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Formation of energy gap in high-dimensional spin-orbital liquids

Shun-Qing Shen\textsuperscript{1,2} and Guang-Ming Zhang\textsuperscript{3}

\textsuperscript{1} Department of Physics, the University of Hong Kong
Pokfulam Road, Hong Kong, PRC
\textsuperscript{2} Institute of Physics, Chinese Academy of Sciences
P. O. Box 603, Beijing 100080, PRC
\textsuperscript{3} Center for Advanced Study, Tsinghua University
Beijing 100084, PRC

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Abstract. – A Schwinger boson mean-field theory is developed for spin liquids in a symmetric spin-orbital model in higher dimensions. Spin, orbital and coupled spin-orbital operators are treated equally. We evaluate the dynamic correlation functions and collective excitations spectra. As the collective excitations have a finite energy gap, we conclude that the ground state is a spin-orbital liquid with a two-fold degeneracy, which breaks the discrete spin-orbital symmetry. Based on this conclusion, we propose that broken symmetry or symmetric frustration instead of geometric quantum frustration provides a new route to explore the spin liquid states. Possible relevance of this spin liquid state to several realistic systems, such as CaV$_4$O$_9$ and Na$_2$Sb$_2$Ti$_2$O, are discussed.

The formation of a spin gap in two- or higher-dimensional quantum spin systems is a long-standing issue in strongly correlated problems [1]. Several physical mechanisms were proposed to explain the spin gap in the low-energy excitations. Most of them focus on one-dimensional spin chains and spin ladders. In higher dimensions Anderson proposed that strong quantum fluctuations for spin-(1/2) systems may destroy the antiferromagnetic long-range order in two dimensions, and lead to form a resonating valence bond (RVB) state in which a spin gap may open [2]. However, it becomes true only for some frustrated spin systems such as on the Kagome lattice or in the Majumdar-Ghosh model with a strong next-nearest-neighbor interaction, otherwise there exists antiferromagnetic long-range orders in the ground state on a square and triangle lattice. Recently, it has been realized that orbital degrees of freedom of $d$- and $f$-electrons in transition metal ions provide a new route to explore the formation of spin gap. Several spin-orbital models [3–10] have shown the tendency to form a spin gap in the ground states due to strong orbital and spin quantum fluctuations. Accumulating numerical calculations show that spin liquid state may be formed in some one-dimensional spin-orbital coupled systems. Behaviors in higher-dimensional systems are relatively less clear. Experimentally, several higher-dimensional spin gap materials, such as Na$_2$Ti$_2$Sb$_2$O [11] and ...
NaV$_4$O$_9$ [12], are synthesized, in which the orbital degree of freedom might play a key role in the formation of the spin gap.

The spin-orbital model Hamiltonian is written as [13,14]

\[
H = J \sum_{\langle ij \rangle} \left( 2 \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2} \right) \left( 2 \tau_i \cdot \tau_j + \frac{1}{2} \right) + J_s \sum_{\langle ij \rangle} \left( 2 \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2} \right) + J_\tau \sum_{\langle ij \rangle} \left( 2 \tau_i \cdot \tau_j + \frac{1}{2} \right). \tag{1}
\]

This model may be derived from an electronic model with double orbital degeneracy. This model possesses an $SU(2) \otimes SU(2)$ symmetry. When $J_s = J_\tau = J_0$, the model has an additional permutation symmetry between spin and orbital. In this case the model can be written as a combination of two symmetric models

\[
H = (J + J_0) \sum_{\langle ij \rangle} \left( 2 \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2} \right) \left( 2 \tau_i \cdot \tau_j + \frac{1}{2} \right) - J_0 \sum_{\langle ij \rangle} \left( 2 \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{2} \right) \left( 2 \tau_i \cdot \tau_j - \frac{1}{2} \right). \tag{2}
\]

The former part is the standard $SU(4)$ spin-orbital model, which is solvable in one-dimension and has been investigated intensively [6]. The second part is the model first proposed by Santoro [3], and also possesses the $SU(4)$ symmetry with different generators on a bipartite lattice. However, the combination of the two models breaks the $SU(4)$ symmetry. From previous studies, it has been found that the interplay between spin and orbital degrees of freedom produces either quantum ordered or disordered phases. Spin liquid states with an energy gap were found in one dimension [3,4,8,10]. To investigate the model systematically, we try to develop a theory, which can describe the disordered state with an energy gap as well as the ordered states. The Schwinger boson theory is an ideal candidate. The theory was first used to the spin $SU(2)$ Heisenberg model [15], and was generalized to study the symmetric spin-orbital model at $J = -J_0$ by the present authors [10]. The advantage of this theory is that it can describe either quantum ordered or disordered states. The results for the two- and three-dimensional Heisenberg model are fully consistent with the spin wave theory. Here we present the Schwinger boson mean-field theory for this spin-orbital system.

For the present model, there are four possible states on each site $i$ according to the eigenvalues of $\mathbf{S}_i^2$ and $\tau_i^2$: $|1\rangle = | + 1/2, +1/2 \rangle$, $|2\rangle = | - 1/2, +1/2 \rangle$, $|3\rangle = | + 1/2, -1/2 \rangle$, $|4\rangle = | - 1/2, -1/2 \rangle$. We introduce four Schwinger bosons to describe these four states: $|\mu\rangle = a_{i\mu}^\dagger |0\rangle$, where $|0\rangle$ is the vacuum states and $\mu = 1, 2, 3, 4$. There is a local constraint for the four bosons, $\sum_{\mu=1}^{4} a_{i\mu}^\dagger a_{i\mu} = 1$, for each site. On these bases the spin and orbital operators can be expressed in terms of these four Schwinger bosons:

\[
\begin{align*}
S_i^+ &= a_{i1}^\dagger a_{i2} + a_{i3}^\dagger a_{i4}, & \tau_i^+ &= a_{i1}^\dagger a_{i3} + a_{i2}^\dagger a_{i4}, \\
S_i^- &= a_{i2}^\dagger a_{i1} + a_{i4}^\dagger a_{i3}, & \tau_i^- &= a_{i3}^\dagger a_{i1} + a_{i4}^\dagger a_{i2}, \\
S_i^z &= \frac{1}{2}(a_{i1}^\dagger a_{i1} - a_{i2}^\dagger a_{i2} + a_{i3}^\dagger a_{i3} - a_{i4}^\dagger a_{i4}), & \tau_i^z &= \frac{1}{2}(a_{i1}^\dagger a_{i1} - a_{i3}^\dagger a_{i3} + a_{i2}^\dagger a_{i2} - a_{i4}^\dagger a_{i4}).
\end{align*}
\]
Thus, the Hamiltonian is rewritten in terms of the Schwinger bosons:

\[
H = -\frac{J + J_0}{2} \sum_{\langle ij \rangle, \mu \nu} A_{ij, \mu \nu}^\dagger A_{ij, \mu \nu} - J_0 \sum_{\langle ij \rangle, \mu \nu} (B_{ij,14}^\dagger - B_{ij,23}^\dagger)(B_{ij,14} - B_{ij,23}) + \\
+ \sum_i \lambda_i \left( \sum_{\mu = 1}^4 a_{i\mu}^\dagger a_{i\mu} - 1 \right) + \frac{1}{2} z N_A (J + 2 J_0),
\]

where \(A_{ij, \mu \nu} = a_{i\mu} a_{j\nu} - a_{i\nu} a_{j\mu}\) and \(B_{ij, \mu \nu} = a_{i\mu} a_{j\nu} + a_{i\nu} a_{j\mu}\). We have introduced antisymmetric and symmetric operators \(A\) and \(B\) for the purpose of the mean-field calculations. The present theory is limited to the case \(J_0 \geq 0\) and \(J + J_0 \geq 0\). The local Lagrangian multipliers are introduced to realize the local constraint for hard-core bosons. In the mean-field approach we shall take them as site-independent \(\lambda_i \equiv \lambda\). The thermodynamic averages of the operators \(A\) and \(B\) are introduced as the order parameters, respectively,

\[
\langle A_{ij, \mu \nu} \rangle \equiv -2i \Delta_{ij, \mu \nu}^o (r_i - r_j), \quad \langle B_{ij, \mu \nu} \rangle \equiv 2 \Delta_{ij, \mu \nu}^e (r_i - r_j).
\]

\(\Delta_{ij, \mu \nu}^o (r_i - r_j)\) and \(\Delta_{ij, \mu \nu}^e (r_i - r_j)\) are odd and even functions with respect to the indices \(\mu, \nu\) or the sites \(r_i, r_j\). In the momentum space, we take

\[
\frac{i}{Z} \sum_{\delta} \Delta_{\mu \nu}^o (\delta) e^{-ik \cdot \delta} = \frac{\Delta_{\mu \nu}^o}{d} \sum_\alpha \sin k_\alpha = \Delta_{\mu \nu}^o \gamma_s (k),
\]

\[
\frac{1}{Z} \sum_{\delta} \Delta_{\mu \nu}^e (\delta) e^{-ik \cdot \delta} = \frac{\Delta_{\mu \nu}^e}{d} \sum_\alpha \cos k_\alpha = \Delta_{\mu \nu}^e \gamma_c (k),
\]

where \(\delta\) points to the nearest-neighbor sites. By utilizing the Pauli matrices \(\sigma_\alpha\) (\(\alpha = x, y, z\)) and the \(2 \times 2\) identity matrix \(\sigma_0\), the mean-field Hamiltonian can be expressed in a compact form of \(8 \times 8\) matrix,

\[
H = \frac{1}{2} \sum_k \Phi_k^\dagger H(k) \Phi_k + \mathcal{E}_0,
\]

where

\[
\Phi_k^\dagger = (a_{k1}^\dagger, a_{k2}^\dagger, a_{k3}^\dagger, a_{k4}^\dagger, a_{-k1}, a_{-k2}, a_{-k3}, a_{-k4}),
\]

\[
H(k) = \lambda \sigma_0 \otimes \sigma_0 \otimes \sigma_0 - i \sigma_y \otimes A(k) + b(k) \sigma_x \otimes \sigma_y \otimes \sigma_y,
\]

\[
A(k) = -2z(J + J_0) \gamma_s (k) \begin{pmatrix} 0 & \Delta_{12}^o & \Delta_{13}^o & \Delta_{14}^o \\ -\Delta_{12}^o & 0 & \Delta_{23}^o & \Delta_{24}^o \\ -\Delta_{13}^o & -\Delta_{23}^o & 0 & \Delta_{34}^o \\ -\Delta_{14}^o & -\Delta_{24}^o & -\Delta_{34}^o & 0 \end{pmatrix},
\]

\[
\frac{\mathcal{E}_0}{N_A} = +z(J + J_0) \sum_{\mu \nu} \Delta_{\mu \nu}^2 + 2z J_0 (\Delta_{14}^e - \Delta_{23}^e)^2 - 3\lambda + \frac{1}{2} z (J + 2 J_0),
\]

where \(b(k) = -2z J_0 (\Delta_{14}^e - \Delta_{23}^e) \gamma_c (k)\). The Kronecker product for block matrices is used [16]. The Hamiltonian can thus be diagonalized analytically, and the free energy is evaluated to
establish the mean-field equations. Due to the symmetry in the Hamiltonian, there exist two sets of solutions: I) \( \Delta_{14}^1 - \Delta_{23}^0 = \Delta_2, \Delta_{12}^0 = \Delta_{34}^0 = \Delta, \Delta_{13}^0 = -\Delta_{24}^0, \Delta_{14}^0 = \Delta_{23}^0; \) with \( \Delta_1 = \sqrt{\Delta_{13}^2 + \Delta_{14}^2}; \) II) \( \Delta_{14}^1 - \Delta_{23}^0 = \Delta_2, \Delta_{12}^0 = -\Delta_{34}^0, \Delta_{13}^0 = \Delta_{24}^0, \Delta_{14}^0 = -\Delta_{23}^0 = \Delta, \) with \( \Delta_1 = \sqrt{\Delta_{12}^2 + \Delta_{13}^2}. \) With the notation \( \Delta, \Delta_1, \) and \( \Delta_2, \) the same branches of spectra are given by \( \omega(k)_{\pm} = \sqrt{\lambda^2 - a^2(k) - b_{\pm}^2(k)}, \) where \( a(k) = -2z(J + J_0)\Delta\gamma_s(k) \) and \( b_{\pm}(k) = -2z(J + J_0)\Delta_1\gamma_s(k) \pm 2zJ_0\Delta_2\gamma_c(k). \) Moreover, these two ground states are degenerated. The degeneracy may originate from the symmetry of permutation of spin and orbital operators.

When \( J_0 = 0, \) the model is reduced to the standard \( SU(4) \) spin-orbital model. The two spectra become degenerated, \( \omega(k) = \sqrt{\lambda^2 - [2z(J + J_0)\gamma_s(k)]^2(\Delta_{12}^2 + \Delta_{13}^2 + \Delta_{14}^2)}. \) In this way we recover the spectra for the \( SU(N = 4) \) model.

In the following we will focus on the ground state with an energy gap, i.e., \( \min(\omega(k)) \neq 0. \) The free energy per site is given by \( E = \int \frac{dk}{(2\pi)^d} (\omega_+(k) + \omega_-(k))/2 + E_0/N_{\lambda}. \) To determine the order parameters, we introduce a set of dimensionless parameters \( \Delta = 2z(J + J_0)\Delta/\lambda, \Delta_1 = 2z(J + J_0)\Delta_1/\lambda, \Delta_2 = 2zJ_0\Delta_2/\lambda, \) and \( \lambda = \lambda/[z(J + J_0)], \) then the dimensionless quasiparticle excitation spectrum becomes

\[
\bar{\omega}_\pm(k) = \sqrt{1 - \bar{\Delta}^2\gamma_s^2(k) - [\bar{\Delta}_1\gamma_s(k) \pm \bar{\Delta}_2\gamma_c(k)]^2}.
\]  

The self-consistent mean-field equations are established by minimizing the free energy,

\[
\int \frac{dk}{(2\pi)^d} \left[ \frac{1}{\bar{\omega}_+(k)} + \frac{1}{\bar{\omega}_-(k)} \right] = 3, \tag{5a}
\]

\[
\int \frac{dk}{(2\pi)^d} \left[ \frac{\gamma_s^2(k)}{\bar{\omega}_+(k)} + \frac{\gamma_s^2(k)}{\bar{\omega}_-(k)} \right] = 2\bar{\lambda}, \tag{5b}
\]

\[
\int \frac{dk}{(2\pi)^d} \left[ \frac{(\bar{\Delta}_1\gamma_s(k) + \bar{\Delta}_2\gamma_c(k))\gamma_s(k)}{\bar{\omega}_+(k)} + \frac{(\bar{\Delta}_1\gamma_s(k) - \bar{\Delta}_2\gamma_c(k))\gamma_s(k)}{\bar{\omega}_-(k)} \right] = 2\bar{\lambda}\bar{\Delta}_1, \tag{5c}
\]

\[
\int \frac{dk}{(2\pi)^d} \left[ \frac{(\bar{\Delta}_1\gamma_s(k) + \bar{\Delta}_2\gamma_c(k))\gamma_c(k)}{\bar{\omega}_+(k)} + \frac{(\bar{\Delta}_1\gamma_s(k) - \bar{\Delta}_2\gamma_c(k))\gamma_c(k)}{\bar{\omega}_-(k)} \right] = \bar{\lambda}\bar{\Delta}_2 \left( 1 + \frac{J}{J_0} \right). \tag{5d}
\]

Substituting eq. (5b) into eq. (5c), we have

\[
\int \frac{dk}{(2\pi)^d} \left[ \frac{1}{\bar{\omega}_+(k)} - \frac{1}{\bar{\omega}_-(k)} \right] \bar{\Delta}_2\gamma_c(k)\gamma_s(k) = 0.
\]

If \( \bar{\Delta}_2 \neq 0, \bar{\Delta}_1 \) must be equal to zero. Oppositely, if \( \bar{\Delta}_2 = 0, \) the solution is for the case of \( J_0 = 0. \) Therefore for \( J_0 > 0, \) the solution is \( \bar{\Delta}_1 = 0, \) with \( \bar{\omega}_+(k) = \bar{\omega}_-(k) \equiv \bar{\omega}(k). \) The two spectra are also degenerated. Two sets of saddle point solutions correspond, corresponding to the spin liquid phase with an energy gap in elementary excitations: I) \( \Delta_{14}^c - \Delta_{23}^c = \Delta_2, \Delta_{12}^c = \Delta_{34}^c = \Delta, \) and \( \Delta_{13}^c = -\Delta_{24}^c = \Delta, \) and \( \Delta_{14}^c = 0 \) otherwise. II) \( \Delta_{14}^c - \Delta_{23}^c = \Delta_2, \Delta_{12}^c = -\Delta_{34}^c = \Delta, \) and \( \Delta_{13}^c = \Delta_{24}^c = 0 \) otherwise. We focus on the first set of solutions and then present the results for the second set of solutions. For a given value of \( J/J_0, \) we have a set of solutions for \( \bar{\lambda}, \bar{\Delta}_1, \bar{\Delta}_2. \)

To determine the physical properties of the state, we define the one-particle Green’s function in an \( 8 \times 8 \) matrix form as

\[
G(k, t) = -i(0|T\{\Phi_k(t)\Phi_k^\dagger(0)\}|0),
\]
where $|0\rangle$ is the ground state of the Hamiltonian. Its Fourier transform is given by

$$G(k, \omega) = ((\omega + i\delta)\sigma_z \otimes \sigma_0 \otimes \sigma_0 - H_k)^{-1}$$

$$= \frac{1}{\omega^2 - \lambda^2 + a^2(k) + b^2(k) + i\delta} \times \left\{ \omega\sigma_z \otimes \sigma_0 \otimes \sigma_0 - \lambda\sigma_0 \otimes \sigma_0 \otimes \sigma_0 + a(k)\sigma_y \otimes \sigma_0 \otimes \sigma_y - b(k)\sigma_x \otimes \sigma_y \otimes \sigma_y \right\},$$

with $a(k) = -2z(J + J_0)\Delta\gamma_s(k)$ and $b(k) = -2zJ_0\Delta\gamma_c(k)$. From these Green functions, we evaluate the dynamic correlation functions for the spin $S_i^z$, orbital $T_i^z$, and spin-orbital density operators $2S_i^zT_i^z$. After some algebra, we have

$$\chi_X(q, \Omega + i\delta) = \frac{1}{8} \int \frac{dk}{(2\pi)^d} \left( \frac{C_X(k, q)}{\omega(k)\omega(k + q) - 1} - 1 \right) \times$$

$$\times \left[ \frac{1}{\Omega + i\delta + \omega(k) + \omega(k + q) - \Omega + i\delta - \omega(k) - \omega(k + q)} \right],$$

with $(X = S, T, ST)$

$$C_S(k, q) = \lambda^2 - a(k)a(k + q) - b(k)b(k + q),$$

$$C_T(k, q) = \lambda^2 + a(k)a(k + q) - b(k)b(k + q),$$

$$C_{ST}(k, q) = \lambda^2 - a(k)a(k + q) + b(k)b(k + q).$$

If the minimum of $\omega(k)$ is non-zero, $\text{Im}[\chi_X(q, \Omega)]$ become non-zero only when $\Omega \geq 2\min(\omega(k))$. Thus the three collective excitations for the density-density correlation function have a finite energy gap, $\Delta_{\text{gap}} = 2\min(\omega(k))$. It is worth mentioning that the solution has broken the discrete permutation symmetry of spin and orbital. This can be seen from the fact that, in general, $\chi_S(q, \Omega) \neq \chi_T(q, \Omega)$. The same expressions are obtained for the second set of solutions if we permute the indices $S$ and $T$. The spectra and free energy as well as the energy gap are identical to the first set of solutions. Thus the two sets of solutions are energetically degenerated. The symmetries in the two states are different. More important, the double degeneracy of the ground state was also observed in one dimension in other approaches [4,17]. Therefore, this two-fold degeneracy is not a consequence of the mean-field approach, and can be regarded as an evidence to support our mean-field theory.

Now we come to evaluate the energy gap by solving the mean-field equations. On a one-dimensional chain, the energy gap can be evaluated analytically by introducing a parameter $x_0 = (\Delta^2 - \Delta_0^2)/(1 - \Delta_0^2)$. The energy gap and the ratio of $J_0/J$ are

$$\Delta_{\text{gap}} = 4z(J + J_0) \times \left\{ \frac{K(x_0) - E(x_0)}{\pi x_0}, \quad \text{if} \quad x_0 < 0, \right\}$$

$$\sqrt{1 - x_0} \frac{K(x_0) - E(x_0)}{\pi x_0}, \quad \text{if} \quad x_0 \geq 0,$$

$$\frac{J}{J_0} = -1 + 2 \frac{K(x_0) - E(x_0)}{E(x_0) - (1 - x_0)K(x_0)},$$

where $K(x)$ is the complete elliptic integral of the second kind and $E(x)$ is the complete elliptic integral of the first kind. We have established a one-to-one correspondence between
Shun-Qing Shen et al.: Formation of energy gap in high-dimensional etc.

Fig. 1 – The energy gap \(2z(J + J_0)\) via the ratio of \(J_0/J\) in two and three dimensions.

the ratio \(J_0/J\) and the energy gap. We find that there is a turning point at \(J_0/J = 1\). The theory fails at the symmetric point \(J_0 = 0\). The energy gap still opens at that point, which is in conflict with the solution of Bethe ansatz [18]. The same problem was encountered in the spin-(1/2) \(SU(2)\) theory in one dimension.

The mean-field equations for two-dimensional square lattice and three-dimensional cubic lattice are solved numerically. The energy gaps for two- and three-dimensional lattices are plotted in fig. 1. We find that the energy gap opens in the regime of \(0.380 < J_0/J < 9.84\) for \(d = 2\) and of \(0.666 < J_0/J < 1.667\) for \(d = 3\). The gap closes at two critical ratios \(J_0/J\). From fig. 1, it is shown that the energy gap appears in a larger parameter range in two dimensions than in three dimensions. This is consistent with the fact that the quantum fluctuations are stronger in two dimensions. Out of the above parameter regimes the Bose condensations have to be considered, otherwise the mean-field equations have no solutions. The Bose condensation of Schwinger bosons is characteristic of the magnetic or orbital long-range orders, as we discussed in the symmetric point \(J = -J_0\) [10]. At the point of \(J = -J_0\), the ground state possesses ferromagnetic coupled spin-orbital long-range order as well as the conventional spin and orbital long-range orders. At another point of \(J_0 = 0\), the ground state possesses antiferromagnetic coupled spin-orbital long-range order. A detailed discussion on these phases will be presented elsewhere. As far as we know, the Schwinger boson mean-field theory is very successful for the spin liquid state for \(s = 1\) in one dimension, and anti- and ferromagnetic states for higher dimensions. Our theory shows that it also works very well for spin-orbital liquid states in higher dimensions.

Physically, it is broken symmetry or symmetric frustration which drives the system to a spin-orbital liquid. Traditionally, we explore the spin liquids based on geometric quantum frustration as proposed by Anderson [2]. The idea was realized in some strongly frustrated systems. In the present theory, the couplings are not frustrated. The spin-orbital liquids arise in the regime which deviates from two high symmetric points, which do not break the spin \(SU(2)\) symmetry. The term in eq. (1) which breaks the high symmetry forces the system to destroy the ordered states to enter frustrated liquid states. This contains a new physics: \textit{symmetric frustration provides a new possible route to explore the spin liquid}. Some higher-spin systems break the high symmetry and may have liquid states [19]. The formation of a spin energy gap indicates that the ground state is a spin-orbital liquid. Experimentally the energy gap can be measured from magnetic susceptibility. There are several higher-dimensional materials such as \(Na_2Ti_2Sb_2O\) and \(CaV_4O_9\) [12]. It is believed that the orbital degrees of freedom play an important role in the formation of spin gap [4, 5]. The low-temperature
phases of these materials may be relevant to the spin-orbital liquid with the energy gap as we discuss in this paper.

In conclusion, we develop a Schwinger boson mean-field theory for spin gap in any dimensions. The ground state of the model is a spin-orbital liquid with an energy gap in an extensive parameter regime. This ground state breaks the discrete symmetry of permutation of spin and orbital, and is doubly degenerated. The broken high symmetry plays a key role in the formation of the spin gap.

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