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MAGNON EXCITATION OF QUASI-ONE-DIMENSIONAL ORGANIC FERROMAGNET*

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A new quasi-one-dimensional organic ferromagnet model is proposed. Magnon excitations for finite and infinite on-site repulsion of electrons on main carbon chain are investigated. It is found that the lowest branch of spectrum preserves the characteristics of the ferromagnetic magnon. It is also shown that with increasing of the on-site repulsion energy, the itinerant π-electrons become tightly localized.

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Recently, there have been growing interests in magnetic properties of low-dimensional organic materials.[1-8] By now, several organic ferromagnets have been synthesized from organic molecules and hydrocarbons.[1-5] In these materials, there is no transition metal possessing itinerant or localized 3d or 4f electrons which contribute to magnetic exchange interaction as in ordinary ferromagnet. Theoretically, several models have been proposed to study the magnetic properties of these materials.[6-9] In general, π-electrons in these systems are itinerant along the main chain, which will significantly affect the characteristics of the organic ferromagnet. In this paper, our purpose is to investigate the magnon excitation of a new quasi-one-dimensional organic ferromagnet model with finite and infinite on-site repulsion of π-electrons on the main carbon chain.

The model (shown in Fig.1) Hamiltonian under present consideration reads

$$\begin{aligned}\widehat{H} = & -\sum_{l,\sigma}\left(\widehat{a}^{+}_{l,2,\sigma}\widehat{a}_{l,1,\sigma}+\widehat{a}^{+}_{l+1,2,\sigma}\widehat{a}_{l,1,\sigma}+\text{H.c.}\right) \\ & +u\sum_{l}\sum_{m}\widehat{n}_{lm\alpha}\widehat{n}_{lm\beta}+j_2\sum_{l}\left[\widehat{S}^{z}_{l,0}\widehat{S}^{z}_{l,1}+\frac{1}{2}\left(\widehat{S}^{-}_{l,0}\widehat{S}^{+}_{l,1}+\widehat{S}^{+}_{l,0}\widehat{S}^{-}_{l,1}\right)\right], \qquad (1)\end{aligned}$$

where $\widehat{a}^{+}_{l,m,\sigma}(\widehat{a}_{l,m,\sigma})$ denotes the creation (annihilation) operator of the electron at the site specified by l, m and spin $\sigma, l(=1,2,\cdots,N)$ labels the unit cell which contains one residual spin and two carbon sites of the main chain, $m(=0,1,2)$ labels the residual spin and two carbon sites as shown in Fig.1 and $\sigma(=\alpha$ and $\beta)$ denotes up and down spins, respectively, $u(>0)$ denotes the effective repulsive energy between two π-electrons when they are on the same carbon site, $j_2(>0)$ is the exchange energy between the residual spin and π-electrons

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at carbon site $m = 1$. Since each radical in one-dimensional organic ferromagnet contains a free electron, spin operators in Eq.(1) can be written as

$$\widehat{S}^z_{lm} = \frac{1}{2}\left(\widehat{n}_{lm\alpha} - \widehat{n}_{lm\beta}\right) , \quad \widehat{S}^+_{lm\sigma} = \widehat{a}^+_{lm\alpha}\widehat{a}_{lm\beta} \qquad (m = 1, 2) . \tag{2}$$

First, we will be concerned with the case of finite on-site repulsion u. Following the procedure in Ref.[9], we describe the magnon excitation for α branch in terms of linear combination[6],

$$\widehat{\gamma}^+_{\alpha q} = A_1\widehat{\eta}^+_q + \sum_k A_2(k)\widehat{C}^+_{k-\frac{q}{2},1,\beta}\widehat{C}_{k+\frac{q}{2},1,\alpha} + \sum_k A_3(k)\widehat{C}^+_{k+\frac{q}{2},2,\beta}\widehat{C}_{k-\frac{q}{2},2,\alpha} , \tag{3}$$

where $\widehat{\eta}^+_q$ is the spin deviation operator of the residual spin after using Holstein-Primakoff transformation, and $\widehat{C}^+_{k,i,\sigma}(\widehat{C}_{k,i,\sigma})$ is the electron creation (annihilation) operator. With the help of motion equations of magnon excitation in the random-phase-approximation (RPA), a set of self-consistent equations for $A_1, A_2(k)$ and $A_3(k)$ can be obtained readily

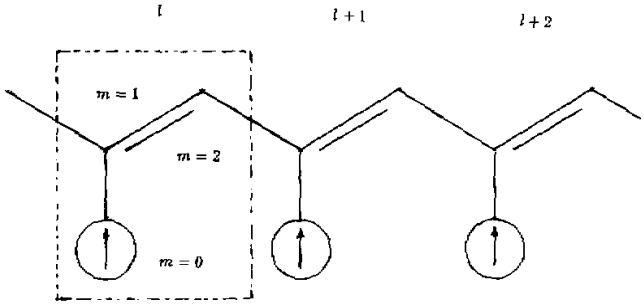

Fig. 1. The quasi-one-dimensional organic ferromagnetic model under our consideration, where l stands for the position of a unit cell and $m = 0, 1$ and 2 label the sites of the residual spin and the two electrons on the main chain in a unit cell.

$$A_1 = [\omega_\alpha(q)]^{-1}\left[-\frac{j_2}{2}\frac{1}{N}\sum_k\left\{A_2(k)V_{11}(k+\frac{q}{2})V_{11}(k-\frac{q}{2}) - A_3(k)V_{12}(k+\frac{q}{2})V_{12}(k-\frac{q}{2})\right\}\right] , \tag{4}$$

$$\begin{aligned} A_2(k) \;=\; & \left[\omega_\alpha(q)+\omega_1(k+\frac{q}{2})+\omega_1(k-\frac{q}{2})\right]^{-1} \\ & \times\Bigg\{u\sum_m V_{m1}(k+\frac{q}{2})V^*_{m1}(k-\frac{q}{2})\frac{1}{N}\sum_{k'}A_2(k')V_{m1}(k'+\frac{q}{2})V^*_{m1}(k'-\frac{q}{2}) \\ & +u\sum_m A_2(k)\left[\frac{(1-2\delta n_m)}{2}V_{m1}(k+\frac{q}{2})V^*_{m1}(k+\frac{q}{2}) - \frac{(1+2\delta n_m)}{2}V_{m1}(k-\frac{q}{2})V^*_{m1}(k-\frac{q}{2})\right] \\ & -u\sum_m V_{m1}(k+\frac{q}{2})V^*_{m1}(k-\frac{q}{2})\frac{1}{N}\sum_{k'}A_3(k')V_{m2}(k'+\frac{q}{2})V^*_{m2}(k'-\frac{q}{2}) \\ & +\frac{A_1 j_2}{2}V_{11}(k+\frac{q}{2})V^*_{11}(k-\frac{q}{2})\Bigg\} , \end{aligned} \tag{5}$$

$$
\begin{aligned}
A_3(k) \;=\; & \left[\omega_\alpha(q)+\omega_2(k+\frac{q}{2})+\omega_2(k-\frac{q}{2})\right]^{-1} \\
& \times\Bigg\{u\sum_m V_{m2}(k+\frac{q}{2})V^*_{m2}(k-\frac{q}{2})\frac{1}{N}\sum_{k'}A_2(k')V_{m1}(k'+\frac{q}{2})V^*_{m1}(k'-\frac{q}{2}) \\
& +u\sum_m A_3(k)\left[\frac{(1-2\delta n_m)}{2}V_{m2}(k+\frac{q}{2})V^*_{m2}(k+\frac{q}{2})-\frac{(1+2\delta n_m)}{2}V_{m2}(k-\frac{q}{2})V^*_{m2}(k-\frac{q}{2})\right] \\
& -u\sum_m V_{m2}(k+\frac{q}{2})V^*_{m2}(k-\frac{q}{2})\frac{1}{N}\sum_{k'}A_3(k')V_{m2}(k'+\frac{q}{2})V^*_{m2}(k'-\frac{q}{2}) \\
& +\frac{A_1 j_2}{2}V_{12}(k+\frac{q}{2})V^*_{12}(k-\frac{q}{2})\Bigg\}\,, \qquad (6)
\end{aligned}
$$

where $\omega_\alpha(q)$ denotes magnon excitation energy for α branch, $\omega_i(k)$ and $V_{ij}(k)$ represent the eigenvalue and corresponding eigenvector of the ground state, respectively, with $m = 1, 2$. Wave vectors $|k|, |k'|$ and $|q|$ take their values from zero to π. The self-consistent equations for $\widehat{\gamma}^+_{\beta q}$ is similar to those for $\widehat{\gamma}^+_{\alpha q}$. It is easy to prove that the coefficients $A_1, A_2(k)$ and $A_3(k)$ satisfy the normalization condition

$$
|A_1|^2-\sum_k|A_2(k)|^2+\sum_k|A_3(k)|^2=1\,. \qquad (7)
$$

Furthermore, solving self-consistent Eqs.(4)–(7), we can obtain the dispersion relation of magnon excitation for different branches.

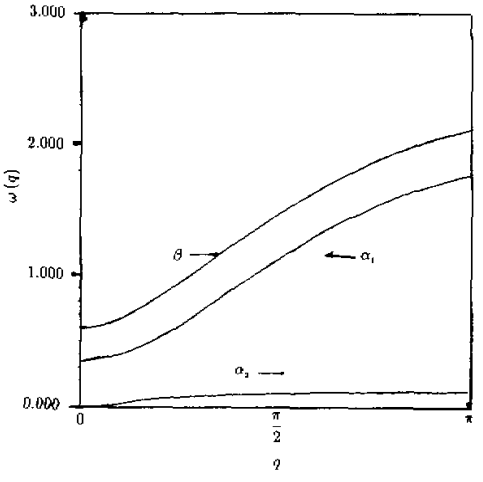

Fig. 2. The dispersion relation of magnon for $u = 2, j_2 = 0.75$.

Figure 2 exhibits a typical dispersion relation of magnon excitation, in which the two lower energy branches of magnons $\alpha_{1,2}$ refer to those of $\widehat{\gamma}^+_{\alpha q}$ and the highest one β attributes to that of $\widehat{\gamma}^+_{\beta q}$. This kind of excitation pattern comes from the topological structure of the system. The lowest branch of magnons is the excitation of radicals correlated indirectly with the conjugated electrons on the main carbon chain.

When the on-site repulsion is very large, the electron on the main chain becomes tightly localized. The energies of the two upper magnons tend to coincide at $q = 0$ (see Fig.3).

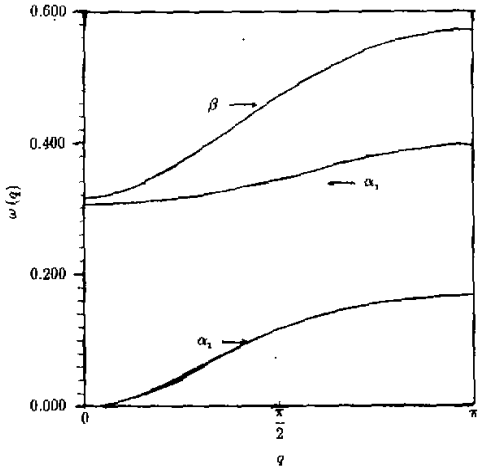

Fig. 3. The dispersion relation of magnon for $u = 10, j_2 = 0.75$.

In the extreme case $u \to \infty$, the electrons on the main chain are completely localized. The exchange interaction between electrons on the main chain can be reduced to an antiferromagnetic Heisenberg correlation with coupling constant $J_a(\equiv 4/u)$. Therefore, the model Hamiltonian (1) can be rewritten as

$$\hat{H}_s = J_a \sum_l (\hat{\vec{S}}_{l,1} \cdot \hat{\vec{S}}_{l,2} + \hat{\vec{S}}_{l+1,1} \cdot \hat{\vec{S}}_{l,2}) + j_2 \sum_l \hat{\vec{S}}_{l,0} \cdot \hat{\vec{S}}_{l,1} \,. \tag{8}$$

According to the standard spin wave theory[10], the model Hamiltonian in the wave vector representation can be expressed as

$$\begin{aligned} \hat{H}_s = & -\frac{N}{2}(2J_a + j_2) + \sum_q (2J_a + j_2)\hat{a}^+_{q1}\hat{a}_{q1} + 2J_a \sum_q \hat{a}^+_{q2}\hat{a}_{q2} \\ & + \sum_q j_2 \hat{a}^+_{q0}\hat{a}_{q0} + \sum_q 2J_a \cos^2(\frac{q}{2})(\hat{a}_{q1}\hat{a}_{q2} + \hat{a}^+_{q2}\hat{a}^+_{q1}) \\ & + \sum_q j_2(\hat{a}_{q1}\hat{a}_{q0} + \hat{a}^+_{q0}\hat{a}^+_{q1}) \,. \end{aligned} \tag{9}$$

Introducing Bogoliubov transformation

$$\begin{aligned} \hat{\gamma}^+_\alpha(q) &= u_1\hat{a}^+_{q1} + u_2\hat{a}_{q2} + u_3\hat{a}_{q0} \,, \\ \hat{\gamma}^+_\beta(q) &= u_4\hat{a}^+_{q1} + u_5\hat{a}_{q2} + u_6\hat{a}_{q0} \end{aligned} \tag{10}$$

to diagonalize the Hamiltonian (9), one obtains

$$(2j_a + j_2 - \omega(q))(2j_a - \omega(q))(j_2 - \omega(q)) - 4j_a^2\cos^4(\frac{q}{2})(j_2 - \omega(q)) - j_2^2(2j_a - \omega(q)) = 0 \,, \tag{11}$$

where $\omega_\sigma(q)$ is the excitation spectrum. The corresponding numerical result is shown in Fig.4. It is clear that the two upper magnons are degenerated at $q = 0$. Our result indicates that the itinerary of π-electrons significantly affects the magnon spectrum, especially at long-wavelength limit.

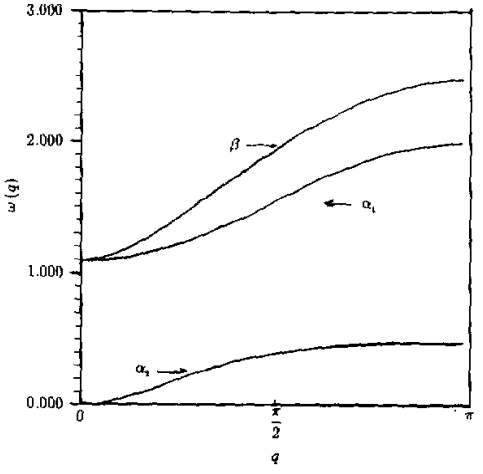

Fig. 4. The dispersion relation of magnon excitation for infinite u and $j_2/j_a = 0.60$.

To summary, a new quasi-one-dimensional organic ferromagnet model for different on-site repulsion is investigated numerically. three branches of magnon for finite and infinite on-site repulsion of π-electrons on main carbon chain are discussed. We find that the lowest branch of spectrum has a dispersion relation $\omega(q) \propto q^2$ as ordinary ferromagnet. In addition, it is shown that with the increase of the on-site repulsion energy the itinerant π-electrons become tightly localized.

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