Antiferromagnetic correlations of the resonating-valence-bond state

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We consider a trial wave function for the two-dimensional spin-$\frac{1}{2}$ antiferromagnetic Heisenberg model (resonating-valence-bond state), which is a sum of all dimerizations. It is shown that the antiferromagnetic correlation length is bounded by that of a classical gas of interacting loops with $n=4$ components. We argue that the range of the antiferromagnetic correlations is finite and triplet excitation has an energy gap.

One of the unsolved problems in condensed-matter physics is the ground-state properties of the spin-$\frac{1}{2}$ antiferromagnetic Heisenberg model in two dimensions whose Hamiltonian is

$$H=\sum_{ij} J_{ij}\sigma_i\cdot\sigma_j \ , \tag{1}$$

where $J_{ij}>0$ is the coupling constant which is usually assigned a nonzero constant value J for nearest neighbors and is zero otherwise. For the one-dimensional case, the exact solution is available[1,2] and the ground state does not have long-range order. On the other hand, the system with three-dimensional lattices and spins greater than $\frac{1}{2}$ is known experimentally and theoretically to have a ground state which is close to the Néel antiferromagnetic state.

There is a recent resurgence of interest in the two-dimensional model due to the discovery of the high-T_c oxide superconductors. The importance of the two-dimensionality of the materials is emphasized for the novel behavior. A simple model for the strongly correlated electrons is the Hubbard model. For the half-filled case (one electron per site), the Hubbard model can be mapped to the antiferromagnetic Heisenberg model in the limit of strong electron-electron repulsion (large-U limit).

Anderson and co-workers[3] have proposed the resonating valence bond (RVB) state as a candidate for explaining the new mechanism of the unusual high-T_c superconductors. The RVB state is based on singlet pairings of spins and does not seem to have long-range order. Also, the term RVB is rather loosely defined and the meaning varies a lot from one work to another.

Many of the researchers, on the other hand, seem to support the existence of antiferromagnetic long-range order, hence the Néel-type state based on the numerical data for the Heisenberg model or the Hubbard model.[4-6] One of the difficulties in deducing the physical results from numerical data is that the numerical methods (exact diagonalization, quantum Monte Carlo, and variational) allow investigation of systems of rather modest sizes.

Recent neutron scattering measurements[7] and magnetic susceptibility measurements[8] on a single crystal of undoped La_2CuO_4 seem to show the existence of unusual spin state (quantum spin fluid) which might play a crucial role in the mechanism of superconductivity.

It is important to understand the nature of the correlation in the RVB state, especially since one may be able to assess the validity of the RVB picture for the high-T_c superconductor by a direct comparison with experiments. We consider a square lattice and the nearest-neighbor coupling of Eq. (1). The version of the RVB state discussed in this paper has singlet couplings on only nearest-neighbor sites and we call this the nearest-neighbor resonating valence bond (NNRVB) state. First consider a state (which certainly is not an eigenstate) corresponding to a dimer covering of a square lattice (see Fig. 1). Here a dimer represents a singlet coupling of spins in neighboring sites, i.e.,

$$\phi_{ij}=(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle) \ , \tag{2}$$

with an obvious notation for the spin states of i and j sites. There are many dimer coverings whose entropy is exactly known[9] and each covering gives a state which is a product of singlet bonds of type (2). The trial energy for this wave function is the singlet energy $\varepsilon_s(=-3J/4)$ per dimer and thus gives $-3J/8$ per site. This is rather poor value for a trial energy since the Néel state without quantum correction has an energy $-J/2$ per site. It is possible, however, to improve the trial energy by a superposition of states with different dimer configurations. Let us write ψ_α and ψ_β for states with different dimer configurations (see Fig.

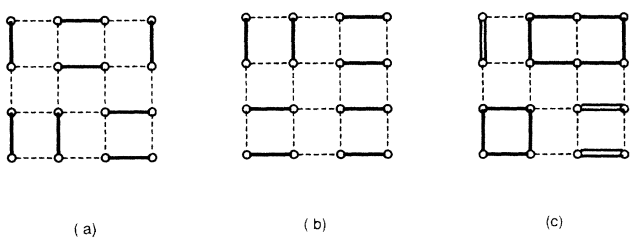

FIG. 1. (a) and (b) Examples of dimer covering; (c) the overlap of dimer coverings (a) and (b).

1). These states are not orthogonal, i.e., $\langle\psi_\alpha|\psi_\beta\rangle\neq 0$ and the overlap is graphically considered as a covering of the square lattice with double dimers and self-avoiding closed loops.[10] This can be seen by noticing that a lattice site is always connected to two dimers which belong to state α and state β. Therefore a loop cannot have end points, and we have only closed loops or double dimers. Since every site must belong to a closed loop or a double dimer the lattice is covered with these objects. Sutherland[10] obtained a rule for estimating $\langle\psi_\beta|H|\psi_\alpha\rangle$. This can be written as a sum of contributions from each closed loop. A contribution from a closed loop in unit of the singlet energy ($\varepsilon_s=-3J/4$) is the length of the loop plus the number of nearest-neighbor bonds which are not on the loop, but still connect two sites on the loop. The quantity $\langle\psi_\beta|H|\psi_\alpha\rangle$ is lower than $\langle\psi_\alpha|H|\psi_\alpha\rangle=\langle\psi_\beta|H|\psi_\beta\rangle$ since the diagonal overlap has the singlets at only half of the bonds. One can improve the variational energy by having the maximum number of cross terms. Thus our trial wave function is $\Psi=\sum_\alpha\psi_\alpha$ where the summation is over all the dimer configurations. Note that we choose the coefficients of the superposition to be simply unity for all α's. We call this the NNRVB state. In fact, the trial energy is compared to the exact numerical estimate of the ground-state energy of a 4×4 lattice of Oitmaa and Betts[4] and the difference is only about 4.7%.[11,12] Although the simple linear combination (all coefficients are the same) seems reasonable, it is, however, far from obvious how this is close to optimal. Note that we also must take into account the normalization $\langle\Psi|\Psi\rangle$ for the energy estimate.

The staggered correlation function is

$$G(\mathbf{r})=\frac{4(-1)^r\langle\Psi|\sigma_z(0)\sigma_z(\mathbf{r})|\Psi\rangle}{\langle\Psi|\Psi\rangle}\ , \tag{3}$$

where $(-1)^r$ is 1 or -1 depending on whether 0 and $\mathbf{r}$ are in the same sublattice of the bipartite square lattice. The integral $\langle\Psi|\Psi\rangle$ is a sum of terms involving $\langle\psi_\beta|\psi_\alpha\rangle$ for various α and β. This overlap integral is graphically represented by a covering of the square lattice by loops and (double) dimers [see Fig. 1(c)]. Each loop or dimer carries a weight of two from the two ways of assigning spins antiferromagnetically on the sites on a loop or a dimer. However, a loop carries an extra weight of two (four in total). There is an extra multiplicative factor of 2 due to two ways to have a loop with an assigned spin orientation. In a loop, color every other bond red, and all the others black. One way to have the loop is that the red bonds come from a bra state and the black bonds from a ket state. Another way is obviously obtained by exchanging the red and the black bonds. Note that a dimer does not have this extra factor of 2. Thus we have

$$\langle\Psi|\Psi\rangle=\sum_\Gamma x^{p_2}y^p\ , \tag{4}$$

where $x=2$, $y=4$, p_2 is the number of dimers and p is the number of loops in a covering Γ. This expression can be thought as a partition function of a statistical mechanical model, and related statistical models for different values of x and y are discussed by Sutherland.[13]

The correlation function (3) is written as

$$G(\mathbf{r})=\sum_\Gamma\chi(\mathbf{r})x^{p_2}y^p\Big/\sum_\Gamma x^{p_2}y^p\ , \tag{5}$$

where

$$\chi(\mathbf{r})=\begin{cases}1 & \text{for 0 and } \mathbf{r} \text{ are on the same loop,}\\ 0 & \text{otherwise.}\end{cases} \tag{6}$$

First it is obvious that we have a constant contribution (1) when the two points are on the same loop since the spins are antiferromagnetically ordered in a loop. There is no contribution to the correlation function if the two spins belong to different loops. Each loop actually represents two spin orientations and the orientation is not correlated for different loops. Thus, they cancel out.

Denote the total length of all the loops as L, then

$$2p_2+L=N\ , \tag{7}$$

where N is the number of sites, and (5) can be written as

$$G(\mathbf{r})=\sum_{\text{loop}}\chi(\mathbf{r})x^{-L/2}y^P d(p_2)\Big/\sum_{\text{loop}}x^{-L/2}y^P d(p_2)\ , \tag{8}$$

where the summations are only for loop configurations and $d(p_2)$ is the number of dimer configurations for a given loop configuration.

Let us explore first the situation in which $d(p_2)$ is independent of the loop configurations. In that case $G(\mathbf{r})$ will simply be the probability for the two sites to belong to the same loop (proportional to energy-energy correlations) in a classical gas of loops[14] (loop gas) with the partition function

$$Z_1(y,x)=\sum_{\text{loop}}y^p x^{-L/2}\ . \tag{9}$$

Such models in two dimension were extensively studied by Nienhuis,[14] who convincingly argues them to be in the universality class of $O(n)$ models with $n=y$. In two dimensions a critical point is present only for $|y|\leq 2$. For $y>2$, the correlations are short-range for any finite x (with the exception of x goes to 0). Note that for $y=2$ the loop gas is just a restriction solid-on solid model[15,16] which is in the same universality class as the XY model and the Kosterlitz-Thouless transition[17] could take place. So the loop multiplicity turns out to be a determinant factor: In the case $y=4$ (and $x=2$), all correlations will be of a finite extent ξ. From the renormalization-group equation for the temperature $\partial T/\partial 1=(n-2)T^2$ (Ref. 18) of the $O(n)$ model, we may obtain a rough estimate for ξ. Using the relation $x\approx T^2$ and the above renormalization-group equation we deduce: $\xi(x)=a_0\times\exp(\frac{1}{2}x^{1/2})$. A rough order-of-magnitude estimate for ξ is therefore $a_0\exp(\frac{1}{2}\sqrt{2})\simeq 1.4a_0$ (a_0 is the lattice spacing).

It may be convincingly argued that ξ of this loop gas is an upper bound for the NNRVB antiferromagnetic correlations. Indeed, the extra factor $d(p_2)$ in (8) will increase the relative weight of the configurations with smaller loops for two reasons: (a) From (7), smaller L means larger p_2 and, on the average, $d(p_2)$ is monotonically increasing with p_2. (b) The smaller the L is, the weaker are the constraints the loops impose in the allowed dimer configurations.

To make a more quantitative estimate, we recall that in the regular square lattice the number of dimer coverings is given by $\exp(2NG/\pi)=(1.79\ldots)^N$ where G is the Catalan constant.[9] However, in the present problem, the space left by the loops is highly irregular and therefore $d(p_2)$ decreases beyond the naive reduction due to the smaller space available to the dimers. As a plausible possibility we conjecture the following "typical" behavior:

$$d(p_2)=\alpha^{2p_2}\ , \tag{10}$$

with

$$1<\alpha<\exp(2G/\pi)=1.79\ldots\ . \tag{11}$$

Assuming this behavior, the antiferromagnetic correlation length will again be that of the same loop gas with the partition function given in (9) but with a larger coupling $x\rightarrow x\alpha^2$ and hence a smaller $\xi\sim a_0\exp(\sqrt{2}/2\alpha)$.

We now consider an excitation spectrum of NNRVB. A candidate of an excitation is to change a singlet to a triplet. Other possible excitation is to have two unpaired spins at some distance. We do not know whether the two-spin interaction is repulsive or attractive. The energetics of the two spins will be considered elsewhere and this type of excitation is not considered here. The operator to create triplets involving site $\mathbf{r}$ in the RVB state is $\sigma_z(\mathbf{r})$ and a candidate excited state is

$$|k\rangle=O_k|\Psi\rangle=\sum_{\mathbf{r}}\exp(i\mathbf{k}\cdot\mathbf{r})\sigma_z(\mathbf{r})|\Psi\rangle\ . \tag{12}$$

The excitation energy $\omega_k=\langle k|H|k\rangle/\langle k|k\rangle-E_0$ (where E_0 is the ground-state energy) is written as

$$\omega_k=\frac{f_k}{s_k}\ , \tag{13}$$

where $f_k=\langle\Psi|[O_{-k},[H,O_k]]|\Psi\rangle/2$ and $s_k=\langle\Psi|O_{-k}O_k\times|\Psi\rangle$. This formula is essentially equivalent to the Bijl-Feynman formula for the excitation spectrum of liquid ^{4}He (Ref. 19). This type of approach in the spin system was recently attempted by Arovas, Auerbach, and Haldane[20] and Sutherland.[10] For NNRVB state, the numerator f_k is exactly calculated as $f_k=-E_0[2-\cos(k_xa_0)-\cos(k_ya_0)]/3$, and behaves as k^2 when k goes to zero. The denominator s_k is the Fourier transform of the correlation function. Since $\langle\Psi|\sum_{\mathbf{r}}\sigma_z(\mathbf{r})|\Psi\rangle=0$ (i.e., singlet ground state) and the correlation is of short range, it can be shown that $s_k\sim k^2$ as $k\rightarrow 0$. Therefore we have a finite gap as $k\rightarrow 0$ for the triplet excitation. In order to obtain the value of the energy gap and the excitation spectrum in this single-mode approximation we need a detailed form of the correlation function which perhaps requires numerical evaluations.

In conclusion we have considered a trial state which involves nearest-neighbor singlet pairings (NNRVB). This quantum-mechanical problem of the antiferromagnetic Heisenberg model in two dimensions is related to the loop gas and to the classical spin model with $O(4)$ symmetry which is disordered for all temperatures. Therefore, the antiferromagnetic correlation in NNRVB is of short range. This is a rather nontrivial result since the dimer covering problem, which is the basis of NNRVB, is known to be equivalent to the six-vertex model at the free fermion point.[21] The six-vertex model is critical, i.e., the correlation length is infinite.

The excitation which changes a singlet to a triplet was also considered. Using the Bijl-Feynman-type analysis, it was shown that this type of excitation has an energy gap.

For the superconductivity problem, it is essential to consider the excitation involving holes. The motion of holes must be treated quantum mechanically. In the effective Hamiltonian for the Hubbard model with large U and near half-filling, the antiferromagnetic spin coupling is second order in t/U, on the other hand the kinetic term for holes appears in first order. Therefore we expect that a hole perturbs the RVB vacuum. If the effect of the kinetic energy is to cause the hole to smear into a local distortion of the RVB state, statistical mechanics may still be a useful tool to study these excitations. Since the experiments suggest antiferromagnetic ordering in undoped samples, it may be the case that the excitations involving holes stablize the RVB state if it exists at all and is relevant to superconductivity.

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