Eur. Phys. J. B **14**, 411–417 (2000)

THE EUROPEAN PHYSICAL JOURNAL B

EDP Sciences

Random phase approximation of the spin-wave excitation for quasi-one-dimensional magnetic polymer

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Received 9 July 1999

Abstract. Based on a theoretical model proposed for quasi-one-dimensional organic polymer ferromagnets, the ground state and low-lying magnetic excitation are studied. Within Hartree-Fock approximation, the ground state of the system is shown to be a stable ferromagnetic state due to the electron-electron correlation and topological structure of the system. The random-phase approximation is employed to explore the magnon excitation and the excitation spectrum is obtained, including an acoustic mode and four optical modes. It is found that the acoustic mode possesses the characteristic of the ferromagnetic magnon.

PACS. 75.30.Ds Spin waves – 71.20.Rv Polymers and organic compounds – 75.50.Dd Nonmetallic ferromagnetic materials

1 Introduction

In recent years, there has been great interest in exploring organic ferromagnets. Several organic ferromagnets, such as poly-BIPO [1], *m*-PDPC [2] and *p*-NPNN [3], have been successfully synthesized. However, only a little has been known about the mechanism of the magnetically ordered state in organic materials. As is well-known, the magnetism in the transition metal compounds mainly originates from the strong interactions between the itinerant or localized d electrons and the itinerant s electrons. In organic polymer molecular ferromagnets there are no magnetic ions as in common ferromagnetic materials. 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 1a. The main chain consists of carbon atoms each with a π electron and R is a kind of side radicals containing an unpaired electron. They treated the π electrons along the main carbon chain as an antiferromagnetic spin chain, and assumed that there are antiferromagnetic interactions between the π-electron spin and the residual spin of radical R. If the radicals are attached to the main carbon chain in such a definite way, all of the residual spins at the side radicals will tend to be in the same direction as shown in Figure 1b and the ferromagnetic order is maintained. Recently, Fang, Liu, and Yao [5,6] proposed a theoretical model to describe this kind of quasi-one-dimensional organic ferromagnets. Within the mean-field theory, the ground state of the system described by the theoretical model was studied. Upon that, Wang *et al.* [7] investigated the spin-wave properties of the system. Their results actually show that the ground state of the system is a stable ferromagnetic state. In their model, the π electrons along the main carbon chain are considered to be itinerant while the unpaired electrons at the side radicals are regarded to have no freedom except their spins.

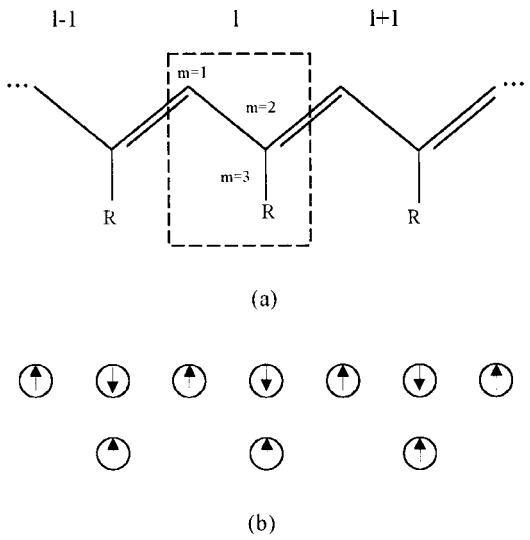

Fig. 1. (a) The simplified structure of a quasi-one-dimensional organic polymer organic ferromagnet and (b) the arrangement of spin.

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The main purpose of this paper is to study the magnon excitation in a quasi-one-dimensional π-conjugated organic ferromagnet based on its simplified structure as shown in Figure 1a. The system is described by a theoretical model in which the unpaired electrons at the side radicals are no longer regarded to be totally localized. In Section 2, we give the model Hamiltonian in which the Hubbard electron-electron repulsion and the hopping of the π electrons on the main chain as well as the unpaired electrons at the side radicals are taken into account. The ground state of the system is also discussed in Section 2. In Section 3, we study the magnon excitation of the system by random-phase approximation (RPA). The results are given in Section 4.

2 The model Hamiltonian and ground state

The model Hamiltonian employed in our study has the following form:

$$\begin{aligned}\hat{H} = & -T\sum_{l,\sigma}\left(\hat{a}^{+}_{l1\sigma}\hat{a}_{(l-1)2\sigma}+\hat{a}^{+}_{l1\sigma}\hat{a}_{l2\sigma}+\text{H.C.}\right)\\ & -T'\sum_{l,\sigma}\left(\hat{a}^{+}_{l2\sigma}\hat{a}_{l3\sigma}+\text{H.C.}\right)\\ & +\sum_{l}\left(U\hat{n}_{l1\alpha}\hat{n}_{l1\beta}+U\hat{n}_{l2\alpha}\hat{n}_{l2\beta}+U'\hat{n}_{l3\alpha}\hat{n}_{l3\beta}\right). \quad (1)\end{aligned}$$

Here, $\hat{a}^{+}_{lm\sigma}(\hat{a}_{lm\sigma})$ denotes the creation (annihilation) operator of an electron at a site specified by l, m and spin σ $(=\alpha,\beta)$ where α and β denote up-spin and down-spin, respectively. l $(=1,2,\ldots,N)$ labels the unit cell which contains two carbon sites of the main chain and one side radical, m $(=1,2)$ labels two carbon sites while m $(=3)$ labels the R side radical. T is the hopping integral between two neighboring π electrons along the main chain while T' is the hopping integral between a π electron on the main chain and an unpaired electron at the side radical. U and U' are the Hubbard effective electron-electron repulsive energy of π electrons on the main chain and unpaired electrons at the side radical, respectively, and $\hat{n}_{lm\sigma}=\hat{a}^{+}_{lm\sigma}\hat{a}_{lm\sigma}$.

It is convenient to use the following transformations:

$$\hat{h}=\frac{\hat{H}}{T},\quad t'=\frac{T'}{T},\quad u=\frac{U}{T},\quad u'=\frac{U'}{T}\cdot \quad (2)$$

Then, the Hamiltonian $\hat{H}$ becomes

$$\begin{aligned}\hat{h} = & -\sum_{l,\sigma}\left(\hat{a}^{+}_{l1\sigma}\hat{a}_{(l-1)2\sigma}+\hat{a}^{+}_{l1\sigma}\hat{a}_{l2\sigma}+\text{H.C.}\right)\\ & -t'\sum_{l,\sigma}\left(\hat{a}^{+}_{l2\sigma}\hat{a}_{l3\sigma}+\text{H.C.}\right)\\ & +\sum_{l}\left(u\hat{n}_{l1\alpha}\hat{n}_{l1\beta}+u\hat{n}_{l2\alpha}\hat{n}_{l2\beta}+u'\hat{n}_{l3\alpha}\hat{n}_{l3\beta}\right). \quad (3)\end{aligned}$$

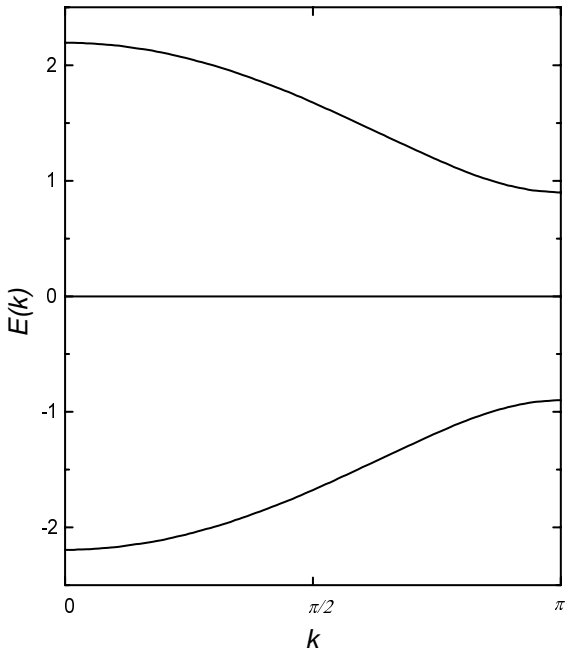

Fig. 2. The electronic energy bands with $t'=0.9$ and $u=u'=0$.

At first, we neglect the electron-electron correlation ($u=u'=0$) and consider only the hopping interactions of the electrons.

By introducing Fourier transformation, we can easily diagonalize the Hamiltonian $\hat{h}$, and get the electron energy spectrum, which contains three bands as shown in Figure 2. The lower band and the higher band, being symmetric with respect to the zero-energy level, mainly come from the π electrons on the main chain. The highly localized band with zero energy mainly comes from the unpaired electrons at the side radicals. Owing to $u=u'=0$, the energy bands are degenerate with respect to spin. So the ground state of the system is nonmagnetic.

Now, we consider the situation when $u\neq 0$ and $u'\neq 0$. In order to deal with the term of the Hubbard electron-electron correlation in Hamiltonian $\hat{h}$, we use Hartree-Fock approximation. By introducing Fourier transformation, within the mean-field theory, the energy spectrum can be obtained through the self-consistent iterative numerical calculation method. The corresponding results are shown in Figure 3 for $t'=0.9$ and $u=u'=1.0$.

As seen from Figure 3, the degeneracy of the energy bands with respect to spin is lifted due to the electron-electron correlation. The energy spectrum contains three up-spin and three down-spin energy bands. In the ground state, the lowest two up-spin energy bands and one down-spin energy band will be filled while the higher three energy bands will be empty. In consideration of the topological structure of the system, the ground state of the system is a high-spin ferromagnetic state.

Here, the energy gap between the middle localized up-spin and down-spin bands is a very important parameter. With decreasing and vanishing of the gap, the middle

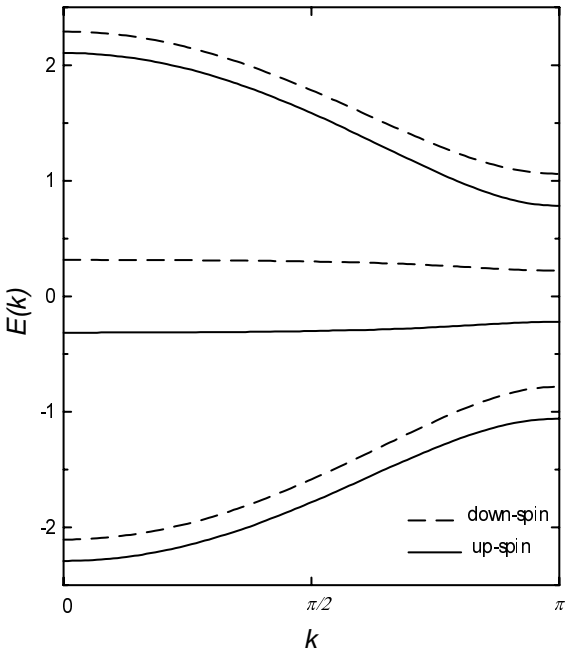

Fig. 3. The electronic energy bands with $t' = 0.9$ and $u = u' = 1.0$.

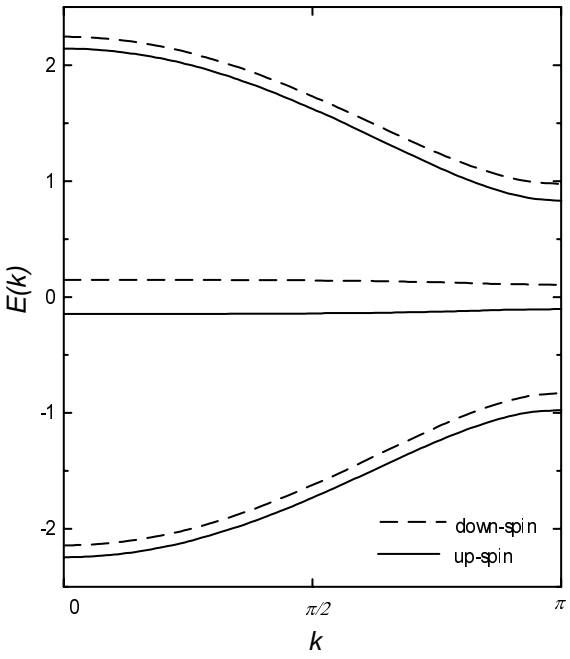

Fig. 4. The electronic energy bands with $t' = 0.9$ and $u = u' = 0.5$.

highly localized up-spin energy band and down-spin energy band in Figure 3 will have a tendency to overlap. In this case, the net spins of the system will decrease, and, as a result, the stability of the high-spin ground state will be spoiled. So the larger the gap is, the more stable the high-spin ground state is. Figure 4 shows the energy spectrum of the system when $t' = 0.9$ and $u = u' = 0.5$ as compared with Figure 3 when $t' = 0.9$ and $u = u' = 1.0$. We can see clearly that the strong electron-electron correlation can increase the energy gap and, consequently, will make the high-spin ferromagnetic ground state of the system more stable.

3 Magnon excitation

We now proceed to the problem of low-lying magnetic excitation above the mean-field theory ground state.

Transforming $\hat{a}^+_{lm\sigma}$ and $\hat{a}_{lm\sigma}$ into the Fourier components with wave vector k,

$$\hat{a}^+_{lm\sigma} = N^{-1/2} \sum_k \mathrm{e}^{-\mathrm{i}kl} \hat{a}^+_{km\sigma}, \tag{4a}$$

$$\hat{a}_{lm\sigma} = N^{-1/2} \sum_k \mathrm{e}^{\mathrm{i}kl} \hat{a}_{km\sigma}. \tag{4b}$$

Then Hamiltonian (Eq. (3)) can be written as

$$\hat{h} = \sum_{k,\sigma} \hat{a}^+_{k\sigma} M(k) \hat{a}_{k\sigma} + \frac{1}{N} \sum_{k,k'} \sum_q \sum_{m=1}^{3} [u + (u' - u)\delta_{m,3}] \times \hat{a}^+_{k+\frac{q}{2},m,\alpha} \hat{a}_{k-\frac{q}{2},m,\alpha} \hat{a}^+_{k'-\frac{q}{2},m,\beta} \hat{a}_{k'+\frac{q}{2},m,\beta}. \tag{5}$$

Here, $\hat{a}^+_{k\sigma}$ is defined as

$$\hat{a}^+_{k\sigma} = \left(\hat{a}^+_{k1\sigma}, \hat{a}^+_{k2\sigma}, \hat{a}^+_{k3\sigma}\right), \tag{6}$$

and $M(k)$ is defined as

$$M(k) = \begin{pmatrix} 0 & -t\left(1 + \mathrm{e}^{-\mathrm{i}k}\right) & 0 \\ -t\left(1 + \mathrm{e}^{\mathrm{i}k}\right) & 0 & -t' \\ 0 & -t' & 0 \end{pmatrix}. \tag{7}$$

From the equation

$$M(k) V_i(k) = E_i(k) V_i(k) \quad (i = 1, 2, 3), \tag{8}$$

we can get eigenvalue $E_i(k)$ $[E_1(k) > E_2(k) > E_3(k)]$ and eigenvector $V_i(k)$ of $M(k)$. $V_i^+(k)$ is a three-dimensional row vector:

$$V_i^+(k) = \left(V_{1i}^*(k), V_{2i}^*(k), V_{3i}^*(k)\right). \tag{9}$$

With the transformation

$$\hat{a}^+_{km\sigma} = \sum_{n=1}^{3} V^*_{mn}(k) \hat{c}^+_{kn\sigma}, \tag{10a}$$

$$\hat{a}_{km\sigma} = \sum_{n=1}^{3} V_{mn}(k) \hat{c}_{kn\sigma}, \tag{10b}$$

$$\hat{\gamma}^{+}_{\alpha q} = \sum_{k} A_1(k)\hat{c}^{+}_{k+\frac{q}{2},1,\beta}\hat{c}_{k-\frac{q}{2},2,\alpha} + \sum_{k} A_2(k)\hat{c}^{+}_{k+\frac{q}{2},1,\beta}\hat{c}_{k-\frac{q}{2},3,\alpha} + \sum_{k} A_3(k)\hat{c}^{+}_{k+\frac{q}{2},2,\beta}\hat{c}_{k-\frac{q}{2},2,\alpha} + \sum_{k} A_4(k)\hat{c}^{+}_{k+\frac{q}{2},2,\beta}\hat{c}_{k-\frac{q}{2},3,\alpha} + \sum_{k} A_5(k)\hat{c}^{+}_{k+\frac{q}{2},3,\beta}\hat{c}_{k-\frac{q}{2},1,\alpha}\,, \tag{12a}$$

$$\hat{\gamma}^{+}_{\beta q} = \sum_{k} A'_1(k)\hat{c}^{+}_{k-\frac{q}{2},2,\alpha}\hat{c}_{k+\frac{q}{2},1,\beta} + \sum_{k} A'_2(k)\hat{c}^{+}_{k-\frac{q}{2},3,\alpha}\hat{c}_{k+\frac{q}{2},1,\beta} + \sum_{k} A'_3(k)\hat{c}^{+}_{k-\frac{q}{2},2,\alpha}\hat{c}_{k+\frac{q}{2},2,\beta} + \sum_{k} A'_4(k)\hat{c}^{+}_{k-\frac{q}{2},3,\alpha}\hat{c}_{k+\frac{q}{2},2,\beta} + \sum_{k} A'_5(k)\hat{c}^{+}_{k-\frac{q}{2},1,\alpha}\hat{c}_{k+\frac{q}{2},3,\beta}\,. \tag{12b}$$

$$\begin{aligned} A_1(k) = {} & \left[\omega_\alpha(q) - E_1\left(k+\frac{q}{2}\right) + E_2\left(k-\frac{q}{2}\right)\right]^{-1}\Bigg[A_1(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]\Bigg\{V^*_{m1}\left(k+\frac{q}{2}\right)V_{m1}\left(k+\frac{q}{2}\right) \\ & \times\frac{1}{N}\sum_{k'}\left[V^*_{m2}(k')V_{m2}(k')+V^*_{m3}(k')V_{m3}(k')\right] - V^*_{m2}\left(k-\frac{q}{2}\right)V_{m2}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V^*_{m3}(k')V_{m3}(k')\Bigg\} \\ & - A_2(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V^*_{m3}\left(k-\frac{q}{2}\right)V_{m2}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V^*_{m3}(k')V_{m3}(k') \\ & + A_3(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V^*_{m1}\left(k+\frac{q}{2}\right)V_{m2}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\left\{V^*_{m2}(k')V_{m2}(k')+V^*_{m3}(k')V_{m3}(k')\right\} \\ & - \sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V^*_{m1}\left(k+\frac{q}{2}\right)V_{m2}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\Bigg\{A_1(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right) \\ & + A_2(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right) + A_3(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right) \\ & + A_4(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right) - A_5(k')V^*_{m1}\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)\Bigg\}\Bigg]\,. \end{aligned} \tag{14}$$

equation (5) can be written as follows:

$$\begin{aligned} \hat{h} = {} & \sum_{k,\sigma}\sum_{i=1}^{3}E_i(k)\hat{c}^{+}_{ki\sigma}\hat{c}_{ki\sigma} \\ & + \frac{1}{N}\sum_{k,k'}\sum_{q}\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right] \\ & \times\sum_{\substack{i_1,i_2,\\ i_3,i_4}}V^*_{mi_1}\left(k+\frac{q}{2}\right)V_{mi_2}\left(k-\frac{q}{2}\right) \\ & \times V^*_{mi_3}\left(k'-\frac{q}{2}\right)V_{mi_4}\left(k'+\frac{q}{2}\right) \\ & \times\hat{c}^{+}_{k+\frac{q}{2},i_1,\alpha}\hat{c}_{k-\frac{q}{2},i_2,\alpha}\hat{c}^{+}_{k'-\frac{q}{2},i_3,\beta}\hat{c}_{k'+\frac{q}{2},i_4,\beta}. \end{aligned} \tag{11}$$

As shown in Figures 3, 4, the ground state of the system turns to be a six-band case with Fermi surface in the gap. Thus, the magnon excitation with spin flip can be described in following forms [8]:

see equations (12a, 12b) above.

Here $A_i(k)$ and $A'_i(k)$ $(i = 1, 2, \ldots, 5)$ are coupling coefficients.

The equations of motion for magnon excitation are

$$\omega_\alpha(q)\hat{\gamma}^{+}_{\alpha q} = \left[\hat{h}, \hat{\gamma}^{+}_{\alpha q}\right], \tag{13a}$$

$$\omega_\beta(q)\hat{\gamma}^{+}_{\beta q} = \left[\hat{h}, \hat{\gamma}^{+}_{\beta q}\right], \tag{13b}$$

where $\omega_\alpha(q)$ and $\omega_\beta(q)$ denote magnon excitation energy for α branch and β branch, respectively. By introducing random-phase approximation (RPA) [9,10], we can obtain equations for $A_i(k)$ $(i = 1, 2, \ldots, 5)$ from equation (13a) as follows:

see equations (14) above and (15–18) next pages.

Here, wave vectors $|k|$, $|k'|$ and $|q|$ take their values from zero to π. Equations for $A'_i(k)$ $(i = 1, 2, \ldots, 5)$ have a similar form to equations (14–18). These equations can be solved numerically.

It can be seen from Figures 3, 4 that there are four possible spin flips from the lowest two occupied up-spin energy bands to the highest and the middle localized unoccupied down-spin energy bands, and, at the same time, there is one possible spin flip from the lowest occupied down-spin energy band to the highest unoccupied up-spin

$$A_2(k)=\left[\omega_\alpha(q)-E_1\left(k+\frac{q}{2}\right)+E_3\left(k-\frac{q}{2}\right)\right]^{-1}\left[A_2(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]\left\{V_{m1}^*\left(k+\frac{q}{2}\right)V_{m1}\left(k+\frac{q}{2}\right)\right.\right.$$
$$\times\frac{1}{N}\sum_{k'}\left[V_{m2}^*(k')V_{m2}(k')+V_{m3}^*(k')V_{m3}(k')\right]-V_{m3}^*\left(k-\frac{q}{2}\right)V_{m3}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V_{m3}^*(k')V_{m3}(k')\Bigg\}$$
$$-A_1(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m2}^*\left(k-\frac{q}{2}\right)V_{m3}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V_{m3}^*(k')V_{m3}(k')$$
$$+A_4(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m1}^*\left(k+\frac{q}{2}\right)V_{m2}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\left\{V_{m2}^*(k')V_{m2}(k')+V_{m3}^*(k')V_{m3}(k')\right\}$$
$$-\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m1}^*\left(k+\frac{q}{2}\right)V_{m3}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\Bigg\{A_1(k')V_{m2}^*\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)$$
$$+A_2(k')V_{m3}^*\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)+A_3(k')V_{m2}^*\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)$$
$$\left.\left.+A_4(k')V_{m3}^*\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)-A_5(k')V_{m1}^*\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)\Bigg\}\right],\tag{15}$$

$$A_3(k)=\left[\omega_\alpha(q)-E_2\left(k+\frac{q}{2}\right)+E_2\left(k-\frac{q}{2}\right)\right]^{-1}\left[A_3(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]\left\{V_{m2}^*\left(k+\frac{q}{2}\right)V_{m2}\left(k+\frac{q}{2}\right)\right.\right.$$
$$\times\frac{1}{N}\sum_{k'}\left[V_{m2}^*(k')V_{m2}(k')+V_{m3}^*(k')V_{m3}(k')\right]-V_{m2}^*\left(k-\frac{q}{2}\right)V_{m2}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V_{m3}^*(k')V_{m3}(k')\Bigg\}$$
$$+A_1(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m2}^*\left(k+\frac{q}{2}\right)V_{m1}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\left\{V_{m2}^*(k')V_{m2}(k')+V_{m3}^*(k')V_{m3}(k')\right\}$$
$$-A_4(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m3}^*\left(k-\frac{q}{2}\right)V_{m2}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V_{m3}^*(k')V_{m3}(k')$$
$$-\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m2}^*\left(k+\frac{q}{2}\right)V_{m2}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\Bigg\{A_1(k')V_{m2}^*\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)$$
$$+A_2(k')V_{m3}^*\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)+A_3(k')V_{m2}^*\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)$$
$$\left.\left.+A_4(k')V_{m3}^*\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)-A_5(k')V_{m1}^*\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)\Bigg\}\right],\tag{16}$$

$$A_4(k)=\left[\omega_\alpha(q)-E_2\left(k+\frac{q}{2}\right)+E_3\left(k-\frac{q}{2}\right)\right]^{-1}\left[A_4(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]\left\{V_{m2}^*\left(k+\frac{q}{2}\right)V_{m2}\left(k+\frac{q}{2}\right)\right.\right.$$
$$\times\frac{1}{N}\sum_{k'}\left[V_{m2}^*(k')V_{m2}(k')+V_{m3}^*(k')V_{m3}(k')\right]-V_{m3}^*\left(k-\frac{q}{2}\right)V_{m3}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V_{m3}^*(k')V_{m3}(k')\Bigg\}$$
$$+A_2(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m2}^*\left(k+\frac{q}{2}\right)V_{m1}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\left\{V_{m2}^*(k')V_{m2}(k')+V_{m3}^*(k')V_{m3}(k')\right\}$$
$$-A_3(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m2}^*\left(k-\frac{q}{2}\right)V_{m3}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V_{m3}^*(k')V_{m3}(k')$$
$$-\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V_{m2}^*\left(k+\frac{q}{2}\right)V_{m3}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\Bigg\{A_1(k')V_{m2}^*\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)$$
$$+A_2(k')V_{m3}^*\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)+A_3(k')V_{m2}^*\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)$$
$$\left.\left.+A_4(k')V_{m3}^*\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)-A_5(k')V_{m1}^*\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)\Bigg\}\right],\tag{17}$$

$$A_5(k) = \left[\omega_\alpha(q) + E_1\left(k+\frac{q}{2}\right) - E_3\left(k-\frac{q}{2}\right)\right]^{-1}\left[A_5(k)\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]\left\{V^*_{m3}\left(k+\frac{q}{2}\right)V_{m3}\left(k+\frac{q}{2}\right)\right.\right.$$
$$\times\frac{1}{N}\sum_{k'}\left[V^*_{m2}(k')V_{m2}(k')+V^*_{m3}(k')V_{m3}(k')\right]-V^*_{m1}\left(k-\frac{q}{2}\right)V_{m1}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}V^*_{m3}(k')V_{m3}(k')\Bigg\}$$
$$-\sum_{m=1}^{3}\left[u+(u'-u)\delta_{m,3}\right]V^*_{m3}\left(k+\frac{q}{2}\right)V_{m1}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\Bigg\{A_1(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)$$
$$+A_2(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)+A_3(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)$$
$$\left.\left.+A_4(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)-A_5(k')V^*_{m1}\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)\Bigg\}\right]\cdot \quad (18)$$

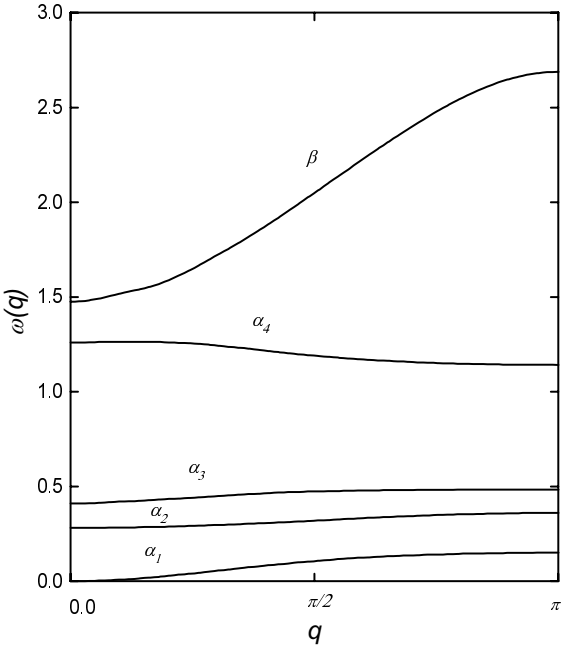

Fig. 5. The dispersion relation of magnon with $t' = 0.9$ and $u = u' = 1.0$.

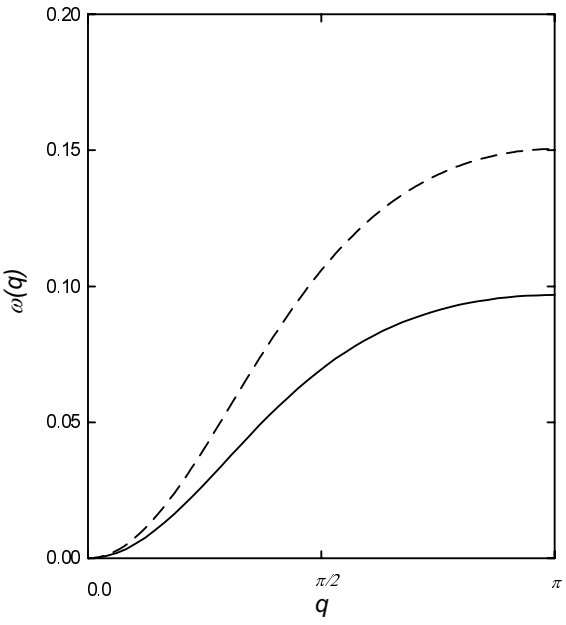

Fig. 6. The two acoustic modes with $t' = 0.9$, $u = u' = 0.5$ (solid line) and $t' = 0.9$, $u = u' = 1.0$ (dashed line) respectively.

energy band. So it can be expected that four α branches and one β branch, which have positive excitation energy, should exist in the combined magnon excitation spectrum. Figure 5 exhibits the dispersion relations of magnon excitation for different branches when $t' = 0.9$ and $u = u' = 1.0$. It is shown that the spectrum of magnon excitation, just as expected, contains five energy branches, one acoustic branch and four optical branches. The four lower energy branches of magnons $\alpha_{1,2,3,4}$ refers to $\hat{\gamma}^+_{\alpha q}$ while the highest one β refers to $\hat{\gamma}^+_{\beta q}$, and the $\hat{\gamma}^+_{\alpha q}$ and $\hat{\gamma}^+_{\beta q}$ correspond to the spin flip of up-spin and down-spin, respectively. As seen from Figure 5, the lowest branch (acoustic mode) of the magnon excitation spectrum shows a quadratic-like dispersion relation in the limit of the long wavelength, which means that the magnon excitation of the system possesses the characteristics of the ferromagnetic magnons.

Figure 6 shows two acoustic modes when $t' = 0.9$, $u = u' = 1.0$ and $t' = 0.9$, $u = u' = 0.5$, respectively. It can be seen that the acoustic mode is significantly influenced by the electron-electron correlation. The excitation becomes larger when u and u' are larger, and, as a result, the high-spin ferromagnetic ground state of the system will be more stable.

It is interesting to study the existence of the three-dimensional LRO in a system made of such chains as shown in Figure 1a. We believe that the three-dimensional LRO should exist because of the strong Hubberd on-site electron-electron repulsions of the π electrons on the main chain and the unpaired electrons at the side radicals that

are favorable to the ferromagnetic correlation in the system. This would be more practical than the quasi-one-dimensional case, though it is not studied by previous works in this field. In our future studies, we will address this problem.

4 Conclusions

We have studied the ferromagnetic properties of a quasi-one-dimensional π-conjugated organic ferromagnetic model based on the simplified structure as shown in Figure 1a. The electron-electron correlation causes the degeneracy of energy bands with respect to spin to be lifted, and hence the system has a stable high-spin ferromagnetic ground state.

The magnon excitation is studied in the random-phase approximation. The magnon excitation spectrum that contains an acoustic branch and four optical branches is obtained. The lowest acoustic branch has been found to possess the characteristics of the ferromagnetic magnon. The strong electron-electron correlation can make the high-spin ferromagnetic ground state of the system more stable.

The authors are grateful to Dr. Y.Q. Wang and Dr. W.Z. Wang for helpful discussions. This work was supported by the National Natural Science Foundation of China under the grant No. 19777101 and 19774023.

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