Effect of inter-site electron-electron interaction on the spin-wave excitation in quasi-one-dimensional organic magnetic polymer

2000 J. Phys.: Condens. Matter 12 5735

(http://iopscience.iop.org/0953-8984/12/26/319)

J. Phys.: Condens. Matter **12** (2000) 5735–5743. Printed in the UK PII: S0953-8984(00)09666-1

Effect of inter-site electron–electron interaction on the spin-wave excitation in quasi-one-dimensional organic magnetic polymer

L Zhao† and K L Yao†‡

† Department of Physics, Huazhong University of Science and Technology, Wuhan 430074, China
‡ International Centre of Material Physics, Academy of Science, Shenyang 110015, China, and CCAST (World Laboratory), PO Box 8730, Beijing 100080, China

Received 17 November 1999, in final form 10 May 2000

Abstract. Considering the inter-site electron–electron interaction, we investigate the ground state and spin-wave excitation of a theoretical model proposed for quasi-one-dimensional π-conjugated organic polymer ferromagnets. Within mean-field theory, the ground state of the system is shown to be a high-spin ferromagnetic state due to the topological structure of the system. By employing the random-phase approximation (RPA), the spin-wave excitation spectrum is obtained. It is found that the acoustic spin-wave mode displays the feature of the ferromagnetic magnon, and the inter-site electron–electron interaction has influence on it. With the increase of inter-site electron–electron interaction, the high-spin ferromagnetic ground state of the system will be unstable.

1. Introduction

In recent years, with the discovery of organic conductors and organic superconductors, there is great interest in the design and construction of organic magnetic materials [1–5]. Several research groups in the world have successfully synthesized organic ferromagnets, such as poly-BIPO [6], *m*-PDPC [7] and *p*-NPNN [8]. Since the known cases of ferromagnetism always involve spins of either d or f electrons, which play an essential role in the mechanism of the ferromagnetic interaction, the mechanism of the magnetically ordered state in organic materials containing only p electrons is, therefore, a great challenge that has attracted considerable attention.

McConnell [9] first proposed to produce intermolecular ferromagnetic interaction in organic molecules in 1963. Ovchinnikov and Spector [10] proposed that organic π-conjugated polymers with organic free radicals might exhibit ferromagnetic interaction, and suggested a simplified structure schematically shown in figure 1(a) to describe the ferromagnetic order in the organic ferromagnets. The main chain consists of carbon atoms each with a π electron and R is a kind of side radical 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 1(b) and the ferromagnetic order is maintained. Recently, Fang *et al* [11] proposed a theoretical model for the quasi-one-dimensional organic ferromagnet and obtained a ferromagnetic ground state. Upon that, Wang *et al* [12] investigated the spin-wave properties of the system. In their model,

0953-8984/00/265735+09$30.00

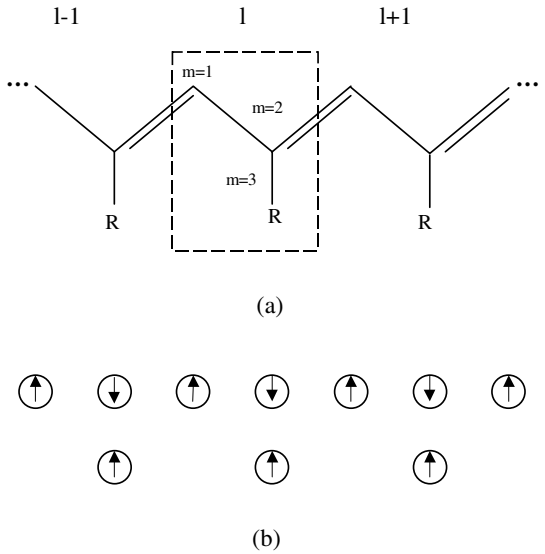

Figure 1. (a) The simplified structure of a quasi-one-dimensional organic polymer ferromagnet and (b) the arrangement of spin.

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.

The main purpose of this paper is to study the spin-wave excitation in a quasi-one-dimensional π-conjugated organic ferromagnet with inter-site electron–electron interaction based on its simplified structure as shown in figure 1(a). The system is described by a theoretical model in which the unpaired electrons at the side radicals are no longer regarded as being totally localized. In section 2, we give the model Hamiltonian in which the Hubbard on-site electron–electron repulsion, the inter-site electron–electron Coulomb 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 spin-wave excitation of the system by the random-phase approximation (RPA). The conclusions are given in section 4.

2. The model Hamiltonian and ground state

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

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

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.

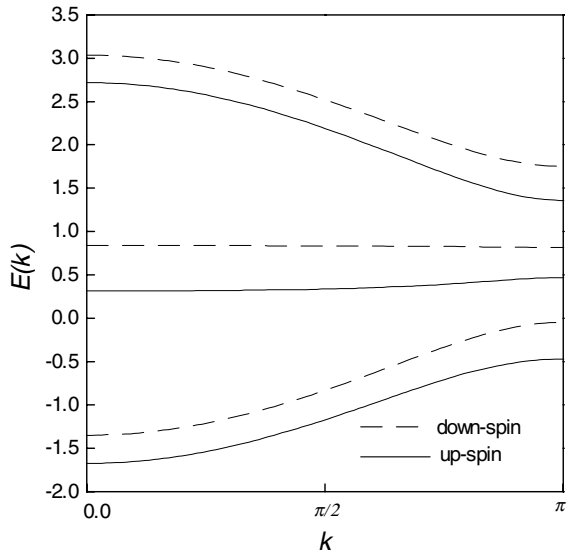

Figure 2. The electronic energy bands with $t' = 0.9$, $u = u' = 0.9$ and $v = v' = 0.2$.

T is the hopping integral between two neighbouring 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 on-site e–e repulsion of π electrons on the main chain and unpaired electrons at the side radical, respectively. V describes the inter-site e–e Coulomb repulsion of π electrons on the main chain while V' describes the inter-site e–e Coulomb repulsion between a π electron on the main chain and an unpaired electron at the side radical, and $\hat{n}_{lm\sigma} = \hat{a}^{+}_{lm\sigma}\hat{a}_{lm\sigma}$.

It is convenient to use the following transformations:

$$\hat{h} = \frac{\hat{H}}{T} \qquad t' = \frac{T'}{T} \qquad u = \frac{U}{T} \qquad u' = \frac{U'}{T} \qquad v = \frac{V}{T} \qquad v' = \frac{V'}{T}. \tag{2}$$

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

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

In order to deal with the term of the Hubbard on-site e–e repulsion and inter-site e–e Coulomb repulsion in Hamiltonian $\hat{h}$, we use the 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 2 for $t' = 0.9$, $u = u' = 0.9$ and $v = v' = 0.2$.

As seen from figure 2, 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.

3. Magnon excitation

We now turn to the problem of low-lying magnetic excitation.

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

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

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

Then the Hamiltonian (equation (3)) can be written as

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

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

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

and $M(k)$ is a 3×3 matrix.

From the equation

$$M(k)V_i(k) = E_i(k)V_i(k) \qquad (i=1,2,3) \tag{7}$$

we can obtain 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:

$$V_i^+(k) = (V^*_{1i}(k), V^*_{2i}(k), V^*_{3i}(k)). \tag{8}$$

With the transformation

$$\hat{a}^+_{km\sigma} = \sum_{n=1}^{3}V^*_{mn}(k)\hat{c}^+_{kn\sigma} \tag{9a}$$

$$\hat{a}_{km\sigma} = \sum_{n=1}^{3}V_{mn}(k)\hat{c}_{kn\sigma} \tag{9b}$$

equation (5) can be written as follows:

$$\begin{aligned}\hat{h} = \sum_{k,\sigma}\sum_{i=1}^{3}E_i(k)\hat{c}^+_{ki\sigma}\hat{c}_{ki\sigma} + \frac{1}{N}\sum_{k,k'}\sum_q\sum_{m=1}^{3}[u+(u'-u)\delta_{m,3}]\\ \times\sum_{\substack{i_1,i_2,\\ i_3,i_4}}V^*_{mi_1}\left(k+\frac{q}{2}\right)V_{mi_2}\left(k-\frac{q}{2}\right)V^*_{mi_3}\left(k'-\frac{q}{2}\right)V_{mi_4}\left(k'+\frac{q}{2}\right)\\ \times\hat{c}^+_{k+q/2,i_1,\alpha}\hat{c}_{k-q/2,i_2,\alpha}\hat{c}^+_{k'-q/2,i_3,\beta}\hat{c}_{k'+q/2,i_4,\beta}\\ +\frac{1}{N}\sum_{k,k'}\sum_q\sum_{\sigma,\sigma'}\sum_{m=1,3}[v+(v'-v)\delta_{m,3}][1+\exp(-\mathrm{i}q)\delta_{m,1}]\\ \times\sum_{\substack{i_1,i_2,\\ i_3,i_4}}V^*_{mi_1}\left(k+\frac{q}{2}\right)V_{mi_2}\left(k-\frac{q}{2}\right)V^*_{2i_3}\left(k'-\frac{q}{2}\right)V_{2i_4}\left(k'+\frac{q}{2}\right)\\ \times\hat{c}^+_{k+q/2,i_1,\sigma}\hat{c}_{k-q/2,i_2,\sigma}\hat{c}^+_{k'-q/2,i_3,\sigma'}\hat{c}_{k'+q/2,i_4,\sigma'}.\end{aligned} \tag{10}$$

As shown in figure 2, the ground state of the system turns to be a six-band case with Fermi surface in the gap. 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, in the meanwhile, there is one possible spin flip from the lowest occupied down-spin energy band to the highest unoccupied up-spin energy band. Thus, the spin-wave excitation with spin flip can be described in the following form [13]:

$$\hat{\gamma}^{+}_{\alpha q} = \sum_{k} A_1(k)\hat{c}^{+}_{k+q/2,1,\beta}\hat{c}_{k-q/2,2,\alpha} + \sum_{k} A_2(k)\hat{c}^{+}_{k+q/2,1,\beta}\hat{c}_{k-q/2,3,\alpha} + \sum_{k} A_3(k)\hat{c}^{+}_{k+q/2,2,\beta}\hat{c}_{k-q/2,2,\alpha} + \sum_{k} A_4(k)\hat{c}^{+}_{k+q/2,2,\beta}\hat{c}_{k-q/2,3,\alpha} + \sum_{k} A_5(k)\hat{c}^{+}_{k+q/2,3,\beta}\hat{c}_{k-q/2,1,\alpha} \tag{11a}$$

$$\hat{\gamma}^{+}_{\beta q} = \sum_{k} A'_1(k)\hat{c}^{+}_{k-q/2,2,\alpha}\hat{c}_{k+q/2,1,\beta} + \sum_{k} A'_2(k)\hat{c}^{+}_{k-q/2,3,\alpha}\hat{c}_{k+q/2,1,\beta} + \sum_{k} A'_3(k)\hat{c}^{+}_{k-q/2,2,\alpha}\hat{c}_{k+q/2,2,\beta} + \sum_{k} A'_4(k)\hat{c}^{+}_{k-q/2,3,\alpha}\hat{c}_{k+q/2,2,\beta} + \sum_{k} A'_5(k)\hat{c}^{+}_{k-q/2,1,\alpha}\hat{c}_{k+q/2,3,\beta}. \tag{11b}$$

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

The equations of motion for magnon excitation are

$$\omega_\alpha(q)\hat{\gamma}^{+}_{\alpha q} = [\hat{h}, \hat{\gamma}^{+}_{\alpha q}] \tag{12a}$$

$$\omega_\beta(q)\hat{\gamma}^{+}_{\beta q} = [\hat{h}, \hat{\gamma}^{+}_{\beta q}], \tag{12b}$$

where $\omega_\alpha(q)$ and $\omega_\beta(q)$ denote magnon excitation energy for the α branch and β branch, respectively. By introducing the random-phase approximation (RPA) [14, 15], we can obtain equations for $A_i(k)$ and $A'_i(k)$ $(i = 1, 2, \ldots, 5)$ (see the appendix) from equations (12). These equations can be solved numerically.

Figure 3 exhibits the dispersion relations of magnon excitation for different branches when $t' = 0.9$, $u = u' = 0.9$ and $v = v' = 0.2$. It is shown that the spectrum of magnon excitation contains five energy branches—one acoustic branch and four optical branches. The four lower energy branches of magnons $\alpha_{1,2,3,4}$ refer 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 3, the lowest branch (acoustic mode) of the magnon excitation spectrum shows a quadratic-like dispersion relation when q is small, which means that the magnon excitation of the system displays the feature of the ferromagnetic magnons.

Figures 4 and 5 show several acoustic modes when $t' = 0.9$, $u = u' = 0.9$ and $t' = 0.9$, $u = u' = 1.2$, respectively. Curves a, b, c, d and e correspond, respectively, to inter-site electron–electron Coulomb repulsion $v = v' = 0.0$, 0.2, 0.4, 0.6 and 0.8. It can be seen that the inter-site electron–electron Coulomb repulsion has an influence on the acoustic mode. The excitation becomes rather easy when the inter-site electron–electron Coulomb repulsions v and v' are large, which indicates that the ferromagnetic correlation in the system is weaker and, furthermore, is unfavourable to the stability of the ferromagnetic ground state. Comparing figure 4 with 5, one can see that the smaller the Hubbard on-site electron–electron repulsion is, the smaller the excitation becomes and, with increasing v and v', the quicker the decreasing of the excitation is. So, it is important to refrain from the strong inter-site electron–electron Coulomb repulsion and, meanwhile, to stimulate the strong Hubbard on-site electron–electron repulsion in order to preserve the high-spin ferromagnetic ground state of the system.

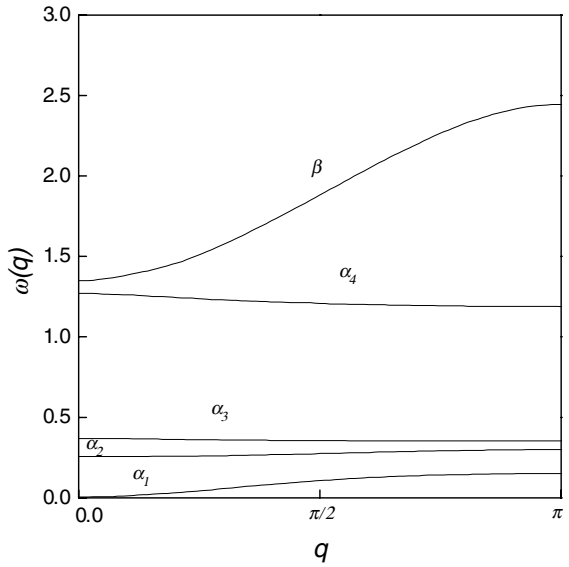

Figure 3. The dispersion relation of the magnon with $t' = 0.9$, $u = u' = 0.9$ and $v = v' = 0.2$.

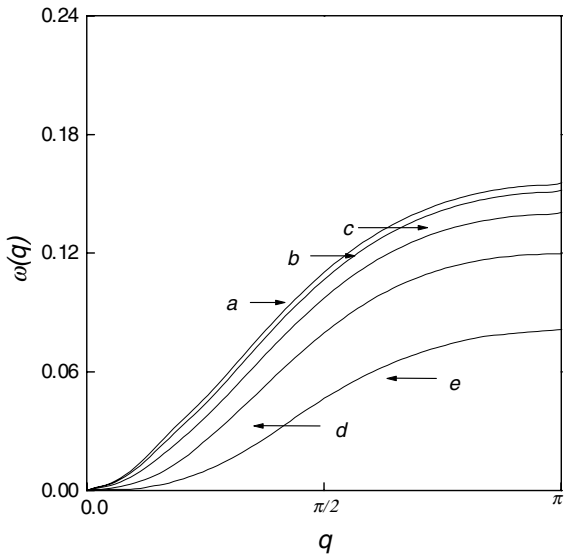

Figure 4. The acoustic modes for different inter-site e–e Coulomb repulsion: (a) $v = v' = 0.0$, (b) $v = v' = 0.2$, (c) $v = v' = 0.4$, (d) $v = v' = 0.6$ and (e) $v = v' = 0.8$ when $t' = 0.9$ and $u = u' = 0.9$.

In addition, when the inter-site Coulomb interaction v or v' is taken into account separately, our calculations show that the Coulomb interaction (v) between the nearest-neighbour π electrons on the main chain plays a more effective role in suppressing the ferromagnetic correlation than that (v') between an unpaired electron at the side radical and a π electron on the main chain does.

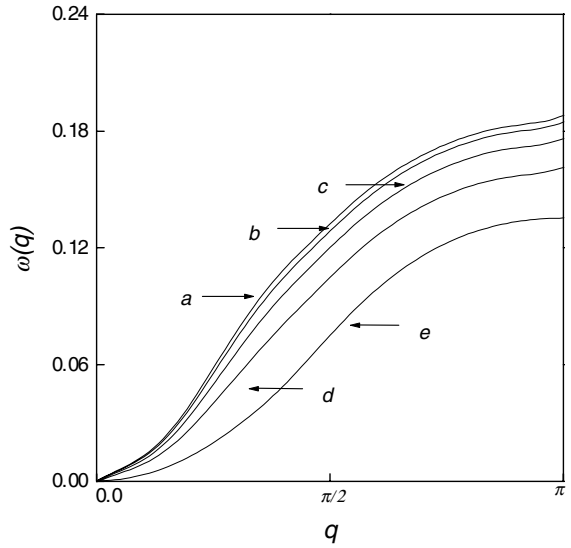

Figure 5. The acoustic modes for different inter-site e–e Coulomb repulsion: (a) $v = v' = 0.0$, (b) $v = v' = 0.2$, (c) $v = v' = 0.4$, (d) $v = v' = 0.6$ and (e) $v = v' = 0.8$ when $t' = 0.9$ and $u = u' = 1.2$.

It should be noticed that the mean-field theory and random-phase approximation (RPA), which neglect the fluctuations, are not sufficient to discuss the complete physical picture associated with the (quasi-) one-dimensional system. However, realistic materials are just the quasi-one-dimensional chain, and for some organic polymers such as polyacetylene [16–18] and organic ferromagnets [11–13], many significant results can be obtained from the mean field approximation and RPA while studying their energy bands or excitation energies. In this paper, within the mean-field approximation and RPA, we just study the ground state and low-lying excited state of the system at absolute zero temperature since most organic ferromagnets exhibit spontaneous magnetization at very low temperature near $T = 0$ K; the fluctuations can be neglected temporarily. We also noticed that the relative physical pictures obtained by employing some other methods, such as quantum Monte Carlo simulation [19], in a quasi-one-dimensional Hubbard chain are similar to those obtained from our Hartree–Fock approximation [11].

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 1(a). The Hubbard on-site electron–electron repulsion as well as the inter-site electron–electron Coulomb repulsion is taken into account. With mean-field theory, the system shows a 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 characteristic of the ferromagnetic magnon. The acoustic mode is influenced by the inter-site electron–electron Coulomb repulsion, and with the increase of the inter-site electron–electron Coulomb repulsion, the stability of the high-spin ferromagnetic ground state of the system will be spoiled.

Acknowledgments

One of the authors (L Zhao) would like to acknowledge 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 Nos 19777101 and 19774023.

Appendix

Equations for $A_i(k)$ $(i = 1, 2, \ldots, 5)$:

$$\begin{aligned}
A_i(k) = &\left[\omega_\alpha(q) - E_\tau\left(k+\frac{q}{2}\right) + E_\nu\left(k-\frac{q}{2}\right)\right]^{-1}\left[\sum_{m=1}^{3}[u+(u'-u)\delta_{m,3}]\right.\\
&\times\left\{A_i(k)\left[V^*_{m\tau}\left(k+\frac{q}{2}\right)V_{m\tau}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_1(k';m)\right.\right.\\
&\left.-V^*_{m\nu}\left(k-\frac{q}{2}\right)V_{m\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_2(k';m)\right]\\
&+(1-\delta_{i,5})\left[A_{i'}(k)V^*_{m\tau}\left(k+\frac{q}{2}\right)V_{m\nu'}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_1(k';m)\right.\\
&\left.-A_{i''}(k)V^*_{m\tau'}\left(k-\frac{q}{2}\right)V_{m\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_2(k';m)\right]\\
&\left.-V^*_{m\tau}\left(k+\frac{q}{2}\right)V_{m\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_3(k';m)\right\}\\
&+\sum_{m=1,3}[v+(v'-v)\delta_{m,3}]\left\{(1+\delta_{m,1})\left(A_i(k)\left[V^*_{2\tau}\left(k+\frac{q}{2}\right)V_{2\tau}\left(k+\frac{q}{2}\right)\frac{1}{N}\right.\right.\right.\\
&\times\sum_{k'}\zeta_1(k';m)+V^*_{m\tau}\left(k+\frac{q}{2}\right)V_{m\tau}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\xi_1(k')-V^*_{2\nu}\left(k-\frac{q}{2}\right)\\
&\left.\times V_{2\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_2(k';m)-V^*_{m\nu}\left(k-\frac{q}{2}\right)V_{m\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\xi_2(k')\right]\\
&+(1-\delta_{i,5})\left[A_{i'}(k)\left[V^*_{2\tau}\left(k+\frac{q}{2}\right)V_{2\nu'}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\zeta_1(k';m)+V^*_{m\tau}\left(k+\frac{q}{2}\right)\right.\right.\\
&\left.\times V_{m\nu'}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\xi_1(k')\right]-A_{i''}(k)\left[V^*_{2\tau'}\left(k-\frac{q}{2}\right)V_{2\nu}\left(k-\frac{q}{2}\right)\right.\\
&\left.\left.\left.\times\frac{1}{N}\sum_{k'}\zeta_2(k';m)+V^*_{m\tau'}\left(k-\frac{q}{2}\right)V_{m\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\xi_2(k')\right]\right]\right)\\
&-V^*_{2\tau}\left(k+\frac{q}{2}\right)V_{m\nu}\left(k-\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\eta_1(k,k';m)\\
&\left.\left.-V^*_{m\tau}\left(k+\frac{q}{2}\right)V_{2\nu}\left(k+\frac{q}{2}\right)\frac{1}{N}\sum_{k'}\eta_2(k,k';m)\right\}\right]
\end{aligned}$$

where

$$\zeta_1(k';m) = V^*_{m2}(k')V_{m2}(k') + V^*_{m3}(k')V_{m3}(k') \qquad \zeta_2(k';m) = V^*_{m3}(k')V_{m3}(k')$$

$$\zeta_3(k';m) = A_1(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right) + A_2(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)$$

$$+A_3(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)+A_4(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)$$
$$-A_5(k')V^*_{m1}\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)$$

$$\xi_1(k')=V^*_{22}(k')V_{22}(k')+V^*_{23}(k')V_{23}(k') \qquad \xi_2(k')=V^*_{23}(k')V_{23}(k')$$

$$\eta_1(k,k';m)=\{1+\exp[-\mathrm{i}(k'-k)]\}\Bigg[A_1(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{21}\left(k'+\frac{q}{2}\right)$$
$$+A_2(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{21}\left(k'+\frac{q}{2}\right)+A_3(k')V^*_{m2}\left(k'-\frac{q}{2}\right)V_{22}\left(k'+\frac{q}{2}\right)$$
$$+A_4(k')V^*_{m3}\left(k'-\frac{q}{2}\right)V_{22}\left(k'+\frac{q}{2}\right)-A_5(k')V^*_{m1}\left(k'-\frac{q}{2}\right)V_{23}\left(k'+\frac{q}{2}\right)\Bigg]$$

$$\eta_2(k,k';m)=\{1+\exp[-\mathrm{i}(k-k')]\}\Bigg[A_1(k')V^*_{22}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)$$
$$+A_2(k')V^*_{23}\left(k'-\frac{q}{2}\right)V_{m1}\left(k'+\frac{q}{2}\right)+A_3(k')V^*_{22}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)$$
$$+A_4(k')V^*_{23}\left(k'-\frac{q}{2}\right)V_{m2}\left(k'+\frac{q}{2}\right)-A_5(k')V^*_{21}\left(k'-\frac{q}{2}\right)V_{m3}\left(k'+\frac{q}{2}\right)\Bigg]$$

and

$$i'=3,\ i''=2,\ \tau=1,\ \tau'=3,\ \nu=2,\ \nu'=2 \qquad \text{for } i=1;$$
$$i'=4,\ i''=1,\ \tau=1,\ \tau'=2,\ \nu=3,\ \nu'=2 \qquad \text{for } i=2;$$
$$i'=1,\ i''=4,\ \tau=2,\ \tau'=3,\ \nu=2,\ \nu'=1 \qquad \text{for } i=3;$$
$$i'=2,\ i''=3,\ \tau=2,\ \tau'=2,\ \nu=3,\ \nu'=1 \qquad \text{for } i=4;$$
$$\tau=1,\ \nu=3 \qquad \text{for } i=5.$$

Here, wavevectors $|k|$, $|k'|$ and $|q|$ take their values from zero to π. Equations for $A'_i(k)$ ($i=1,2,\ldots,5$) have a similar form to the above equations.

References

[1] Macedo A M S, dos Santos M C, Coutinho-Filho M D and Macedo C A 1995 *Phys. Rev. Lett.* **74** 1851
[2] Fukutome H, Takahashi A and Ozaki M 1987 *Chem. Phys. Lett.* **133** 34
[3] Koneravka M, Kellnerova V, Kopcansky P and Kuczyski T 1995 *J. Magn. Magn. Mater.* **140–144** 1455
[4] Bagus P S and Torrance J B 1989 *Phys. Rev.* B **9** 7301
[5] Cao Y, Wang P, Hu Z, Li S Z, Zhang L Y and Zhao J G 1988 *Synth. Met.* **27** B625
[6] Korshak Y V, Medvedeva T V, Ovchinnikov A A and Spector V N 1987 *Nature* **326** 370
[7] Iwamura H, Sugawara T, Ioth K and Takui T 1985 *Mol. Cryst. Liq. Cryst.* **125** 379
[8] Takahashi M, Turk P, Nakazawa Y, Tamura M, Nozawa K, Shioni D, Ishikawa M and Kinoshita M 1991 *Phys. Rev. Lett.* **67** 746
[9] McConnell H M 1963 *J. Chem. Phys.* **39** 1910
[10] Ovchinnikov A A and Spector V N 1988 *Synth. Met.* **27** B615
[11] Fang Z, Liu Z L and Yao K L 1994 *Phys. Rev.* B **49** 3916
[12] Wang Y Q, Xiong Y S, Yi L and Yao K L 1996 *Phys. Rev.* B **53** 8481
[13] Nasu K 1986 *Phys. Rev.* B **33** 330
[14] Mattis D C 1981 *The Theory of Magnetism I* (Berlin: Springer)
[15] Tahir-Kheli R A 1976 *Phase Transition and Critical Phenomena* vol 5B, ed C Domb and M S Green (London: Academic)
[16] Safstrom S 1985 *Phys. Rev.* B **32** 4060
[17] Boudreaux D S, Chance R R and Bredas J L 1983 *Phys. Rev.* B **28** 6927
[18] Hayashi H and Nasu K 1985 *Phys. Rev.* B **32** 5295
[19] Liang D L, Wang Z D, Wang Q H and Shen S Q 1999 *Phys. Rev.* B **59** 3321