Comparison of the Multi-Band Hubbard and Heisenberg Models on Small Finite Clusters

J. Matysiak[*] and R. Lemański

Institute of Low Temperature and Structure Research, Polish Academy of Science,
Okólna 2, 50-422 Wrocław, Poland

We perform exact numeric calculations for a few versions of the two-orbital Hubbard model on the four site cluster. We show that the conventional spin 1 Heisenberg Hamiltonian can be obtained in the strong coupling limit, but only for some versions of the two-orbital Hubbard model. Then we propose a modified version of the Heisenberg Hamiltonian, that we call the two-orbital Heisenberg model, which is relevant in those cases, where the conventional spin 1 Heisenberg model fails.

1. Introduction

It is well known that the spin $1/2$ Heisenberg Hamiltonian can be derived from the single-band Hubbard Hamiltonian at half filling in the large interaction limit [1–6]. However, as far as we know, not much consideration has been given to studies of relationships between multibands Hubbard models and the Heisenberg model with spins higher than $1/2$. This question is relevant for correlated materials with orbital degeneracy, i.e. for most of the transition-metal compounds. In particular, it is important in the theoretical analysis of single molecular magnets (SMMs). Magnetic molecules are usually described by phenomenological spin model Hamiltonians, which contain the Heisenberg term as a dominant one (e.g. [7, 8]). There are also attempts to describe these systems from the first principles using the DFT method and by applying the Falicov-Kimball model [9]. But recently, it has been proposed to describe SMMs using the multi-band Hubbard model (HM) in the large interaction limit combined with DFT calculations [10]. Here we examine this approach by performing exact numerical calculations for the four-site ring with two orbitals per site, so we deal with the two-band HM. Such a small ring size was chosen to allow for performing the exact numeric calculations. Since we deal with small clusters, henceforth we replace the name 'multi-band HM' by 'multi-orbital HM'.

In this paper, we study two versions of the two-orbital HM: the standard HM represented by the Hamiltonian H_{HM}, that contains only the direct Coulomb-type interactions, and the developed HM (DHM) represented by the Hamiltonian H_{DHM}, that includes in addition the exchange interactions (H_{HM} was already considered in [11–13] and H_{DHM} in [14, 15] but for other systems). In fact, we consider two types of H_{DHM} assuming two dif-

ferent possible cases for the hopping amplitudes of electrons. The Hamiltonians H_{HM} and H_{DHM} are as follows:

$$H_{HM} = \sum_{i \neq j, m, m', \sigma} t^{mm'} c_{im\sigma}^{\dagger} c_{jm'\sigma} + U \sum_{im} n_{im\downarrow} n_{im\uparrow}$$

$$+ \tfrac{1}{2} \sum_{i, m \neq m', \sigma} \left[U' n_{im\sigma} n_{im'\bar\sigma} + U'' n_{im\sigma} n_{im'\sigma} \right], \quad (1)$$

$$H_{DHM} = H_{HM} \quad (2)$$

$$+ \tfrac{1}{2} \sum_{i, m \neq m', \sigma} \left[J c_{im\sigma}^{\dagger} c_{im'\bar\sigma}^{\dagger} c_{im\bar\sigma} c_{im'\sigma} + J c_{im\sigma}^{\dagger} c_{im\bar\sigma}^{\dagger} c_{im'\bar\sigma} c_{im'\sigma} \right],$$

where i and j denote nearest-neighbour sites, m, m' label orbitals and $\sigma, \bar\sigma$ label spins of electrons ($\bar\sigma = -\sigma$). U, U' and U'' describe the Coulomb type on-site interactions between two electrons: U - on the same orbital and $U'(U'')$ - on different orbitals with opposite (parallel) spins, respectively. J represents the on-site exchange coupling, but it also enters the interaction constants in H_{HM}. Indeed, we take $U' = U - 2J$, $U'' = U - 3J$, as it was derived in [12]. So the Hamiltonian H_{HM} is not equivalent to H_{DHM} with $J = 0$.

Our aim is to compare the lowest parts of energy spectra of H_{HM} and H_{DHM} with the spectra of effective spin models derived from the perturbation theory. Since for all these models, subspaces with various total spin S are orthogonal to each other, we simplify presentation of our results by considering only states with the total spin $S=0$. However, other subspaces should of course be included if one would want to compare calculated results with experimental ones.

2. The lowest parts of energy spectra

Contrary to the single orbital model, in the multi orbital case, the states with different spin configurations have, in general, different energies even in the limit where $t^{mm'} = 0$ for all hopping constants. Then, in order to obtain the effective Hamiltonian with only spin degrees of freedom, we need to restrict our considerations to a

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[*]corresponding author; e-mail: j.matysiak@int.pan.wroc.pl

subspace with the lowest energy, where all unperturbed states are degenerate.

The number of all states of the cluster with the total spin $S = 0$ is 4900. When all $t^{mm'} = 0$, then energies of these states assume 26 different values both in the case of H_{HM} and H_{DHM}, but they are not the same in the two cases. It also turns out that the degenerations of these states are different in the both cases. Indeed, the ground state degeneracy amounts 6 for H_{HM} and 19 for H_{DHM}.

Figure 1 presents the lowest parts of energy spectra of the two orbital HM in the limit where all hopping constants $t^{mm'} = 0$ (i.e. $t^{11} = t^{22} = t^{12} = t^{21} = 0$), obtained for the four site chain with two orbitals on each site. The parameters $U = 6$ eV, $J = 0.3$ eV are chosen in accordance with the calculations reported in [10].
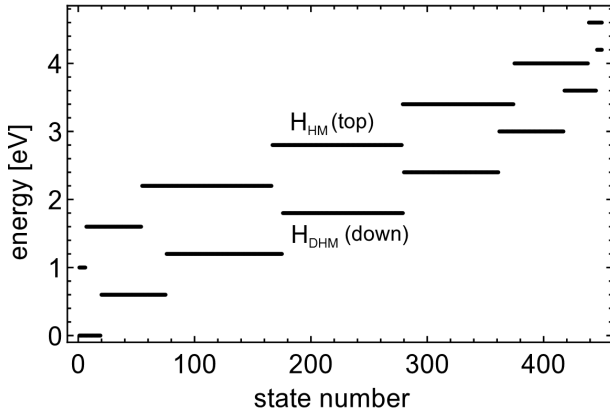

Fig. 1. Comparison of the lowest parts of energy spectra of H_{HM} and H_{DHM} for the subspace with the total spin $S = 0$ obtained in the limit where all hopping constants $t^{mm'} = 0$, $U = 6$ eV and $J = 0.3$ eV. To enable the comparison the spectra are separated (here the spectrum of H_{HM} is moved up by 1 eV).
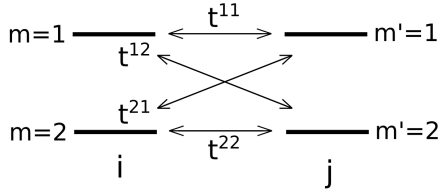

Fig. 2. Labelling of the hopping amplitudes $t^{mm'}$ between the sites i and j in the two orbital HM. m, m' are orbital indices.

In the two orbital HM there are, in general, four independent hopping amplitudes $t^{mm'}$, which are illustrated in Fig. 2. Here we restrict our studies only to the two special cases: a) $t^{11} = t^{22} = t$, $t^{21} = t^{12} = 0$ and b) $t^{11} = t^{22} = t^{21} = t^{12} = t$. For the finite hopping amplitudes $t^{mm'}$, we observed a slight splitting of each degenerate subset of eigenvalues presented in Fig. 1. Obviously, the splittings in the cases a) and b) are different.

Now we would like to focus our attention on the DHM. Our task is to compare the lowest part of energy spectrum of H_{DHM} with the spectrum of the spin-1 Heisenberg Hamiltonian $H_{Heis}^{S=1}$ applied to the ring of four spins $S_i = 1$

$$H_{Heis}^{S=1} = \Gamma \sum_{ij} \mathbf{S_i S_j}, \tag{3}$$

and the lowest part of the spectrum of the multi-orbital Heisenberg Hamiltonian H_{Heis}^{morb}

$$H_{Heis}^{morb} = \sum_{mm'ij} \Gamma_{mm'} s_{im} s_{jm'}, \tag{4}$$

where $\mathbf{S}_i = (S_i^x, S_i^y, S_i^z)$ refers to the ion's spin 1 at the site i and s_{im} refers to the electron's spin $1/2$ on a single orbital m. We point out here that the dimensions of Hilbert spaces of admitted states of the Hamiltonian (4) and (3) are different. Indeed, in our system the number of all admissible states amounts 19 in the case of $H_{Heis}^{S=1}$ and 70 in the case of H_{Heis}^{morb}. However, the Hamiltonian (4) is acting only within the subspace of the lowest energy eigenstates of H_{DHM} (they are degenerated in the atomic limit) that also contains 19 states.

Since in our calculations we put $U = 6$ eV, $J = 0.3$ eV, $t^{mm} = 0.1$ eV, then from the perturbation theory for H_{Heis}^{morb} we get the exchange parameters being approximatively equal to $\Gamma_{mm'} = \dfrac{2(t^{mm'})^2}{U + J} = 0.00318$ eV.
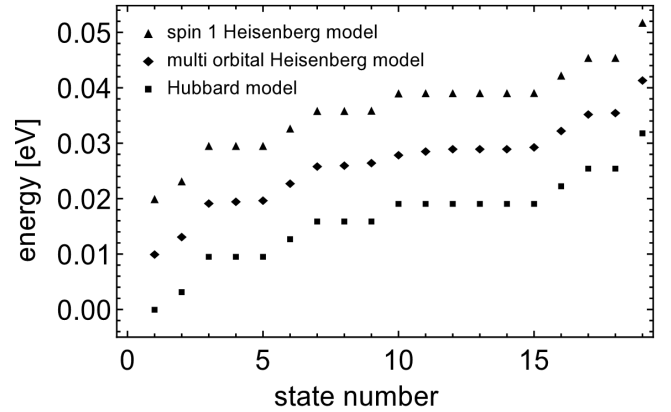

Fig. 3. Comparison of the lowest parts of energy spectra of the models H_{DHM}, $H_{Heis}^{S=1}$ and H_{Heis}^{morb} for $t^{mm} = 0.1$ eV, $t^{mm'} = 0$ for $m \neq m'$, $U = 6$ eV, $J = 0.3$ eV and $\Gamma = 0.00318$ eV. To enable the comparison the spectra are separated from each other by 0.01 eV.

The result of the comparison of the energy spectra is illustrated in Figs. 3 and 4. Figure 3 shows the data for the case a) and Fig. 4 for the case b). In both figures we compare the lowest 19 eigenvalues of the models: H_{DHM}, $H_{Heis}^{S=1}$ and H_{Heis}^{morb}. Based on this comparison we admit that the Hamiltonian $H_{Heis}^{S=1}$ may, indeed, play a role of the effective Hamiltonian when $t^{11}, t^{22} \gg t^{12}, t^{21}$, but it is not the case in the region where $t^{11}, t^{22} \sim t^{12}, t^{21}$. On the other hand, the Hamiltonian H_{Heis}^{morb} seems to be more universal, as it is relevant in the both cases.
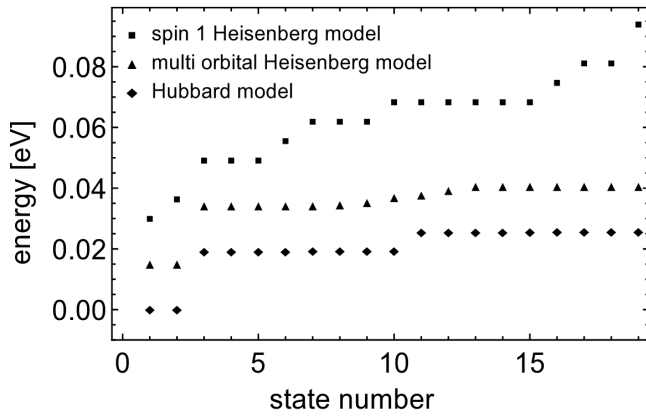
Fig. 4. Comparison of the lowest parts of energy spectra of the models H_{DHM}, $H_{Heis}^{S=1}$ and H_{Heis}^{morb} for $t^{mm'} = 0.1$ eV for all m, m', $U = 6$ eV, $J = 0.3$ eV and $\Gamma = 0.00318$ eV. To enable the comparison the spectra are separated from each other by 0.015 eV.

We also performed calculations for the Hamiltonian H_{HM}, where the exchange interaction terms are omitted [11, 12]. It's overall spectrum is similar to the one obtained for $H_{Heis}^{S=1}$, but their lower parts differ considerably. In particular, the lowest energy subspace now contains only 6 states, as opposed to 19 in the previous case (for the total spin of the whole molecule $S = 0$).

Then, the result of comparison of energy spectra of the Hamiltonians H_{HM}, $H_{Heis}^{S=1}$ and H_{Heis}^{morb} applied to those the lowest 6 energy levels is illustrated in Fig. 5.
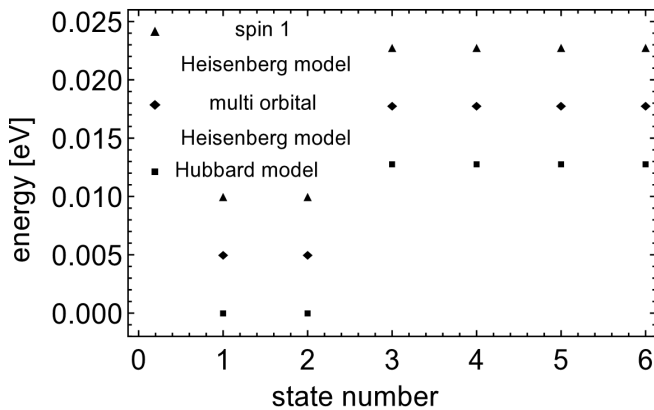

Fig. 5. Comparison of the lowest parts of energy spectra of H_{HM}, $H_{Heis}^{S=1}$ and H_{Heis}^{morb} for $t^{mm} = 0.1$ eV, $t^{mm'} = 0$ for $m \neq m'$, $U = 6$ eV and $J = 0.3$ eV. To enable the comparison the spectra are separated from each other by 0.005 eV.

3. Summary and conclusions

We performed exact numeric calculations for the four site ring, with two orbitals and two electrons per site, using the two-orbital Hubbard Hamiltonians H_{HM} and H_{DHM}. We have found that the energy spectra of the spin 1 Heisenberg model $H_{Heis}^{S=1}$ and the lowest part of the spectra of H_{DHM} are equivalent only when the hopping amplitudes between orbitals of the same type are much larger than between different ones. On the other hand, the multi-orbital Heisenberg model H_{Heis}^{morb} reproduces satisfactory the lowest part of energy spectrum of H_{DHM} both for the case a) $t^{11} = t^{22} = t$, $t^{21} = t^{12} = 0$ and b) $t^{11} = t^{22} = t^{21} = t^{12} = t$.

We also found that the lowest part of energy spectrum of the simplified multi-orbital Hubbard Hamiltonian H_{HM} (without the exchange terms) can also be approximately reproduced by H_{Heis}^{morb}.

So we suggest to use the model H_{Heis}^{morb} in further studies of SMM's materials, as it seems to represent more adequately a wider class of microscopic models of correlated electrons in the large interaction limit than the spin S Heisenberg model H_{Heis}^{S}.

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