CROATICA CHEMICA ACTA
CCACAA, ISSN 0011-1643, e-ISSN 1334-417X
Croat. Chem. Acta **86** (4) (2013) 431–434.
http://dx.doi.org/10.5562/cca2323

Original Scientific Article

U= ∞ Hubbard Model for 1D Frustrated Magnets†

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RECEIVED JULY 3, 2013; REVISED SEPTEMBER 15, 2013; ACCEPTED SEPTEMBER 16, 2013

Abstract. The energy spectrum of Hubbard model with infinite repulsion on two types of distorted necklace ladder with additional diagonal interactions is studied analytically and numerically. For the diamond type lattice we demonstrate the appearance of magnetic polarons at negative ratio of hopping parameters describing the interactions of neighbor unit cells. For second type of the lattice, despite of similar triangular topology, the corresponding polarons have nonmagnetic character. (doi: 10.5562/cca2323)

Keywords: Hubbard model, infinite electron repulsion, magnetic polarons, nanomagnets

INTRODUCTION

Polymeric quasi-one-dimensional compounds of transition metals with macroscopic value of ground state spin permanently attract much interest as the constitutional components of nanomagnets. Traditional theoretical approach to the simulation of its magnetic properties is based on 1D quantum spin models[1] and Lieb theorem.[2,3] For example, the isotropic spin-1/2 Heisenberg Hamiltonian on diagonal ladder lattice with three site unit cells (necklace ladder) describes adequately low-temperature magnetic properties of quasi-one-dimensional ferrimagnets like of IPA2CuCL4 (IPA-isopropylammonium).[4] The appearance of itinerant electrons due to the acceptor doping may change significantly the magnetic properties of these materials. This effect was described theoretically on the base of *t*-J model.[5,6] The simplest approach to the study of magnetism of itinerant electrons is a Hubbard model with infinite electron repulsion ($U = \infty$ Hubbard model). It represents restricted hopping in a space with no double occupied sites. Despite the simple algebraic structure of the model, the study of its spectrum is a complicated task, and corresponding exact results only in a few special cases are known. As has been shown by Nagaoka[7] and Thouless[8] for some type of lattices, the ground state of the one-band model with the one hole in a half-filled band has the maximal value of the total spin. Recently[9] ferromagnetic ground state was found for isotropic *n*-leg ladder $U = \infty$ Hubbard model with the density of electrons per site ρ in the range $0.8 \le \rho < 1$. In our previous works[10,11] it was shown that for $U = \infty$ Hubbard model on the rectangular lattice strips consisting of weakly interacted *n*-site segments there is a cascade of concentration transitions with the oscillation of the ground state multiplicity between minimal and maximal values due to the creation of magnetic polarons. In other words, this model demonstrates the ferromagnetism for finite concentrations of holes.

Necklace ladder with additional diagonal interactions is known as diamond chain. Since its discovery in a natural mineral azurite, this frustrated spin system is attracting a big attention.[12] It is of interest for a design of new magnetic materials to study the effect of itinerant electrons on the lowest energy states of frustrated electron system like diamond chain described by $U = \infty$ Hubbard model.

In our work we report the results of our study of low energy states of the Hubbard model with infinite repulsion defined on two types of frustrated lattices depicted on Figure 1. The first one is a distorted diamond chain formed by weakly interacting three-site unit cells. It also can be treated as distorted necklace ladder with 'vertical' diagonal interactions. Second lattice can be treated as a distorted necklace ladder with additional 'horizontal' diagonal interactions. By means of degenerate perturbation theory (PT) and cyclic spin permutation formalism we derived effec-

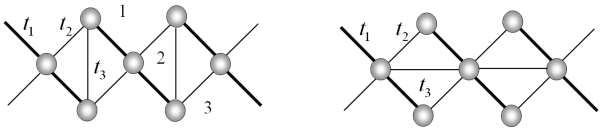

Figure 1. Two types of distorted necklace ladder with additional 'vertical' and 'horizontal' diagonal interactions.

† Dedicated to Professor Douglas Jay Klein on the occasion of his 70th birthday.

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tive Hamiltonians describing low-energy states of above systems. We have shown that despite of similar 'triangular' topology of the lattices, both models may demonstrate significantly different magnetic behavior. Thus for some region of model parameters the ground state spin of first model is unchanged with the increase of lattice size, whereas the ground state spin of second model is increased.

EFFECTIVE HAMILTONIANS

Let us enumerate all the site of chain by pair of indexes (i, k), where first index determines the number of unit cell and second one corresponds to the number of site of this cell. Let also enumerate all the electrons in succession of the unit cells. In cyclic spin permutation formalism the model Hamiltonian for diamond chain has the form

$$\begin{gathered}\mathbf{H} = t_1\mathbf{H}_0 + t_2\sum_{i=1}^{L-1}\mathbf{H}_{i,i+1} + t_3\sum_{i=1}^{L-1}\bar{\mathbf{H}}_{i,i+1}\\ \mathbf{H}_0 = \sum_{i=1}^{L}(\mathbf{a}_{i,1}^{+}\mathbf{a}_{i,2} + \mathbf{a}_{i,2}^{+}\mathbf{a}_{i,3}) + H.c.\\ \mathbf{H}_{i,i+1} = \mathbf{a}_{i+1,1}^{+}\mathbf{a}_{i,2}\mathbf{Q}_{m,n} + \mathbf{a}_{i+1,2}^{+}\mathbf{a}_{i,3}\mathbf{Q}_{m',n'} + H.c.\\ \bar{\mathbf{H}}_{i,i+1} = \mathbf{a}_{i+1,1}^{+}\mathbf{a}_{i,3}\mathbf{Q}_{m'',n''} + H.c.\end{gathered} \tag{1}$$

where $\mathbf{a}_{i,j}^{+}$ is a spin-free Fermi operator which acts only on the coordinate part of model wave function that describes the distribution of N electrons over the $3L$ lattice sites; $\mathbf{Q}_{m,n}$ is an operator of cyclic permutation of spin variables σ_k ($k = m, m+1,..., n$) of electrons, located on the lattice sites from the interval (i, j), where ith and jth sites are neighbors[13,14]

$$\mathbf{Q}_{m,n}\left|\sigma_m,\sigma_{m+1},\ldots\sigma_{n-1},\sigma_n\right\rangle = \left|\sigma_n,\sigma_m,\sigma_{m+1},\ldots\sigma_{n-1}\right\rangle;$$

For second type of lattice the Hamiltonian $\bar{\mathbf{H}}_{i,i+1}$ has the form:

$$\bar{\mathbf{H}}_{i,i+1} = \mathbf{a}_{i+1,2}^{+}\mathbf{a}_{i,2}\mathbf{Q}_{m''',n'''} + H.c. \tag{2}$$

This is the exact representation for the Hubbard Hamiltonian with infinite repulsion in cyclic spin permutation formalism[11] which allows constructing its matrix elements in a spin-symmetry-adapted basis by means of branching diagram technique.

For $t_2 = t_3 = 0$ the diamond chain transforms to the set of L non interacting three-site unit cells. These cells have degenerate energy spectrum with respect to the spin value. Let us first consider the fragment of diamond chain formed by two neighbor cells with one and two electrons per cell respectively. In this case, the interaction between cells removes above degeneracy in first PT order in the limit $|t_2|,|t_3| << |t_1|$. Similar to the PT consideration of $U = \infty$ Hubbard ladders,[13] the corresponding effective Hamiltonian for pair of interacting cells can be written in the form

$$\mathbf{H}_1 = \frac{\sqrt{2}\,t_2}{8}\begin{pmatrix}0 & \mathbf{A}\\ \mathbf{A} & 0\end{pmatrix} \tag{3}$$

$$\mathbf{A} = \left(\mathbf{Q}_{1,2} + \mathbf{Q}_{2,3} - 2\mathbf{I}\right)sign\left(t_1\right) + 2\sqrt{2}\alpha\mathbf{I},\ \ \alpha = t_3 / t_2$$

$\mathbf{I}$ is the unit matrix.

Due to simple algebraic structure of (3) it can be shown that the ground state spin of this Hamiltonian changes from $S_0 = 1/2$ to $S_0 = 3/2$ when $\alpha < -\frac{3\sqrt{2}}{8}$ for $t_1 < 0$ and $\alpha > \frac{3\sqrt{2}}{8}$ for $t_1 > 0$. Note also that the exact energy spectrum of the lattice Hamiltonian (1) satisfies the relations

$$\begin{aligned}E\left(t_1,t_2,t_3\right) &= E\left(t_1,-t_2,-t_3\right)\\ &= E\left(-t_1,t_2,-t_3\right)\\ &= E\left(-t_1,-t_2,t_3\right)\end{aligned}$$

Therefore, let us restrict our further consideration by the case of $t_1 < 0$.

The interaction between neighbor cells with equal electron filling is described in second PT order in t_2,t_3. For the filling with one electron per each cell, the corresponding effective low-energy Hamiltonian corresponds to the uniform Heisenberg spin chain

$$\begin{aligned}\mathbf{H}_2 = &\frac{19\sqrt{2}\left(t_2\right)^2 + 18\,t_2t_3}{48\left|t_1\right|}\left(\mathbf{S}_1\mathbf{S}_2 - \frac{1}{4}\right) -\\ &-\frac{\sqrt{2}}{32}\left[8\left(t_2\right)^2 + 8t_2t_3\sqrt{2} + 5\left(t_3\right)^2\right]\end{aligned} \tag{4}$$

The ground state spin of this Hamiltonian is decreased from $S_0 = 1$ to $S_0 = 0$, when $t_3 / t_2 > -\frac{19\sqrt{2}}{18}$.

Let us now consider the diamond chain formed by L unit cells with total number of electrons $N = L + 1$. In the ground state $L - 1$ cells are occupied by single electrons and one cell has two electrons. Electron hops between unit cells with non-equal filling in first PT order leads to the effective movement of two-electron cell state along the chain. At $-\frac{3\sqrt{2}}{8} > \alpha > -\frac{19\sqrt{2}}{18}$ this movement is accompanied by the competition of the interactions between neighbor unit cells with equal and non-equal filling. According to Krivnov and Ovchinnikov[10] this competition leads to the creation of ferromagnetic shell around the cell with two electrons

(magnetic polaron). If this polaron occupies X unit cells, the ground state energy of the chain could be written approximately in the form

$$E_0 = -|t_3|\cos\left(\frac{\pi}{X+1}\right) - R_1(L-X) - R_2 \qquad (5)$$

where

$$R_1 = \frac{19\sqrt{2}\,(t_2)^2 + 18\,t_2 t_3}{48|t_1|}\ln 2$$

$$R_2 = \frac{\sqrt{2}}{32}(L-2)\left[8(t_2)^2 + 8t_2 t_3\sqrt{2} + 5(t_3)^2\right]$$

To minimize this functional, we can evaluate the size of ferromagnetic region and the energy and spin of the lattice ground state. For large values of X such a minimization gives the following result:

$$X = \left(\frac{\pi^2|t_3|}{R_1}\right)^{\frac{1}{3}} \qquad (6)$$

The quantity X determines the ground state spin of the chain S_0 and does not depend on chain size. Therefore the increase of L does not effect on S_0. At $X = L$ the polaron occupies all the chain and its ground state spin takes maximal value.

If $N > L + 1$, there are several unit cells with two electrons. Similar to Reference 10, it can be shown that all these cells form a common ferromagnetic region and there is the critical value of electron density ρ, above which the distorted diamond chain considered above has a ferromagnetic ground state.

$$\rho = 1/3 + \left(\frac{R_1}{9\pi^2|t_3|}\right)^{\frac{1}{3}} \qquad (7)$$

Note, that in case of necklace ladder without frustration ($t_3 = 0$), the magnetic polarons do not appear. At $\alpha \le -\frac{19\sqrt{2}}{18}$ all the interactions between unit cells lead to the ferromagnetic ordering and the ground state spin of distorted diamond chain should take maximal value.

Table 1. Lowest energy lewels of finite fragment of distorted diamond chain. The ground state for each value of α is underlined

$\alpha \setminus S$	0	1	2	3
–0.5	<u>–7.1353</u>	–7.1329	–7.1266	–7.122
–0.6	–7.1273	–7.1298	<u>–7.1307</u>	–7.1302
–0.7	–7.1304	–7.1362	–7.1380	<u>–7.1383</u>

In order to check the accuracy of above consideration we calculated the exact lowest energy states of the Hamiltonian (1) for lattice fragment with $N = 6$, $L = 5$, $t_1 = -1$, $t_2 = 0.1$, $-\alpha = 0.5$–0.7 by Davidson method. The results of the calculations are given in Table 1 and are in agreement with above predictions of polaron approximation.

For second type of the necklace ladder lattice with 'horizontal diagonals' first PT order gives the following effective Hamiltonian for three- electron problem on two neighbor unit cells:

$$\mathbf{H}_1 = -\frac{1}{16}\begin{pmatrix} 0 & \mathbf{A} \\ \mathbf{A}^+ & 0 \end{pmatrix} \qquad (8)$$

$$\mathbf{A} = 2\sqrt{2}\,t_2\left(\mathbf{Q}_{1,2} + \mathbf{Q}_{2,3} - 2\right) + t_3\left(\mathbf{Q}_{1,2} + \mathbf{Q}_{2,3} - \mathbf{I} - \mathbf{Q}_{1,3}\right)$$

It can be shown that the energy state of (8) with $S_0 = 3/2$ has zero energy at arbitrary values of $\boldsymbol{t}_3$. On the other side, simple calculations with trial spin function $\Phi = |\alpha\alpha\beta\rangle$, where α and β are spin variables, which correspond to spin up and down respectively, give non-zero value of average $\langle\mathbf{A}\rangle = -2\sqrt{2}\,t_2$. This means that the ground state of the Hamiltonian (8) corresponds to minimal value of total spin $S_0 = 1/2$ at arbitrary values of $\boldsymbol{t}_3$ in contrast of distorted diamond chain.

The effective two-electron Hamiltonian for two neighbor cells has a form

$$\mathbf{H}_2 = \frac{19\sqrt{2}\,(t_2)^2 + 40\,t_2 t_3 + 2\sqrt{2}\,(t_3)^2}{48|t_1|}\left(\mathbf{S}_1\mathbf{S}_2 - \frac{1}{4}\right) - \frac{\sqrt{2}}{8}\left[2(t_2)^2 + (t_3)^2\right] \qquad (9)$$

The ground state spin for the Hamiltonian (9) equals to $S_0 = 1$ at $-0.7071 > t_3/t_2 > -13.435$. Therefore, similar to our consideration of distorted diamond chain with $N = L + 1$, we may suppose the creation of nonmagnetic polaron in our ladder system. This process destroys the ferromagnetic ordering of the cells occupied by single electrons and decreases the ground state spin of the ladder in contrast to magnetic polaron in distorted diamond chain. Similar to previous consideration we may estimate the size of nonmagnetic polaron

$$X = \left(\frac{2\pi^2|\varepsilon|}{R}\right)^{\frac{1}{3}}, \qquad (10)$$

where ε is the ground state energy of the Hamiltonian (8),

$$R = \frac{19\sqrt{2}\,(t_2)^2 + 40\,t_2 t_3 + 2\sqrt{2}\,(t_3)^2}{48|t_1|}\ln 2$$

This quantity does not depend on the size of the ladder. Therefore the increase of L at fixed values of hopping integrals leads to the increase of the ground state spin of the ladder.

CONCLUSIONS

On the base of perturbation theory and cyclic spin permutation formalism we derived effective Hamiltonians described low-energy states of Hubbard model on two types of distorted necklace ladder formed by N weakly interacting unit cells with additional diagonal interactions and total number of electrons $N = L + 1$. For the lattice of diamond type we demonstrated the appearance of magnetic polarons at negative ratio of hopping parameters describing the interactions of neighbor cells. For second type of the lattice, despite of similar triangular topology, the corresponding polarons have nonmagnetic character.

Acknowledgements. This paper is dedicated to Professor Douglas Klein on his seventieth birthday in gratitude for his valuable and stimulating discussions on the physics of strongly correlated electron systems.

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