and  $S_{lR}^z$  as follows [8, 5]:

$$n_{l\sigma} = \langle n_{l\sigma} \rangle + \Delta n_{l\sigma} \quad (10a)$$

$$S_{lR}^z = \langle S_{lR}^z \rangle + \Delta S_{lR}^z. \quad (10b)$$

Here  $\langle \dots \rangle = \langle G | \dots | G \rangle$  is the average with respect to the ground state  $|G\rangle$  and  $\Delta n_{l\sigma}$  and  $\Delta S_{lR}^z$  are fluctuations from the average values. Then (3) and (4) can be rewritten as follows:

$$H_2 = V \sum_l (\langle n_{l\alpha} \rangle n_{l\beta} + \langle n_{l\beta} \rangle n_{l\alpha} - \langle n_{l\alpha} \rangle \langle n_{l\beta} \rangle) \quad (11)$$

$$H_3 = \frac{J}{2} \sum_l \langle S_{lR}^z \rangle (n_{l\alpha} - n_{l\beta}) \delta_l. \quad (12)$$

Transforming  $c_{l\sigma}^+$  and  $c_{l\sigma}$  into the Fourier components with wave vector  $\mathbf{k}$ ,

$$c_{l\sigma}^+ = N^{-1/2} \sum_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{l}} c_{\mathbf{k}\sigma}^+ \quad (13)$$

$$c_{l\sigma} = N^{-1/2} \sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{l}} c_{\mathbf{k}\sigma} \quad (14)$$

where  $N$  is the number of the lattice sites,  $l$  is the vector of the  $l$ th lattice site. Then, in (1), the Hamiltonian associated with the electron can be written as

$$H' = \sum_{\mathbf{k}\sigma} \mathbf{a}_{\mathbf{k}\sigma}^+ \mathbf{M}_\sigma(\mathbf{k}) \mathbf{a}_{\mathbf{k}\sigma}. \quad (15)$$

Here  $\mathbf{a}_{\mathbf{k}\sigma}^+$  is a vector defined as

$$\mathbf{a}_{\mathbf{k}\sigma}^+ = (a_{1\mathbf{k}\sigma}^+, a_{2\mathbf{k}\sigma}^+) \quad (16a)$$

$$a_{1\mathbf{k}\sigma}^+ = c_{\mathbf{k}\sigma}^+ \quad (16b)$$

$$a_{2\mathbf{k}\sigma}^+ = c_{\mathbf{k}'\sigma}^+$$

for  $k'_x = k_x$ ,  $k'_y = k_y$ ,  $k'_z = k_z + \pi/d$ ,  $\mathbf{M}_\sigma(\mathbf{k})$  is a  $2 \times 2$  energy matrix defined as

$$\mathbf{M}_\sigma(\mathbf{k}) = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} \quad (17)$$

$$M_{11} = -2t_0 \cos k_z d - 2\eta t_0 (\cos k_x a + \cos k_y a) + V \langle n_{\bar{\sigma}} \rangle + (J/4) \langle S_{lR}^z \rangle (\delta_{\sigma\alpha} - \delta_{\sigma\beta})$$

$$M_{12} = M_{21}^* = 4\gamma u_0 i \sin k_z d$$

$$M_{22} = 2t_0 \cos k_z d - 2\eta t_0 (\cos k_x a + \cos k_y a) + V \langle n_{\bar{\sigma}} \rangle + (J/4) \langle S_{lR}^z \rangle (\delta_{\sigma\alpha} - \delta_{\sigma\beta}).$$

Here the constant term  $\langle n_{l\alpha} \rangle \langle n_{l\beta} \rangle$  in (11) is omitted;  $d$  and  $a$  are lattice constants along the chain and in the directions perpendicular to the chain respectively.

It is convenient to cast all quantities into dimensionless forms as:

$$\begin{aligned} h' &= H'/t_0 & v &= V/t_0 & j &= J/t_0 \\ \lambda &= (2\gamma/t_0)u_0 & \mathbf{m}_\sigma(\mathbf{k}) &= \mathbf{M}_\sigma(\mathbf{k})/t_0. \end{aligned} \quad (18)$$

Then we can diagonalize the Hamiltonian  $h'$  as follows:

$$h' = \sum_{i\mathbf{k}\sigma} [E_{i\sigma}(\mathbf{k}) b_{i\mathbf{k}\sigma}^+ b_{i\mathbf{k}\sigma}] \quad (19)$$

where  $b_{i\mathbf{k}\sigma}^+$  is a new operator defined as

$$b_{i\mathbf{k}\sigma}^+ = \sum_j V_{ij\sigma}(\mathbf{k}) a_{j\mathbf{k}\sigma}^+ \quad (i, j = 1, 2). \quad (20)$$

Here  $V_{ij\sigma}(\mathbf{k})$  is defined by the eigenvector  $\mathbf{V}_{i\sigma}(\mathbf{k})$  of the matrix  $\mathbf{m}_\sigma(\mathbf{k})$ :

$$\mathbf{V}_{i\sigma}(\mathbf{k}) = (V_{i1\sigma}^*(\mathbf{k}), V_{i2\sigma}^*(\mathbf{k})) \quad (i = 1, 2) \quad (21)$$

$$\mathbf{m}_\sigma(\mathbf{k}) \mathbf{V}_{i\sigma}(\mathbf{k}) = E_{i\sigma}(\mathbf{k}) \mathbf{V}_{i\sigma}(\mathbf{k}). \quad (22)$$

The ground state can be written as

$$|G\rangle = \prod_{k\sigma} \prod_{i=1}^{occ} b_{ik\sigma}^+ |0\rangle. \quad (23)$$

Here  $|0\rangle$  is the true electron vacuum state and *occ* means those states occupied by electrons, and the eigenvalues  $E_{i\sigma}(\mathbf{k})$  ( $i = 1, 2$ ) of  $\mathbf{m}_\sigma(\mathbf{k})$  give the energy bands of the electron:

$$E_{1\sigma}(\mathbf{k}) = -A + B_\sigma \quad (24)$$

$$E_{2\sigma}(\mathbf{k}) = A + B_\sigma \quad (25)$$

where

$$A = [(2 \cos k_z d)^2 + 4\lambda^2 (\sin k_x d)^2]^{1/2} \quad (26)$$

$$B_\sigma = 2\eta(\cos k_x a + \cos k_y a) + v\langle n_{\bar{\sigma}} \rangle + \frac{1}{4}j\langle S_{iR}^z \rangle(\delta_{\sigma\alpha} - \delta_{\sigma\beta}) \quad (27)$$

for  $-\pi/a < k_x, k_y \leq \pi/a$  and  $-\pi/2d < k_z \leq \pi/2d$ , and

$$\langle n_\sigma \rangle = \langle G | n_{l\sigma} | G \rangle = \frac{1}{N} \sum_{\mathbf{k}} \sum_{i=1}^{occ} \sum_{j=1}^2 V_{ij\sigma}^*(\mathbf{k}) V_{ij\sigma}(\mathbf{k}). \quad (28)$$

Hence  $\langle n_\sigma \rangle$  must be solved from (28) and (22) self-consistently.

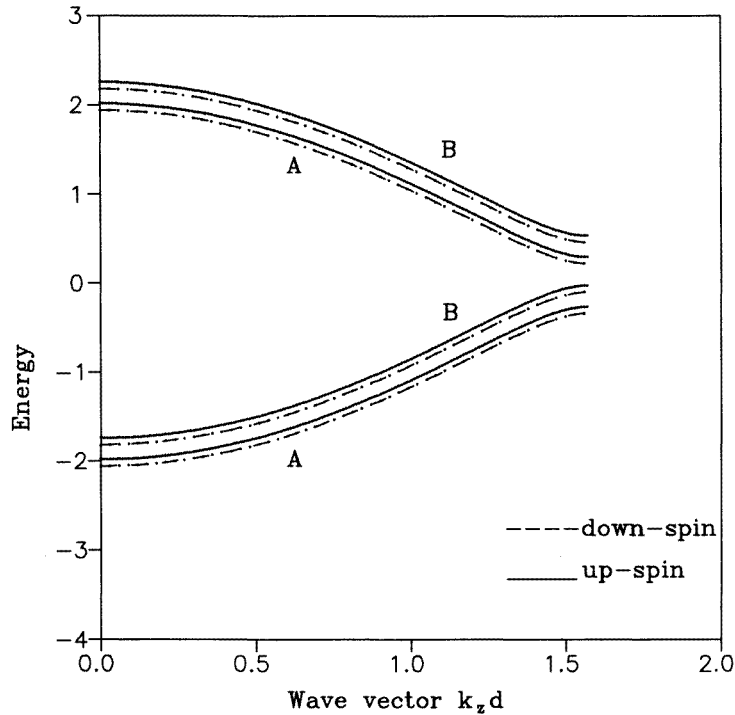
### 3. Results and discussion

From (24) and (25), we can see the electronic energy spectra contains four energy bands (figure 2). The dimerization results in an energy gap between the valence band and conduction band which split with respect to the up spin and down spin. The energy splitting originates from the antiferromagnetic correlation between the  $\pi$  electrons along the main chain and the unpaired electrons at the side radicals. To calculate the energy spectra numerically, we must determine the parameters  $\lambda$ ,  $v$ ,  $\eta$ ,  $j$ ,  $\langle n_\sigma \rangle$  and  $\langle S_{iR}^z \rangle$ . Although there are no experimental data on the dimerization and the interchain interaction, from figure 1 we can see that the main chain of this kind of simplified structure of the organic polymer ferromagnet is the same as that of polyacetylene, so we can estimate the dimerization order parameter  $\lambda$  (18) and the interchain interaction as those [7] of polyacetylene:  $\lambda = 0.144$ ,  $\eta = 0-0.1$ . Since there is only one  $\pi$  electron at a lattice site along the main chain, two lower bands must be filled by electrons while two higher bands are empty. In this case, from (28) we can obtain

$$\langle n_\alpha \rangle = \langle n_\beta \rangle = \frac{1}{2}. \quad (29)$$

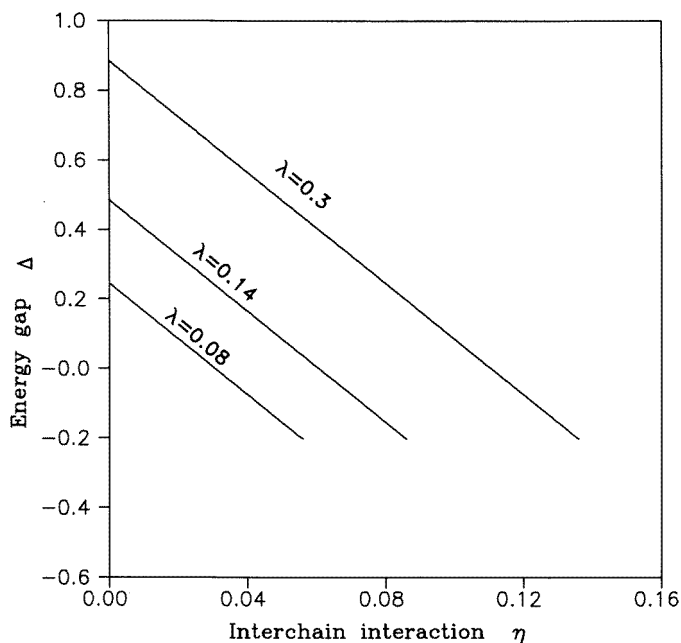
From the result of [5], we determine  $\langle S_{iR}^z \rangle = 1/2$  since all the residual spins of side radical R point up. We assume  $v = 0.2$ ,  $j = 0.3$  and  $\eta = 0.03$ . With the above parameters, we calculate the energy spectra along the  $k_z$  axis shown in figure 2. Curves A and B correspond to  $k_x a = k_y a = 0$  and  $k_x a = k_y a = \pi$  respectively. We can see clearly that the energy spectra is anisotropic in three-dimensional wave vector space. The Fermi surface has a complicated shape. The energy band along the  $k_z$  axis for  $k_x a = k_y a = 0$  (curve A) is lower than that for  $k_x a = k_y a = \pi$  (curve B). this difference results from the interchain electron transfer between  $\pi$  electrons of nearest chains. From (24) and (25), the minimum gap between the valence and conduction bands is the difference between the energy of the conduction band with down spin at  $k_x a = k_y a = 0$ ,  $k_z d = \pi/2$  and the energy of the valence band with up spin at  $k_x a = k_y a = \pi$ ,  $k_z d = \pi/2$  (see figure 2):

$$\Delta = 4\lambda - 8\eta - \frac{1}{2}j\langle S_{iR}^z \rangle - v(\langle n_\beta \rangle - \langle n_\alpha \rangle). \quad (30)$$



**Figure 2.** The electronic energy spectra along the  $k_z$  axis. Curves A and B correspond to  $k_x a = k_y a = 0$  and  $\pi$  respectively.

From (30), we can see clearly that the energy gap becomes small while the interchain coupling  $\eta$  increases, and for a definite  $\lambda$  there is a critical  $\eta = \eta_c$  at which the gap disappears. The smaller the dimerization  $\lambda$  is, the smaller the critical interchain interaction will be. This shows that the competition between the interchain interaction and the dimerization along the chain enables the band structures to exhibit a quite different feature from that of [5]. When the interchain interaction  $\eta$  is smaller than  $\eta_c$ , the gap exists everywhere. In this case, because the energy splitting with respect to different spin is smaller than the gap, all  $\pi$  electrons fill the valence band, so, in the ground state, half of the valence band is occupied by the up-spin electrons, while the other half is occupied by down-spin electrons (see (29)). There is no net spin density along the main chain. The ground state of the system is a stable ferromagnetic state, since all of the  $\pi$  electrons of the main chain exhibit nonmagnetism and all the residual spins of the radical point up. However, when the interchain interaction is strong enough ( $\eta > \eta_c$ ), due to the anisotropic feature of the energy spectra (shown in figure 2 and equations (24)–(27)), the energy gap  $\Delta < 0$  is satisfied and the Fermi surface has only partial gaps, hence the conduction and valence bands are connected. Figure 3 shows the energy gap  $\Delta$  versus interchain coupling  $\eta$  for different dimerization order parameters  $\lambda$ . From figure 3, we can see that for  $\lambda = 0.14$ ,  $v = 0.2$ ,  $j = 0.3$ ,  $\Delta < 0$  is satisfied at  $\eta_c = 0.06$ , which is indeed small, and the gap disappears. In this case, because the down-spin energy band is lower than the up-spin energy band (see figure 2), there are more electrons occupying the down-spin band than the up-spin band, so, in the ground state, there is spontaneous magnetization along the main chain. The



**Figure 3.** The energy gap  $\Delta$  versus interchain coupling  $\eta$  for  $j = 0.3$ ,  $v = 0.2$ ,  $\langle S_{lR}^z \rangle = 1/2$  and different values of  $\lambda$ .

$\pi$  electrons along the main chain form a weak ferromagnetic order. Unfortunately, because there are residual down spins along the main chain due to the interchain coupling while the residual spins of radicals point up, the sum of the net up spins of the system is decreased. This shows that the interchain coupling  $\eta > \eta_c$  will destabilize the ferromagnetic ground state of the system.

From the above discussion, it is obvious that the energy gap  $\Delta < 0$  is the condition under which the ferromagnetic order along the main chain appears. Using this criterion, we can estimate in which regime of the parameters  $v$ ,  $\eta$ ,  $\lambda$ ,  $j$  the ferromagnetic order of the main chain appears. We discuss the situation in which the absolute value of the energy gap  $|\Delta|$  is near zero. In this case, (29) is approximately satisfied, and the last term in (30) can be neglected. From (30), we can find that for  $j > 16\lambda - 32\eta$ , the energy gap  $\Delta < 0$  is satisfied. We must notice that only for  $j > 0$  does the splitting of the energy band with respect to spin happen. Therefore,  $j > 16\lambda - 32\eta > 0$  is the essential condition leading to the ferromagnetic order along the main chain. It should be noticed that when the on-site interaction  $v$  and the interchain hopping  $\eta$  are large with respect to the exchange interaction  $j$  between spin of the  $\pi$  electron and the spin of the side radical, the assumption that the spin density  $\langle n_{l\sigma} \rangle$  in (28) is independent of the site  $l$  in the main chain is inadequate. Fang *et al* [9] point out that for electron-phonon coupling constant 0.3 corresponding to the dimerization parameter  $\lambda = 0.14$  in our paper, when the on-site interaction  $v > 2$  is satisfied, all of the  $\pi$  electrons of the main chain form an antiferromagnetic spin density wave. Hence, the on-site interchain  $v < 2$  is another condition resulting in the ferromagnetic order along the main chain. In our paper, for the small dimerization  $\lambda = 0.14$ , which is a typical value of quasi-one-dimensional organic polymers (see [7]), the condition  $j > 16\lambda - 32\eta > 0$  leads



to  $\eta < 0.07$ , which is indeed small. Apparently, the parameters for figures 2 and 3 can satisfy the above conditions.

In conclusion, considering the interchain interaction, we develop a theoretical model to deal with the quasi-one-dimensional organic polymer ferromagnet. The results show that the energy spectra contain four energy bands which exhibit an obviously anisotropic feature in wave vector space. When the interchain interaction increases the energy gap becomes narrower. In the case of small dimerized distortion, the interchain electron transfer makes the energy gap disappear. The conjugated  $\pi$  electrons themselves show ferromagnetic order, but the net spin density of  $\pi$  electrons is negative while the spin density of the side radical is positive, so the interchain coupling destabilizes the ferromagnetic ground state of the system. We believe this is a possible reason why the synthesis of organic materials is so difficult that only a few products show ferromagnetism. In this paper we just consider one kind of interchain interaction. In our future study we will discuss which interchain interaction is favourable to stabilize the ferromagnetic ground state.

### Acknowledgment

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