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DMRG studies on interchain coupling model for quasi-one-dimensional organic magnet

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Abstract. We carry out Density Matrix Renormalization Group (DMRG) calculations on the interchain coupling model for the quasi-one-dimensional organic ferromagnets under open boundary condition. Considering the topology of the system, the two-step DMRG technique is adopted. It is found that in the high spin ground state of the system the interchain coupling is in favor of the stability of ferromagnetic state, and the on-site electron-electron repulsive interaction has the same effect. However the inter-site electron-electron interaction causes a charge density distribution along the main chain and weakens the stability of the ferromagnetic ground state. The lattice distortion along the main chain is also discussed.

PACS. 75.40.Mg Numerical simulation studies – 75.30.Ds Spin waves

1 Introduction

Since the ferromagnetic ordering was discovered in an organic compound, p-NPNN(p-nitrophenyl nitronylnitroxide) [1,2] about 10 years ago, more than 20 organic ferromagnets based on ferromagnetic intermolecular interactions have been synthesized [3]. Thus the challenge of understanding the organic ferromagnet has led to much interest. For a zigzag chain consisting of π-conjugated carbon atoms and a radical at every other site, the Hamiltonian is described by the Su-Schrieffer-Heeger model supplemented by spin coupling between the π-electron and the radical electrons. The effect of interchain coupling in this structure has also been studied for an itinerant electron model including the lattice displacement and electron-phonon coupling within the Hatree-Fock approximation [4–6]. Although the ferromagnetic order in the ground state of these models is obtained numerically and analytically, and most of efforts are made by means of mean-field theory, some other interesting properties are not known yet and more appropriate theoretical methods are needed to study these systems.

In recent years, the density-matrix renormalization group (DMRG) method introduced by White [7,8] has proven a remarkable efficient one in the computation of the ground state, the thermodynamic and the dynamic properties of one-dimensional interacting electron models [9]. It has also been extended to various systems including quasi-one-dimensional organic and inorganic materials [10], weakly coupled ladder system [11], carbon nano-tube ropes [12], quasi-one-dimensional spin-Peierls systems [13], etc. However the investigation of organic ferromagnets by DMRG is only at the beginning.

In this article, we use DMRG to study an interchain coupling model considering the interchain electron transfer between the corresponding sites on the nearest neighboring chains. The simplified structure of the quasi-one-dimensional organic polymer ferromagnets is schematically shown in Figure 1. As is known, the DMRG method has trouble dealing with periodic boundary condition, so we discuss the total energy, the spin configuration and the lattice distortion with open boundary condition. In Section 2, the theoretical model and the computational process are given. In Section 3 we discuss the effect of interchain coupling parameters, boundary condition, electron-phonon interaction on the system. Finally we will give a brief summary of our results in Section 4.

2 The theoretical model and computational method

Considering two neighboring coupling chains, the Hamiltonian is written as:

$$H = H_1 + H_2 + H' \tag{1}$$

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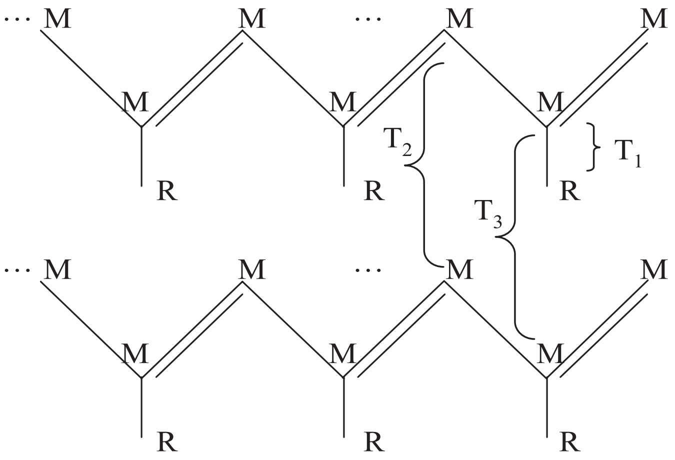

Fig. 1. Two neighboring chains of an organic ferromagnet. "M" labels the site on the main chain and "R" is the side radical. T_1 denotes the hopping integral between the π-electron on the main chain and the unpaired electron at the side radical, and T_2 (T_3) are interchain electron hopping from the odd site(even site) in the first chain to the corresponding odd site (even site) of the second chain.

$$H_1 = -\sum_{jl\sigma}[t_0+\gamma(u_{jl}-u_{jl+1})]\left(c^{+}_{j1l\sigma}c_{j1l+1\sigma}+\text{h.c.}\right) + \frac{\kappa}{2}\sum_{jl}(u_{jl}-u_{jl+1})^2 - \sum_{jl\sigma}\left(T_1 c^{+}_{j1l\sigma}c_{j2l\sigma}\delta_l+\text{h.c.}\right) \tag{2}$$

$$H_2 = U\sum_{jl}(n_{j1l\uparrow}n_{j1l\downarrow}+n_{j2l\uparrow}n_{j2l\downarrow}\delta_l) + V\sum_{jl\sigma\sigma'}(n_{j1l\sigma}n_{j1l+1\sigma'}+n_{j1l\sigma}n_{j2l\sigma'}\delta_l) \tag{3}$$

$$H' = -\sum_{l\sigma}[T_2(1-\delta_l)+T_3\delta_l](c^{+}_{11l\sigma}c_{21l\sigma}+\text{h.c.}). \tag{4}$$

The first term H_1 describes the interchain hopping of the $\pi-$electrons on each main chain and the unpaired electrons at side radicals, the electron-phonon interaction and the distortion of the lattice, where $c^{+}_{jil\sigma}(c_{jil\sigma})$ denotes the creation(annihilation) operator of a $\pi-$electron ($i=1$) along each main chain or an unpaired electron ($i=2$) at a side radical with spin σ on the lth site of the jth chain, t_0 is the nearest neighboring hopping integral of the $\pi-$electron along each main chain when there is no distortion, T_1 is the nearest neighboring hopping integral between the $\pi-$electron on the main chain and the unpaired electron at side radical, γ is the electron-phonon coupling constant, u_{jl} is the displacement of the lth site of the jth main chain, κ is the elastic constant of the lattice. We assume that the side radicals connect with the even carbon atoms, then $\delta_l=1$ for an even site and $\delta_l=0$ for odd site.

The second term H_2 describes the Hubbard on-site electron-electron repulsion (the first term in H_2) and the nearest-neighboring Coulomb repulsion (the second term in H_2) of the $\pi-$electron and the unpaired electron at side radical. $n_{jil\sigma}=c^{+}_{jil\sigma}c_{jil\sigma}$ where σ denotes spin up or spin down. H' describes the interchain interaction, where $T_2(T_3)$ is the interchain electron transfer from the odd site (even site) of the first main chain to the corresponding odd site (even site) of the second main chain.

It is convenient to cast all quantities into dimensionless forms as

$$h=\frac{H}{t_0}, u=\frac{U}{t_0}, v=\frac{V}{t_0}, t_i=\frac{T_i}{t_0}, (i=1,2,3,4), \tag{5}$$

$$\lambda=\frac{2\gamma^2}{t_0\pi\kappa}, y_{jl}=\frac{(-1)^l(u_{jl}-u_{jl+1})\gamma}{t_0}. \tag{6}$$

Then, the Hamiltonian is transformed as follows:

$$h=-\sum_{jl\sigma}[1+y_{jl}]\left(c^{+}_{j1l\sigma}c_{j1l+1\sigma}+\text{h.c.}\right)+\frac{1}{\pi\lambda}\sum_{jl}y^2_{jl} - \sum_{jl\sigma}\left(t_1c^{+}_{j1l\sigma}c_{j2l\sigma}\delta_l+\text{h.c.}\right)+u\sum_{jl}(n_{j1l\uparrow}n_{j1l\downarrow} + n_{j2l\uparrow}n_{j2l\downarrow}\delta_l)+v\sum_{jl\sigma\sigma'}(n_{j1l\sigma}n_{j1l+1\sigma'}+n_{j1l\sigma}n_{j2l\sigma'}\delta_l) - \sum_{l\sigma}[t_2(1-\delta_l)+t_3\delta_l]\left(c^{+}_{11l\sigma}c_{21l\sigma}+\text{h.c.}\right) \tag{7}$$

where λ is defined as the electron-phonon coupling parameter. Using the Hellmann-Feynman theorem, we have the self-consistency equation:

$$y_{jl}=(-1)^l\pi\lambda\left(p_{jl}-\frac{1}{N}\sum_m p_{jm}\right). \tag{8}$$

Here, N is the number of the sites along each main chain and the bond order is defined as:

$$p_{jl}=\left\langle\sum_{\sigma}\left(c^{+}_{j1l\sigma}c_{j1l+1\sigma}+\text{h.c.}\right)\right\rangle. \tag{9}$$

According to the above Hamiltonians and the topological structure of the interchain coupling system, we use open boundary condition by DMRG algorithm. The whole computational process consists of two steps. In the first step only the single chain is constructed in finite DMRG scheme. We start with four sites of the main chain with two radicals and systematically add one atom, or one atom and one radical bond to it in the middle of the system, building up the molecule in such a way that the right block states are obtained by the reflection of the left block symmetrically. In this implementation we store the matrices corresponding to the operators of the left block for convenience of the next step, so it takes up a large storage space. Subsequently in the second step, numerical renormalization algorithm is implemented for the two coupled chains

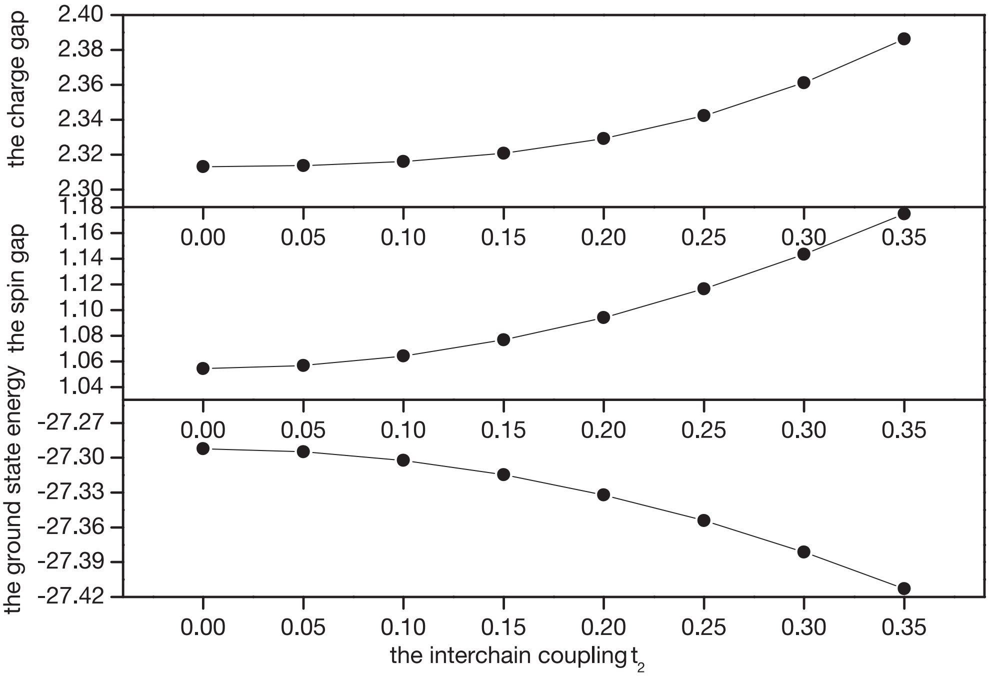

Fig. 2. The ground state energy, the spin gap Δ_s and the charge gap Δ_c as a function of the the coupling t_2 with $u = 1.0$, $v = 0.0$, $t_3 = 0.3$, $\lambda = 0.4$.

and a transformation is made by use of the eigenstates of the single chain in the previous step. Then the spin density of every site and the total energy of the system and other related properties can be calculated by means of the ground state wave function. The similar conception has been exhibited in Moukouri and Caron's work [14], where they studied the anisotropic quantum spin half Heisenberg model on a square lattice by combing the DMRG method and the renormalization group (RG) method.

The feasibility and the rationality of the two-step DMRG method are judged as follows: for our Hubbard model Hamiltonian, the dimension of the Hilbert space is of the order of more than 4^L, where L is the number of atoms of the system. It is very difficult for the traditional DMRG to treat this kind of systems. However in this work we first obtain the low energy Hamiltonian of the single chain with radicals connected to even site, so that the ground state properties is reserved in the block form and the dimension of the Hamiltonian is largely reduced. Furthermore, to enhance the accuracy of our calculations, in the first step we have done DMRG calculation with at most 120 density matrix eigenvectors reserved after making sure that the energy converges for this truncation. The estimated truncation error is typically of the order of 10^{-5}. The stability of the optimized configuration is ensured since the difference between two successive iterations is less than 10^{-8} for lattice configuration. In the second step, we increase the up-spin electron number from 0 to the total electron number L in the case of half-filling. The results show that the lowest energy state lies in the subspace of the total spin $S^Z = 1/2 \times L/3$, and most of the states are in its nearby subspaces, which is the case of weak interchain interaction.

3 Results and discussions

We study two neighboring open chains shown in Figure 1. Each chain includes 10 carbon atoms and 5 side radicals due to the restriction of computer memory. Since the investigation on the organic ferromagnetic materials is at the initial stage, in our calculation we take the electron-phonon interaction λ, the Hubbard electron-electron correlation u, which are unknown yet, as the same order as those of polyacetylene. The hopping term t_1 is set as 0.9, which is smaller than that between two neighboring π-electrons since the radical electron is relatively localized.

First, we study the total energy and the ferromagnetic stability of the system. Due to the topological structure of the system, different interchain couplings with respect to different sites of the chain will be also discussed. This is similar to the situations for typical organic polymers with chain-like structure, such as polyacene, in which the interchain interaction is the interchain hopping of electrons, but phonons are strictly one dimensional. With these parameters, we find the ground state is in the subspace of the total spin $S^Z = 1/2 \times L/3 = 5$, where L is the total electron number. So the ground state of the system is a high spin ferromagnetic state. Our results are shown in Figure 2a for the lowest energy of the system with different interchain coupling t_2 and definite $t_3 = 0.3$, $u = 1$, $\lambda = 0.4$. We can see clearly that the interchain coupling decreases the total energy of the system so that the stability of ferromagnetic state becomes stronger.

Generally due to dimerization and Peierls instability, there exists an energy gap in quasi-one-dimensional systems. From the results of the mean-field theory study on interchain coupling model, it is analyzed that there is a

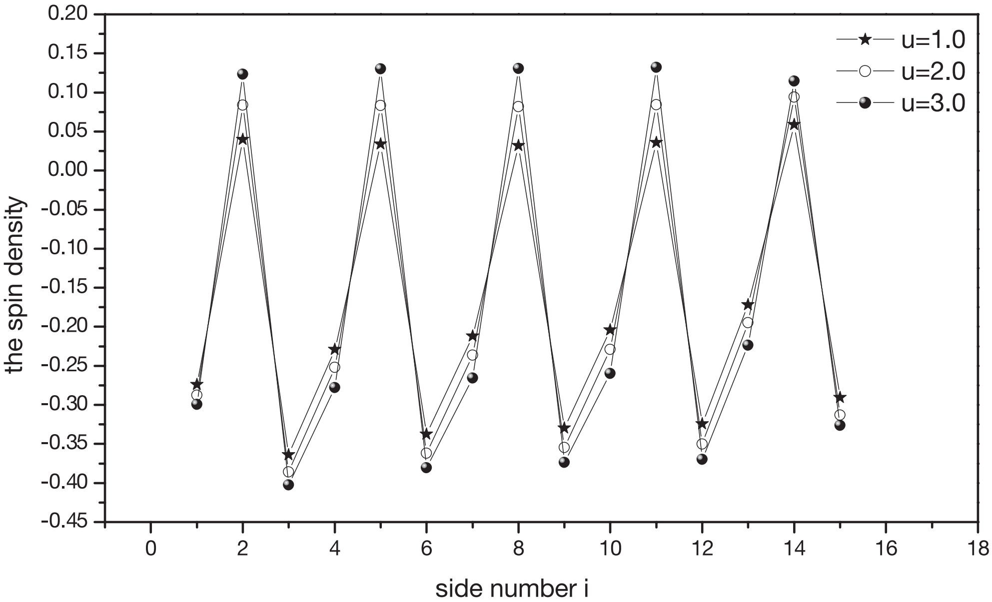

Fig. 3. The spin density of the main chain ($n = 3m + 1$, $3m + 2$, $m = 0$, 1, 2, 3 ...) and the radical sides ($n = 3m$, $m = 1$, 2, 3 ...) with $t_2 = t_3 = 0.2$, $v = 0.0$ and $\lambda = 0.2$.

split between spin-up and spin-down states near the Fermi level and the energy gap exists between the occupied states and the unoccupied states [5]. For the estimation of the energy gap, we define the spin gap $\Delta_s(L)$ and the charge gap $\Delta_c(L)$ at the half filling [14] as follows:

$$\Delta_s(L) = E_0\left(\frac{2L}{3}+1, \frac{L}{3}-1\right) - E_0\left(\frac{2L}{3}, \frac{L}{3}\right)$$
$$\Delta_c(L) = E_0\left(\frac{2L}{3}+1, \frac{L}{3}\right) + E_0\left(\frac{2L}{3}-1, \frac{L}{3}\right) - 2E_0\left(\frac{2L}{3}, \frac{L}{3}\right)$$

where $E_0(N_\uparrow, N_\downarrow)$ is the lowest energy of the system with $N_\uparrow$ up and $N_\downarrow$ down spin electrons, and the $E_0(\frac{2L}{3}, \frac{L}{3})$ is the ground state energy which is obtained from computational results. In Figures 2b and c we give the spin gap $\Delta_s(L)$ and the charge gap $\Delta_c(L)$ as a function of the interchain coupling t_2, respectively. It can be seen that the charge gap $\Delta_c(L)$ is bigger than spin gap $\Delta_s(L)$ and the both gaps increase with the increase of the interchain coupling. So we think the system shows the insulating property and the interchain coupling is in favor of the stability of the ferromagnetic ground state.

Second, we discuss the distribution of the spin density and the charge density. When neglecting the inter-site electron-electron interaction ($v = 0$) and considering only the on-site electron-electron repulsive interaction, it is found that there appears antiferromagnetic spin density wave (SDW) along the main chain. Mediated by the SDW, the ferromagnetic ordering is formed by the concentrated distribution of the spin density on the side radical [4]. Here, the appearance of the SDW state is the result of the on-site electron-electron repulsive interaction. Figure 3 shows the distribution of the spin density on the main chain and the side radical, which corresponds to $n = 3m + 1, 3m + 2$, and $n = 3m + 3$, respectively (m is taken as an integer from 0 to 9). It can be seen that the amplitude of the SDW along the main chain and the side radical will increase with the increasing of the on-site electron-electron repulsive interaction u. Thus the ferromagnetic exchange interaction of the spins of the unpaired electrons at side radicals will become stronger and the ferromagnetic ground state of the system will be more stable. When adding the inter-site electron-electron interaction, the charge density is no longer distributed uniformly. Figure 4 gives the distribution of the spin density and the charge density on the main chain and the side radical for different inter-site electron-electron interaction v. We can see that the increase of the inter-site repulsion v decreases the spin density amplitude of the radical spin and strengthens the amplitude of the charge density along the main chain. So it is argued that with the increasing of the inter-site repulsion v, the system will transfer from a SDW into a CDW state. Consequently the stability of the ferromagnetic state will be weakened.

Finally we discuss the dimerization of the system along each chain. The dimerization accompanied with the Peierls transition is a unique feature in one dimensional system. Figure 5 displays that with small electron-phonon interaction the amplitude of the distortion changes alternatively along the main chain and the dimerization-like distortion occurs. When increasing the electron-phonon coupling λ, the dimerization parameter decreases.

4 Conclusions

We have studied the interchain coupling model for a quasi-one-dimensional organic ferromagnet using DMRG

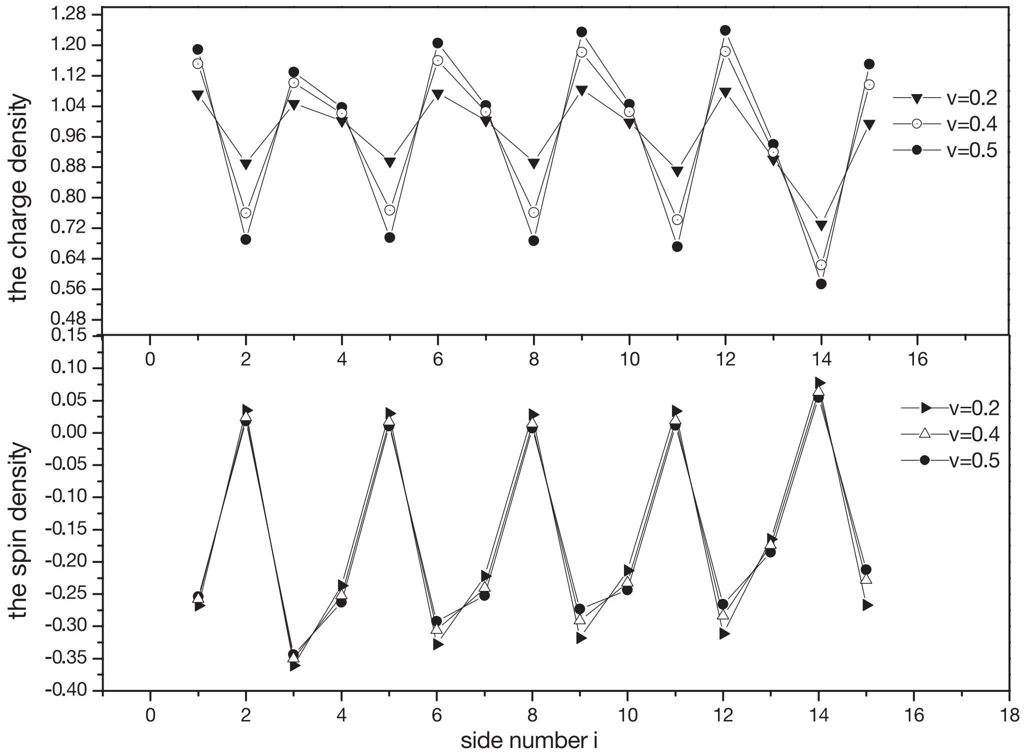

Fig. 4. The charge density and the spin density of the main chain sites ($n = 3m + 1$, $3m + 2$, $m = 0$, 1, 2, 3 ...) and side radical sites ($n = 3m$, $m = 1$, 2, 3 ...) with $u = 1.0$, $t_2 = t_3 = 0.2$ and $\lambda = 0.2$.

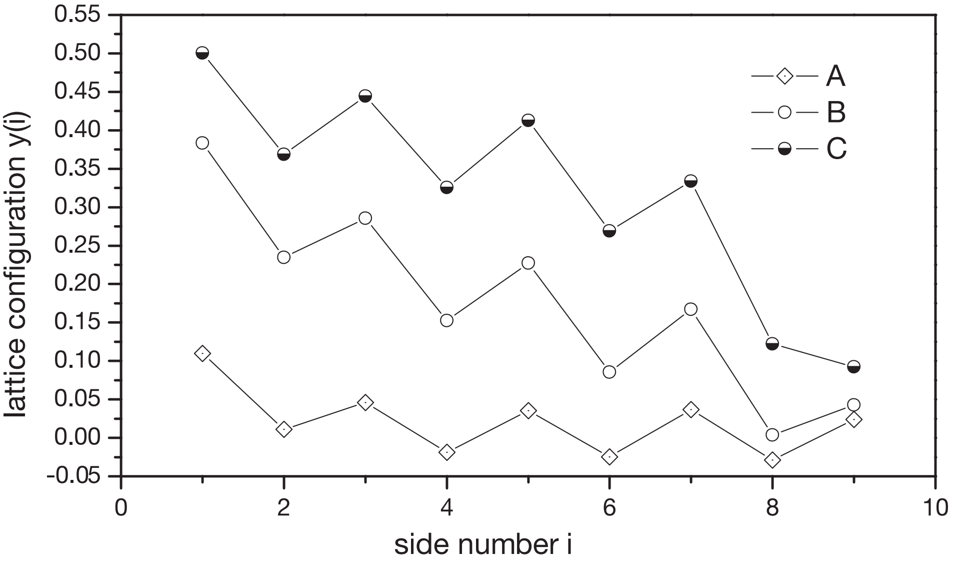

Fig. 5. The dimerization parameter y(i) along the main chain. The parameters are taken as $t_2 = t_3 = 0.1$, $u = 1.0$, $v = 0.0$. For curve *A*, *B*, *C*, $\lambda = 0.2$, 0.4, 0.45, respectively.

method with open boundary condition. The results of the calculation indicate that the ground state of the interchain coupling system discussed above is a high spin ferromagnetic state. The interchain coupling is in favor of the stability of the ferromagnetic state for definite parameters. The onsite and the intersite electron interaction and the electron-phonon interaction have competing influence on the ferromagnetism of the system. the larger on-site electron-electron repulsive interaction will make the ferromagnetic ground state of the system more stable, while the inter-site electron-electron interaction causes a charge density distribution along the main chain and consequently weakens the stability of the ferromagnetic ground state. With increase of electron-phonon coupling there is no longer a perfect dimerization along the main chain of the system. These results are in agreement with the solution of the mean-field theory. The two-step DMRG method is useful in understanding the mechanism of the ferromagnetic organic materials.

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