Exact Ground States in Spin Systems with Orbital Degeneracy

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(Received)

We present exact ground states in spin models with orbital degeneracy in one and higher dimensions. A method to obtain the exact ground states of the models when the Hamiltonians are composed of the products of two commutable operators is proposed. For the case of the spin-1/2 model with two-fold degeneracy some exact ground states are given, such as the Valence-Bond (VB), the magnetically ordered, and the orbitally ordered states under particular parameter regimes. We also find the models with the higher spin and degeneracy which have the new types of VB ground states in the spin and the orbital sectors.

KEYWORDS: orbital degeneracy, spin systems, valence bond state, orbital order

Quantum spin systems with orbital degeneracy have attracted our theoretical and experimental interests \[\] . These systems have rich phases such as the spin order, the orbital order, the spin liquid, and the orbital liquid. Examples of these systems are the Mott insulator of the transition metal (Mn, Cu, Cr, etc.) oxides, the dynamical Jahn-Teller molecular systems with a phonon coupling (TDAE-C$_{60}$)\[1\] , and the heavy fermion systems in the insulator phase \[2\] . The strong interplay between the orbital and the magnetic ordering is interesting. Other interest is the existence of the spin and the orbital disorder states which are induced by the frustrations with the large quantum fluctuation arising from the spin-orbital interactions. There might already exist a physical realization of a quantum spin-orbital liquid in three dimension: LiNiO$_2$\[3\] . The simplest model of the electron system with orbital degeneracy is the two-fold degenerate Hubbard Hamiltonian with Hund’s rule coupling. In the strong coupling limit, the charge excitation has a large gap at 1/4-filling and this Hamiltonian is an effective model in the insulator phase such as the above spin systems.

In the present letter, we consider the spin-orbital models and give the exact ground states for the spin-1/2 and two-fold degenerate case. We extend the model to more than three-fold degenerate and half and higher spin cases which facilitate the higher dimensional VB states under certain parameter regimes. These VB states are the new types of the VB ground states and kinds of the Resonating Valence Bond (RVB) states.

First, we consider the Hamiltonian is defined by

$$\mathcal{H} = \sum_{i,j} (\mathcal{U}_{ij} - \lambda_0^u)(\mathcal{V}_{ij} - \lambda_0^v),$$  \hspace{1cm} (1)

where the sum runs over all nearest-neighbor bonds \((i,j)\). \(\mathcal{U}_{ij}\) and \(\mathcal{V}_{ij}\) denote some Hermitian local operators whose lowest eigenvalues are given by \(\lambda_0^u\) and \(\lambda_0^v\), respectively. If \(\mathcal{U}_{ij}\) and \(\mathcal{V}_{ij}\) commute with each other: \([\mathcal{U}_{ij}, \mathcal{V}_{ij}] = 0\), the eigenstate of \(\mathcal{H}\) is described by \(|\Psi\rangle = |u\rangle \otimes |v\rangle\), where \(|u(v)\rangle\) denotes a state vector in \(u(v)\) subspace. The local Hamiltonian is positive semi-definite, and hence the total Hamiltonian is also positive semi-definite. The following two types of states are the possible ground states of the Hamiltonian.

Type(i).—One possible ground state is written by

$$|\Psi_G(A, B)\rangle = |\Phi^u(A)\rangle \otimes |\Phi^v(B)\rangle, \quad |\Phi^u(A)\rangle \quad \text{and} \quad |\Phi^v(B)\rangle$$

are given by

$$|\Phi^u(A)\rangle = \prod_{(i,j) \in A} |i, j\rangle_0^u, \quad |\Phi^v(B)\rangle = \prod_{(i,j) \in B} |i, j\rangle_0^v,$$ \hspace{1cm} (2)

where \(|i, j\rangle_0^u = \sum_n C_n^{ij} |\phi_n^{(u)}\rangle_{i,j}\rangle \quad (n = 1, ..., N)\) and \(|\phi_n^{(u)}\rangle_{i,j}\rangle\rangle\rangle\) is the lowest eigenstate of the local operator \(\mathcal{U}(\mathcal{V})\) on sites \(i,j\), whose degeneracy is \(N\). \(A\) and \(B\) denote a bond-covering (a configuration of the bonds) in \(u\) and \(v\) subspaces, respectively, which are constrained by \(A \cup B = \{ \text{all bonds} \}\). One site has \(z_u\) and \(z_v\) bonds on the bond-covering \(A\) and \(B\), respectively, obeying \(z = z_u + z_v\), where \(z\) denotes a lattice coordination number. Obviously, \(\mathcal{H}_G(A, B) = 0\), thus \(|\Psi_G(A, B)\rangle\) is the ground state, because the eigenvalues of the Hamiltonian are positive semi-definite and \(|\Psi_G(A, B)\rangle\) is the lowest eigenstate with an eigenvalue of zero. The ground-state degeneracy depends on the number of the bond-coverings.

Type(ii).—If the state \(|\Phi^u(v)\rangle\) in one sector obeys

$$\mathcal{H}_G|\Phi^u(v)\rangle = \mathcal{H}_0|\Phi^u(v)\rangle = 0, \quad \text{where} \quad |\Phi^u(v)\rangle = \prod_{(ij) \in A} |i, j\rangle_0^u, \quad \text{the ground state of the global Hamiltonian is given by} \quad |\Psi_G\rangle = |\Phi^u(v)\rangle \otimes |\nu(u)\rangle, \quad \text{where} \quad |\nu(u)\rangle \quad \text{denotes an arbitrary state in the v(u) space. Because \(\mathcal{H}\) is positive semi-definite and} \quad |\Psi_G\rangle = 0, \quad \text{and hence} \quad |\Psi_G\rangle$$

is the lowest eigenstate with an eigenvalue of zero. In addition, the ground state of the Hamiltonian \(\mathcal{H} \rightarrow \mathcal{H} + \alpha \mathcal{H}_0\left(\alpha \geq 0\right)\) is given by \(|\Phi^u(v)\rangle \otimes |\Phi^v(u)\rangle\), where \(|\Phi^v(u)\rangle\) is the ground state of the \(\mathcal{H}_{v(u)}\) with the ground-state energy \(E^u_{v(u)}\), since \(\mathcal{H} - \alpha E_0\left(\alpha \geq 0\right)\) is positive semi-definite and \((\mathcal{H} - \alpha E_0^{v(u)})|\Psi_G\rangle = 0\), and hence \(|\Psi_G\rangle\) is the lowest eigenstate with the eigenvalue \(\alpha E^v_{v(u)}\).

Moreover, the ground states of the Hamiltonian: \(\mathcal{H} = \alpha E_{v(u)}\).
and let us set the Hubbard model with Hund’s rule coupling in the strong limit. This point the system has a first-order phase transition. In the limit \( \Delta \to \infty \), the ground state is equivalent to that of the antiferromagnetic (AF) Heisenberg model for \( J_\alpha > -\Delta'/4 \) and of the ferromagnetic Heisenberg model for \( J_\alpha < -\Delta'/4 \). For the ferromagnetic spin states the ground-state energy is given by \( E_0 = -\frac{1}{4} \left[ J_\alpha + J_\gamma + \Delta' \left( \frac{3}{4} - \frac{J_\gamma}{\Delta'} \right) \right] L \) in any dimension. In \( D = 1 \) the AF Heisenberg model is solved by the Bethe Ansatz and has unique, massless singlet ground state. The one-dimensional ground-state energy for \( J_\alpha > -\Delta'/4 \) is given by \( E_0 = -\frac{1}{4} \left[ J_\alpha (4 \ln 2 - 1) + J_\gamma + \Delta' \left( \frac{3}{4} - \frac{J_\gamma}{\Delta'} \right) \right] \). For \( D > 2 \) the existence of an antiferromagnetic long range order (AFLRO) in the AF Heisenberg model has been rigorously proved but in \( D = 2 \) there is no proof of the existence of the AFLRO. Adding an interaction term \( \delta \sum_{(i,j)\in\text{bridges}} S_i^z S_j^z \) to the Hamiltonian at \( J_\alpha = -\frac{\Delta'}{4} \) and \( J_\gamma = \frac{\Delta'}{4} \), the ground states in the spin sector are equivalent to those of the AF Ising model. Thus, the system has the AFLRO on the bipartite lattices in arbitrary dimensions with infinitesimal anisotropy of the \( z \)-direction. On the other hand, at \( J_\alpha = -\frac{\Delta'}{4} \) and \( J_\gamma = \frac{\Delta'}{4} \) the spin liquid states are induced by the infinitesimal perturbations such as an inhomogeneous interaction or a frustration reflecting a lattice topology. For example, let us consider the \( 1/3 \)-depleted square lattice which has the same topology as the CaV\(_4\)O\(_9\) lattice and consists of the four-site squares and the bridge bonds among the squares. If one adds an interaction term \( J_1 \sum_{(i,j)\in\text{squares}} S_i \cdot S_j \) to the Hamiltonian, the exact ground state is a product of the four-spin singlet plaquette RVB (PRVB) states on the squares. On the other hand, adding an interaction term \( J_2 \sum_{(i,j)\in\text{bridges}} S_i \cdot S_j \) to the Hamiltonian, the exact ground state is a product of the singlet dimer states on the bridges. We can exactly construct the PRVB and the dimer states without the next-nearest-neighbor interaction. An orbital long range order (ORLO) makes the systems have the spin liquid state under certain parameter regions.

**Ferromagnetic ground state in the spin sector**— Next we set \( J_{\gamma} = -1 \) and the lowest eigenstates of \( \mathcal{U}_{i,j} \) are triplet whose eigenvalue is \(-\frac{1}{4}\). When the Hamiltonian (3) can be expressed as

\[
\mathcal{H} = \sum_{(i,j)} (\mathcal{U}_{i,j} + \frac{3}{4}) (\mathcal{V}_{i,j} - \frac{\Delta'}{4}) + \alpha \mathbf{S}_i \cdot \mathbf{S}_j + \beta \mathcal{H}_{\text{XXZ}}^{\tau}(\gamma) + \text{constant},
\]

where \( \alpha = J_\alpha + \frac{\Delta'}{4} (\Delta' \leq -1) \), \( \beta = J_\gamma - \frac{1}{2} \geq 0 \), and \( \gamma \leq -1 \) and \( \mathcal{H}_{\text{XXZ}}^{\tau}(\gamma) \) denotes the Hamiltonian of the XXZ model in the pseudo-spin space and is defined by

\[
\mathcal{H}_{\text{XXZ}}^{\tau}(\gamma) = \sum_{(i,j)} (\tau_i^x \tau_j^x + \tau_i^y \tau_j^y + \gamma \tau_i^z \tau_j^z),
\]

namely, for \( J_\gamma \geq \frac{\Delta'}{4} \), \( J_{\sigma} = 1 \), and \( \Delta = \frac{3}{4} \left( \Delta' - \gamma \right) \) \((\Delta' \leq -1)\), the ground state is type (ii) and is a ferromagnetic state in the pseudo-spin space. In the spin space the ground state is equivalent to that of the antiferromagnetic (AF) Heisenberg model for \( J_\alpha > -\Delta'/4 \) and of the ferromagnetic Heisenberg model for \( J_\alpha < -\Delta'/4 \). For the ferromagnetic spin states the ground-state energy is given by \( E_0 = -\frac{1}{4} \left[ J_\alpha + J_\gamma + \Delta' \left( \frac{3}{4} - \frac{J_\gamma}{\Delta'} \right) \right] L \) in any dimension. In \( D = 1 \) the AF Heisenberg model is solved by the Bethe Ansatz and has unique, massless singlet ground state. The one-dimensional ground-state energy for \( J_\alpha > -\Delta'/4 \) is given by \( E_0 = -\frac{1}{4} \left[ J_\alpha (4 \ln 2 - 1) + J_\gamma + \Delta' \left( \frac{3}{4} - \frac{J_\gamma}{\Delta'} \right) \right] \). For \( D > 2 \) the existence of an antiferromagnetic long range order (AFLRO) in the AF Heisenberg model has been rigorously proved but in \( D = 2 \) there is no proof of the existence of the AFLRO. Adding an interaction term \( \delta \sum_{(i,j)\in\text{squares}} S_i^z S_j^z \) to the Hamiltonian at \( J_\alpha = -\frac{\Delta'}{4} \) and \( J_\gamma = \frac{\Delta'}{4} \), the ground states in the spin sector are equivalent to those of the AF Ising model. Thus, the system has the AFLRO on the bipartite lattices in arbitrary dimensions with infinitesimal anisotropy of the \( z \)-direction. On the other hand, at \( J_\alpha = -\frac{\Delta'}{4} \) and \( J_\gamma = \frac{\Delta'}{4} \) the spin liquid states are induced by the infinitesimal perturbations such as an inhomogeneous interaction or a frustration reflecting a lattice topology. For example, let us consider the \( 1/3 \)-depleted square lattice which has the same topology as the CaV\(_4\)O\(_9\) lattice and consists of the four-site squares and the bridge bonds among the squares. If one adds an interaction term \( J_1 \sum_{(i,j)\in\text{squares}} S_i \cdot S_j \) to the Hamiltonian, the exact ground state is a product of the four-spin singlet plaquette RVB (PRVB) states on the squares. On the other hand, adding an interaction term \( J_2 \sum_{(i,j)\in\text{bridges}} S_i \cdot S_j \) to the Hamiltonian, the exact ground state is a product of the singlet dimer states on the bridges. We can exactly construct the PRVB and the dimer states without the next-nearest-neighbor interaction. An orbital long range order (ORLO) makes the systems have the spin liquid state under certain parameter regions.

**Dimerized ground states in one dimension.**—For \( J_\alpha = \frac{\Delta'}{4} + \frac{1}{2} \) (\( \Delta' \geq -1 \)) and \( J_\gamma = \frac{1}{4} \), the Hamiltonian (3) can be written as \( \mathcal{H} = \sum_{(i,j)} \left( \mathcal{U}_{i,j} + \frac{3}{4} \right) \left( \mathcal{V}_{i,j} + \frac{1}{4} \right) + \text{constant} \). The ground states are type (i) and given by

\[
| \Psi_G \rangle = \prod_{i,j} \left[ \text{singlet} \right]_{i,j+1}^\tau \otimes \left[ \text{singlet} \right]_{j,i+1}^\tau,
\]

where \( i \) =even(odd) and \( j \) =odd(even). The ground states are the products of nearest-neighbor singlet dimers and two-fold degenerate. The translational symmetry is spontaneously broken although the interaction is only nearest-neighbor and no bond-alternation. The ground-state energy is \( E_0 = -\frac{3}{4} (\Delta + 2) L \), where \( L \) denotes the number of sites. At \( \Delta = 1 \), Kolezhuk and Mikeska have already given the exact ground states in the ladder model with a leg-leg biquadratic interaction by the matrix product ansatz. In the limit \( \Delta \to \infty \), the lowest states of the local Hamiltonian in the orbital part are two-fold degenerate \((|+\rangle_{j,j+1} \text{ and } |-\rangle_{j,j+1})\). The ground states of the global Hamiltonian in the orbital space are also two-fold degenerate and the orbital order (the Néel order in the pseudo-spin part) exists. At this point the system has a first-order phase transition. In other words, if an infinitesimal xy-element exists in the pseudo-spin space, then the ground states are the singlet, disordered states. The interaction of the transverse direction between spins and orbitals drastically enhances the quantum fluctuation.
ground state is equivalent to that of the XXZ Hamiltonian \( \mathcal{H}_{XXZ}(\gamma) \). On a square lattice the AF XXZ model has the AFLRO for \( \gamma > 1.66 \) that has been proved by Kubo and Kishi and Nishimori et al., using the infrared bounds method. This AFLRO corresponds to an OLRO in the orbital space. The state with the OLRO is what the electrons alternately occupy two orbital levels.

Next, we consider the model with the spin-\( \frac{1}{2} \) and the pseudo-spin-1 in one and higher dimensions described by

\[
\mathcal{H} = \sum_{(i,j)} [S_i \cdot S_j + \alpha] [\tau_i \cdot \tau_j + \beta (\tau_i \cdot \tau_j)^2 + \gamma].
\]

This model is regarded as the effective model with the spin-\( \frac{1}{2} \) for the 3-fold orbital degeneracy. For simplicity, we discuss only the isotropic case.

In one dimension for \( \alpha = 3/4, 0 \leq \beta < 1/3, \) and \( \gamma = 2 - 4\beta \) the exact ground states are type(i) and the singlet dimer states in both the spin and the orbital spaces, because one can write the Hamiltonian as

\[
\mathcal{H} = \sum_{(i,j),n} a_n [S_i \cdot S_j + 3/4] [\tau_i \cdot \tau_j + 2]^n,
\]

where \( a_n \geq 0 \) and \( n > 0 \). These states are two-fold degenerate and the translational symmetry is spontaneously broken. For the case of \( \alpha = 3/4 \) and \( \beta = 1/3, \) the orbital ground state is equivalent to a Valence Bond Solid (VBS) state of the Affleck-Kennedy-Lieb-Tasaki model.\(^{18}\) In the spin sector the ground state is equivalent to that of the AF Heisenberg model for \( \gamma > 2/3 \) and of the ferromagnetic Heisenberg model for \( \gamma < 2/3 \) (type(ii)). At \( \beta = 1/3 \) and \( \gamma = 2/3 \) in the orbital sector the VBS state and the dimerized states are degenerate and in the spin sector all states are degenerate, and hence this point is multi-critical.

At \( (\alpha, \beta, \gamma) = (1 \frac{1}{2}, 1 \frac{1}{2}, 1 \frac{1}{2}) \) the Hamiltonian is reduced to \( \mathcal{H} = 2 \sum_{(i,j)} p_{i,j}^{(S=1)}/p_{i,j}^{(r=2)}. \) Here \( p_{i,j}^{(S=1)} \) and \( p_{i,j}^{(r=2)} \) are the projection operators onto the subspace of the total spin \( S = 1 \) and the total pseudo-spin \( r = 2 \) on the two sites \( i,j \), respectively. In arbitrary bipartite lattices with the coordination number three, e.g., a 2-leg ladder, a hexagonal lattice, a 1/5-depleted square lattice, we can find that the ground states are type(i) and given by

\[
|\Psi_G(A, B)\rangle = |\text{Dimer}\rangle^S \otimes |\text{VB}\rangle^T,
\]

where \( |\text{Dimer}\rangle = \prod_{(i,j) \in A} (a_i^\dagger b_j^\dagger - b_i^\dagger a_j^\dagger)|0\rangle, \)

\[
|\text{VB}\rangle = \prod_{(i,j) \in B} (c_i^\dagger d_j^\dagger - d_i^\dagger c_j^\dagger)|0\rangle,
\]

with the Schwinger boson creation operators that create the spin \( S = 1/2 \) up(down) spin and the \( r = 1/2 (+(-)) \) pseudo-spin at the site \( i \), respectively. A and B denote a configuration of dimers \( (z_s = 1) \) and a configuration of VB's \( (z_r = 2) \) which has two bonds per site, respectively, constrained as \( A \cup B = \{\text{all bonds}\} \) and \( A \cap B = \emptyset \). The ground-state degeneracy is equal to the number of the dimer-coverings. On a ladder and the higher dimensional lattices the number of the bond-coverings is infinity as the lattice size increasing to infinity. These ground states are kinds of the RVB states and the new types of the VB states induced by the interplay between the spin and orbital degrees of freedom. Taking some additional spin interaction terms to the Hamiltonian, one can restrict the number of the degenerate states. The ground state of the Hamiltonian: \( \mathcal{H} \rightarrow \mathcal{H} + x \sum_{(i,j) \in \text{rung}} (p_{i,j}^{(S=1)} - \frac{3}{2}) + y \sum_{(i,j) \in \text{legs}} (p_{i,j}^{(r=2)} - \frac{3}{2}), \)

\( x, y > 0 \) is a product of all rung dimers in the spin sector and two decoupled VBS states in the orbital sector. Therefore, the excitation energy has a finite gap.\(^{26}\) Another Hamiltonian: \( \mathcal{H} \rightarrow \mathcal{H} + x \sum_m \mathcal{H}^{(m)}(x > 0) \) with \( \mathcal{H}^{(m)} \) denotes the Hamiltonian of the Majumder-Ghosh model on the chain \( m \) \( m = 1, 2 \).\(^{26}\) The ground states are four-fold degenerate and have two types. One is a checkerboard-type product of the singlet dimers among the legs in the spin sector and the VBS state connecting a chain in the orbital sector (fig.1(a)). The other is a product of the two neighboring dimers along the legs in the spin sector and a product of the 4-site plaquette VB states in the orbital sector (fig.1(b)). These are the localized states as the dimer, the VB, and the finite VBS states. In the case of above two Hamiltonians, it is expected that each excitation energy has a finite gap.

In general, the model can be easily extended to the higher spin and pseudo-spin cases. We can construct the models with the spin-\( S \) and the pseudo-spin-\( T \) on the lattices whose coordination number is \( z \), which have the following exact VB ground states:

\[
|\Psi_G(A, B)\rangle = |\Phi^A(A)\rangle \otimes |\Phi^T(B)\rangle,
\]

where \( |\Phi^A(A)\rangle = \prod_{(i,j) \in A} (a_i^\dagger b_j^\dagger - b_i^\dagger a_j^\dagger)^M_z|0\rangle, \)

\[
|\Phi^T(B)\rangle = \prod_{(i,j) \in B} (c_i^\dagger d_j^\dagger - d_i^\dagger c_j^\dagger)^M_z|0\rangle.
\]

Here \( M_z \) and \( M_r \) obey \( M_z = 2S/z_s = \text{integer} \) and \( M_r = 2T/z_r = \text{integer} \), respectively, and \( z \) satisfies \( z \leq z_s + z_r \). \( A(B) \) denotes a configuration of the bonds which has \( z_s(z_r) \) bonds per site and \( A \cup B = \{\text{all bonds}\} \). The Hamiltonian with these ground states is given by

\[
\mathcal{H} = \sum_{(i,j)} \sum_{J=0}^{2S} \sum_{I=0}^{2T} K_{J,I} P_{i,j}^{(S=J)} P_{i,j}^{(r=I)},
\]

where \( J_0 = 2S - M_z + 1, I_0 = 2T - M_r + 1 \), and \( K_{J,I} \geq 0 \). The simplest example of the one-dimensional models is given by \( \mathcal{H} = \sum_{(i,j)} [S_i \cdot S_j + S(S+1)] [\tau_i \cdot \tau_j + T(T+1)] \), whose ground states are the singlet dimer states with two-fold degeneracy in both the spin and the pseudo-spin spaces, where \( (z, z_s, z_r) = (2, 1, 1) \). Moreover, one can easily extend to the anisotropic cases. It is natural that the SU(2) symmetry is broken in the orbital sector. The models have the same ground states of the isotropic cases as long as the lowest level of the two-site local Hamiltonian does not cross the higher levels.

In summary, we have proposed a method to obtain the exact ground states of the models which are composed of the products of two commutable operators. Applying this method to the quantum spin model models with the orbital degeneracy as the Hamiltonian (3), we have obtained various ground states of such models and found that these models have the following interesting proper-
ties under particular parameter regimes. The frustration arising from the spin-orbital interaction of the transverse direction increases the quantum fluctuation and induces the singlet dimer ground states. The long range orders in one sector enhance various instabilities in the other sector in any dimension, e.g., the orbital orders make the systems have the spin liquid states and the AFLRO exist with infinitesimal anisotropy of the $z$-direction on the bipartite lattices. In addition, we have constructed models with arbitrary spin and orbital degeneracy which have the exact VB ground states whose degeneracy is infinity in the thermodynamic limit on a ladder and higher dimensional lattices. These VB states include the correlations between the spin and orbital degrees of freedom and are the new types of the RVB states in $D > 1$. The exact ground states are obtained under restrict parameter regions, but we expect that these states are adiabatically connected with the ground states in the large parameter space.

The author would like to thank M. Takahashi, T. Kawarabayashi, M. Nakamura and N. Muramoto for their encouragement and helpful comments.

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Fig. 1. Two types of the VB states on the ladder lattice (see text). Pairs in the spin sector indicate dimer states. Solid lines in the orbital sector denote (a) VBS, (b) finite VBS states.