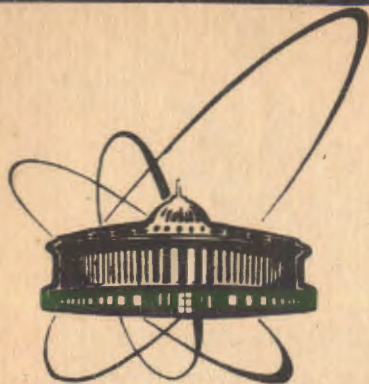


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QUANTUM ANTIFERROMAGNETS IN TWO
DIMENSIONS: THE RESONATING VALENCE
BOND SCHEME

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1. INTRODUCTION

The problem of the ground state of the quantum spin Heisenberg antiferromagnet (AFM) is one of the fundamental questions in the many-body theory. Despite of the continuous interest in this problem over about 60 years only a few exact results are available. There is the famous exact Bethe-Hulthen solution [1,2] for the one-dimensional problem yielding a power-law decay for the spin correlation in the ground state and an energy per spin $E/N = -\ln 2 + 1/4$. According to Marshall's theorem [3,4] the ground state is a singlet (total spin $S^2 = 0$) for N spins on a bipartite lattice with nearest-neighbour (nn) antiferromagnetic interactions. However, as indicated by various small-cluster calculations (see e.g. [5-7]) this could be true even in systems with frustration as the triangular lattice. Though we are far from an exact knowledge of the ground state in three dimensions, our picture on the antiferromagnetic long-range order (LRO) is well established by several approaches as perturbation theory [8], spin-wave theory [9] or variational methods [10]. In two dimensions the problem seems to be most complicated. On the one hand there are serious hints on LRO in the ground state [11-16] but according to Mermin and Wagner [17] no spontaneous magnetization is possible for finite temperatures. However, Mermin's and Wagner's theorem doesn't exclude an infinite zero-field susceptibility at finite temperatures suggesting that the two-dimensional (2d) Heisenberg magnet could be not far from LRO even for $T \neq 0$.

It is just this 2d quantum AFM which attracted a lot of interest in the last three years in connection with the high- T_c superconducting materials. The Cu spin in the Cu-O planes being responsible for the superconduction is $1/2$, the in-plane exchange is strong (about $10^3 K$ in La_2CuO_4 [18]) and exceeds the exchange perpendicular to the plane by much. The anisotropy in the spin space is small. For example the widely exploited La_2CuO_4 shows antiferromagnetic LRO in the undoped case with a Neel temperature up to about 300K [19]. For the stability of LRO for $T \neq 0$ the (weak) off-plane exchange coupling is responsible which is evident by neutron scattering showing a three-dimensional magnetic ordering [20]. Doping the material by Ba or Ca by about 1%-2% removes magnetic LRO [21] indicating a great sensibility of LRO to defects which should be typical for quasi-2d systems. A similar magnetic behaviour is

observed also YBaCuO [22], BiSrCaYCuO [23] and TlBaCaYCuO [24]. The magnetic state in the weakly doped (not yet superconducting) regime is characterized by a well-pronounced 2d antiferromagnetic short-range order (SRO) but no LRO and was called by Anderson [25,26] resonating-valence-bond (RVB) state. This notation was taken from chemical bonding and was first introduced in magnetism for the triangular-lattice AFM [27-29]. At present there is a broad discussion of RVB states in connection with 2d quantum antiferromagnetism in materials with high- T_c superconduction [30-40].

In the present paper we discuss the possibility of realization of short-range correlated RVB states in spin clusters up to 16 spins on square lattice. We compare results obtained by exact numerical diagonalization with results for RVB trial wave functions. It is known that the short-range correlated RVB state deviates significantly from the exact ground state in the pure square-lattice AFM. Taking the extrapolated values from [11] for E_{exact} and from [35] and [38] for E_{RVB} one finds for $N \rightarrow \infty$ $E_{\text{RVB}}/E_{\text{exact}} \sim 0.90 \dots 0.92$. Therefore we have to look for mechanisms introduced by doping which could support a short-range RVB state. In particular, we discuss different types of disorder, anisotropy, frustration as well as holes which can modify locally the exchange coupling.

2. EXACT DIAGONALISATION PROCEDURE AND GENERAL FEATURES OF RVB STATES

2.1 Exact diagonalization

We consider the isotropic Heisenberg Hamiltonian

$$H = \sum_{i \neq j} I_{ij} \mathbf{S}_i \cdot \mathbf{S}_j \quad (1)$$

with spin 1/2. For the ordinary AFM we assume $I_{ij}=1$ for i, j being nearest neighbours and $I_{ij}=0$, else. In order to construct the ground state wave function $|\Psi_0\rangle$ we can expand it to any complete set $|\varphi_n\rangle$ in the spin space

$$|\Psi_0\rangle = \sum_n c_n |\varphi_n\rangle. \quad (2)$$

Usually for the $|\varphi_n\rangle$ the direct product of local eigenstates of S_{iz} is used

$$|\varphi_n\rangle = |+\rangle|-\rangle|-\rangle \dots |+\rangle. \quad (3)$$

The number of the $|\varphi_n\rangle$ is 2^N and they represent the set of all Ising eigenstates. To find the coefficients c_n one has to diagonalize the matrix $H_{nm} = \langle \varphi_n | H | \varphi_m \rangle$. Because the size of the matrix increases exponentially the problem quickly exceeds the capacity of computers. By use of conservation laws and symmetries [11,41] the size of H_{nm} can be reduced drastically. In particular, one can take advantage of the commutations $[H, S_{2j}^-] = 0$ and $[H, S_{2j}^2] = 0$; where $S = \sum_i S_i$ represents the total spin. Using all other symmetries Poilblanc [41] could calculate recently all eigenvalues of 10 spins on square lattice with periodical boundary conditions analytically. However, if there are defects or disorder in the system (e.g. in high- T_c materials by doping) no symmetries can be exploited and the problem needs much more computational effort.

To calculate the ground state it is not necessary to diagonalize the whole matrix and the problem further simplifies. Very efficient Lanczos algorithms are mainly used for this task. In the present paper we used a modified version proposed by Gagliano et al. [40]. We start with an initial vector $|\phi_0\rangle$ which must have a non-vanishing overlap to the exact ground state. In the next step a new state $|\phi_1\rangle$ is constructed by applying the Hamiltonian H on the initial state $|\phi_0\rangle$

$$|\phi_1\rangle = b (H |\phi_0\rangle - a |\phi_0\rangle), \quad (4)$$

where $a = \langle \phi_0 | H | \phi_0 \rangle$ and $b = (\langle \phi_0 | H^2 | \phi_0 \rangle - a^2)^{-1/2}$ are chosen to normalize $|\phi_1\rangle$ and to orthogonalize $|\phi_0\rangle$ and $|\phi_1\rangle$. By diagonalizing the two-by-two matrix of H in the basis $|\phi_0\rangle, |\phi_1\rangle$ an improved eigenstate

$$|\tilde{\phi}_0\rangle = c_0 |\phi_0\rangle + c_1 |\phi_1\rangle \quad (5)$$

with lower energy as the initial state $|\phi_0\rangle$ is obtained. Repeating the procedure with $|\tilde{\phi}_0\rangle$ as the new initial state the ground state is approached after a few steps. This method is quite general and can be used for different magnetic models as well as other quantum mechanical systems (e.g. Hubbard model [43], t-J model [44]), but is restricted to small systems.

2.2 The RVB scheme

An alternative way to construct the ground state wave function is the concept of RVB trial wave functions based in some sense on

physical intuition. In their initial papers Anderson and Fazekas [27-29] tried to construct the magnetic ground state for the triangular lattice. They claimed that this ground state in analogy to the one-dimensional system is characterized by a well-pronounced SRD but no LRD (spin liquid). For a system of N spins (N even) a set of pair-bond states (PBS) $|f_\alpha\rangle$ is constructed where $|f_\alpha\rangle$ is any direct product of singlet states of paired spins i,j

$$|f_\alpha\rangle = |i_1j_1\rangle|i_2j_2\rangle\cdots|i_{N/2}j_{N/2}\rangle \quad (6)$$

with

$$|ij\rangle = \frac{1}{\sqrt{2}}(|i+\rangle|j-\rangle - |i-\rangle|j+\rangle) = -|j\rangle \quad (7)$$

being a singlet state of the spin pair (i,j), i.e. $(S_i + S_j)^2 |ij\rangle = 0$. Every $|f_\alpha\rangle$ represents a certain dimer covering of the system. In order to construct a trial wave function the PBS are superposed to a RVB state

$$|\Psi_{RVB}\rangle = \sum_{\alpha} c_{\alpha} |f_{\alpha}\rangle \quad (8)$$

The PBS have the following general features:

- i. The PBS are singlet eigenfunctions of the total spin $S = \sum_i S_i$, i.e. $S^2 |f_\alpha\rangle = 0$. Therefore $|\Psi_{RVB}\rangle$ is a singlet state, too.
- ii. The PBS are nonorthogonal

$$\langle f_\alpha | f_\beta \rangle = 2^{L-N} \quad (9)$$

with L being the number of loops in the loop covering generated by the superposition of the two dimer coverings corresponding to $|f_\alpha\rangle$ and $|f_\beta\rangle$ [32,33].

- iii. The total number of PBS is $Z_{PBS} = (N-1)!!$. The total number of independent singlet states is $Z_{S=0} = (N!) / \{(N/2)!(N/2+1)!\}$ [45], i.e. $Z_{PBS}/Z_{S=0} \sim (N/2e)^N$ for large N. The PBS are linearly dependent.
- iv. The PBS obey the relations [29]

$$|ij\rangle|ik\rangle + |ik\rangle|jl\rangle + |il\rangle|jk\rangle = 0 \quad (10a)$$

demonstrating the linear dependence of the PBS. The above relation can be expressed grafically by

$$\begin{array}{c} i \text{ --- } j \\ k \text{ --- } l \end{array} + \begin{array}{c} i \\ | \\ k \end{array} \begin{array}{c} j \\ | \\ l \end{array} + \begin{array}{c} i \quad j \\ \diagdown \quad \diagup \\ k \quad l \end{array} = 0 \quad (10b)$$

- v. Spin operators act on a PBS as follows [27]:

$$\langle f_\alpha | S_i | f_\alpha \rangle = 0 \quad (11a)$$

$$(1/4 - S_i S_j) |ij\rangle = |ij\rangle \quad (11b)$$

$$(1/4 - S_j S_k) |ij\rangle |kl\rangle = (1/2) |jk\rangle |li\rangle \quad (11c)$$

In any PBS the spin correlation of two paired spins takes its maximum value of $(-3/4)$.

- vi. The selection of the independent PBS can be done by Rumer diagrams [46] as follows: All spins are numbered and the numbers are written in a fixed sequence on a circumference of a circle (cf. fig.1). The pairing of two spins is represented by a line between the corresponding numbers. Any Rumer diagram with crossing lines can be expressed step by step via relation (10) by non-crossing diagrams, yielding finally a set of only non-crossing diagrams representing the set of independent PBS.
- vii. Labelling in a system of two sublattices A and B all A-sites with even numbers and all B-sites with odd numbers it becomes evident from fig.1b, that no intrasublattice pairing has to be taken into account.
- viii. The number of linearly independent PBS is equal to the number of independent singlet states [47], i.e. the linearly independent PBS are complete in the $S=0$ subspace.
- ix. The number of nn PBS of the square lattice is $Z_{nnPBS} \sim (1.792)^N$ [48] and for the square ladder $Z_{nnPBS} \sim 0.724 * (1.272)^N$. The energy $E_\alpha = \langle f_\alpha | H | f_\alpha \rangle$ per bond of a nn PBS for the Heisenberg AFM on the square lattice is $(1/4)(-3/4)I$ and on the square ladder is $(1/3)(-3/4)I$ and increases if non-nearest-neighbour pairing is allowed. This is slightly larger than the energy of the Neel state for the square lattice and equal to the Neel energy for the square ladder. Because the set of the PBS is complete in the $S=0$ subspace the RVB scheme allows (as other basis sets like e.g. the Ising eigenstates (3), too), in principle, to construct any $S=0$ eigenstate exactly.

This can be done really for small systems by computer. However, the use of PBS as basis states has an advantage, which can be very useful for approximative eigenstates for larger systems: Every basis state corresponds to a dimer covering of the system, i.e. to a graph with a certain physical meaning. For the construction of any trial wave function a subset of physically relevant PBS can be selected and/or a certain ansatz for the coefficients c_α can be chosen [32,33,35,40]. The choice of the relevant PBS as well as the ansatz for the coefficients depend on the physical ingredients of the system and are an object of physical intuition as well as optimization of $E_{RVB}(c_\alpha)$. Since in any PBS the spin correlation of paired spins is strong, for a short-range correlated state the restriction to pairing of neighbored spins is reasonable. Taking into account successively the pairing of spins over longer distances the magnetic correlation length increases [35].

3. RESULTS FOR FINITE SQUARE-LATTICE SYSTEMS

We investigate in this section short-range correlated nn RVB states

$$|\Psi_{RVB}\rangle = \sum_{\alpha} c_{\alpha} |f_{\alpha}^{nn}\rangle \quad (12)$$

with $|f_{\alpha}^{nn}\rangle$ being a PBS with nn pairing, only. As standard ansatz for the coefficients often $c_{\alpha}=1$ is used [32,33]. This ansatz is good in periodic systems, though even there an inequivalence of the nn PBS exists [30]. Additionally we consider optimized coefficients c_{α} obtained by minimizing

$$E_{RVB} = \langle \Psi_{RVB} | H | \Psi_{RVB} \rangle / \langle \Psi_{RVB} | \Psi_{RVB} \rangle = \frac{(\sum_{\alpha,\beta} c_{\alpha} c_{\beta} H_{\alpha\beta})}{(\sum_{\alpha,\beta} c_{\alpha} c_{\beta} S_{\alpha\beta})}$$

$$H_{\alpha\beta} = \langle f_{\alpha}^{nn} | H | f_{\beta}^{nn} \rangle; \quad S_{\alpha\beta} = \langle f_{\alpha}^{nn} | f_{\beta}^{nn} \rangle. \quad (13)$$

As discussed in the introduction the short-range correlated RVB state differs in its energy significantly from the long-range correlated ground state of the pure square lattice. However, as it is evident for example from the phase diagram of LaCuO due to doping a short-range correlated magnetic state is established being according to Anderson [25,26] a RVB state. Therefore we have to look for possible mechanisms to favour a short-range correlated RVB state. Possible deviations from the ideal isotropic square lattice

Heisenberg AFM in doped high- T_c materials could be: anisotropy, disorder, frustration and, particularly, the direct influence of holes. We discuss the role of all of them by comparing the exact energy E_{exact} with the energy E_{RVB} of the trial RVB state for spin clusters of 3×4 spins with open boundary conditions (figures 2-4, 6-9) as well as 4×4 spins (fig.5) with periodic boundary conditions

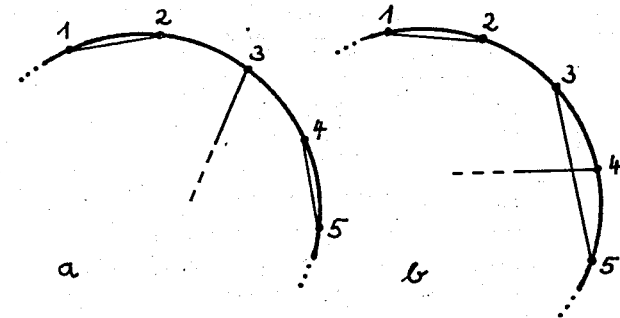


Fig.1: Illustration of two sections of different Rumer diagrams with (b) and without (a) crossing bonds (cf.text).

on the square lattice. The open boundary conditions for the 3×4 cluster are chosen to avoid artificial frustration.

In [20] for LaCuO a weak Ising anisotropy along the orthorhombic c-axis is suggested. In principle, any anisotropy should more or less rule out a singlet RVB state, because the commutation rule $[H, S^2] = 0$ doesn't hold for an anisotropic Hamiltonian and the ground state cannot be a pure singlet. Results are presented in fig.2. Obviously, in the limit of strong easy-axis anisotropy (Ising) the singlet RVB state is far from being a good trial state. In the case of easy-plane anisotropy (xy) the effect is similar but less drastic. However, it becomes evident that small anisotropies of about 30% do not rule out the RVB ansatz as trial wave function. Next we discuss disorder of three different kinds: random anisotropy, random field and random exchange. In a disordered system the standard ansatz with equal-weight coefficients is expected to be bad and the version with optimized coefficients c has to

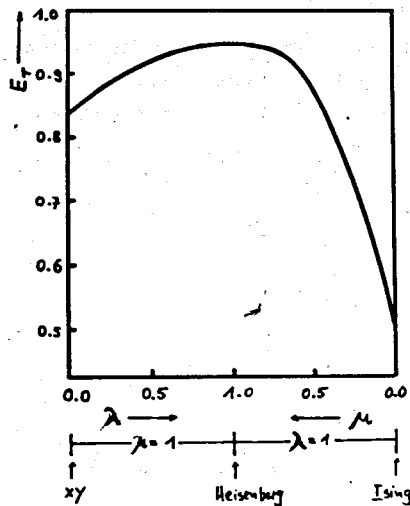


Fig. 2: $E_r = E_{RVB}/E_{exact}$ in dependence on anisotropy for a 3×4 cluster. The anisotropy parameters λ and μ are introduced in the Hamiltonian by $H = \sum_{ij} I_{ij} [\lambda S_{iz} S_{jz} + \mu (S_{ix} S_{jx} + S_{iy} S_{jy})]$. $\lambda=1, \mu=1$ correspond to the isotropic Heisenberg model, $\lambda=1, \mu=0$ to the Ising model and $\lambda=0, \mu=1$ to the xy model.

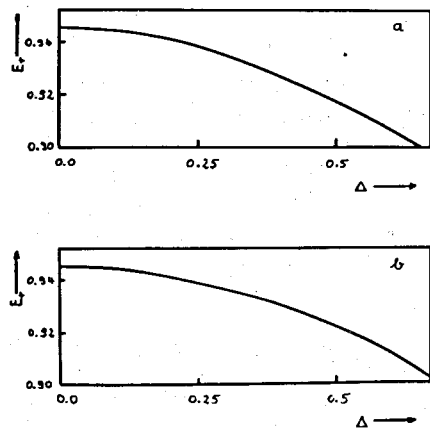


Fig. 3: $E_r = E_{RVB}/E_{exact}$ in dependence on random field (a) and random anisotropy (b) for a 3×4 cluster. Random field is introduced by $H = \sum_{ij} I_{ij} S_i S_j - \sum_i h_i S_{iz}$ with $\langle h_i \rangle = 0$ and random anisotropy by $H = \sum_{ij} I_{ij} [S_{iz} S_{jz} + A_i A_j (S_{ix} S_{jx} + S_{iy} S_{jy})]$ with $\langle A_i \rangle = 1$. The strength of disorder Δ is determined by $\Delta = \langle h_i^2 \rangle^{1/2}$ and $\Delta = \langle (A_i - 1)^2 \rangle^{1/2}$, respectively.

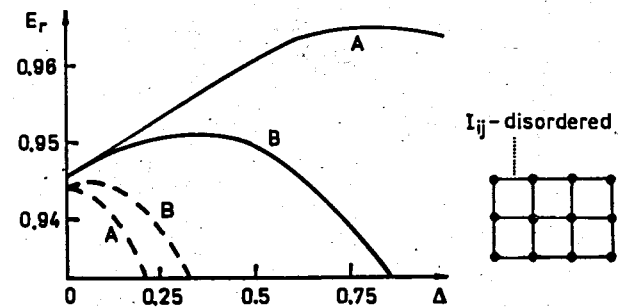


Fig. 4: $E_r = E_{RVB}/E_{exact}$ in dependence on random exchange for two different realizations of randomness on a 3×4 cluster. The strength of disorder is $\Delta = \langle (I_{ij} - \langle I_{ij} \rangle)^2 \rangle^{1/2}$ with $\langle I_{ij} \rangle = 1$ and i, j being nearest neighbours. The solid lines correspond to the RVB state with optimized coefficients and the dashed lines to the RVB state with equal coefficients.

be preferred. Any disorder should decrease long-range correlations and, in principle, a stabilization of the RVB state by disorder could be possible. However, random field or random anisotropy violate $[H, S^2] = 0$ and forbid pure singlet states. Results are shown in fig. 3. Evidently, weak random fields or anisotropies do not rule out the RVB ansatz, but don't support it. More relevant with respect to a stabilization of the RVB state is exchange disorder, which conserves the full rotational symmetry of the Heisenberg Hamiltonian but acts against LRO. Fig. 4 shows results for two different realizations of random exchange parameters. Exchange disorder, indeed, can favour the RVB state. Most drastic is the influence of frustration shown in fig. 5. Frustration in form of next-nearest neighbour (nnn) antiferromagnetic bonds was estimated in [49] to be of the order $I_{nnn}/I_{nn} \sim 5...8\%$ in undoped La_2CuO_4 and should be increased by doping [50-52]. Homogeneous frustration of about $I_{nnn}/I_{nn} = 0.4$ brings the nn RVB state with equal-weight coefficients very close to the exact eigenstate. (We note that $I_{nnn}/I_{nn} \sim 0.4$ is just the value for which the magnetic LRO in the ground state is expected to vanish [53]). For the considered periodic 4×4 cluster the RVB energy differs only by 0.3% and the overlap $\langle \Psi_{RVB} | \Psi_{exact} \rangle$ is 99.6%. Combination of both frustration and exchange disorder can additionally favour the RVB ansatz [40].

Finally we consider the influence of holes directly. As discussed by Emery [54] and supported by photoemission experiments [55] the

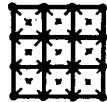
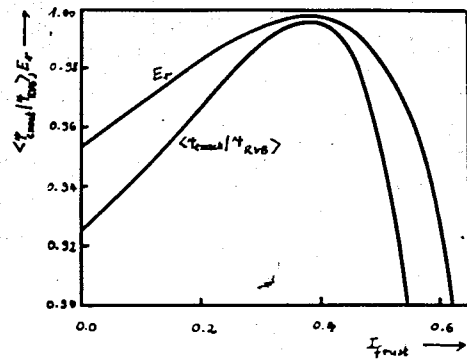


Fig. 5: $E_r = E_{RVB}/E_{exact}$ and overlap $\langle \Psi_{RVB} | \Psi_{exact} \rangle$ in dependence on frustration introduced by next nearest neighbour antiferromagnetic exchange for a periodic 4×4 cluster.

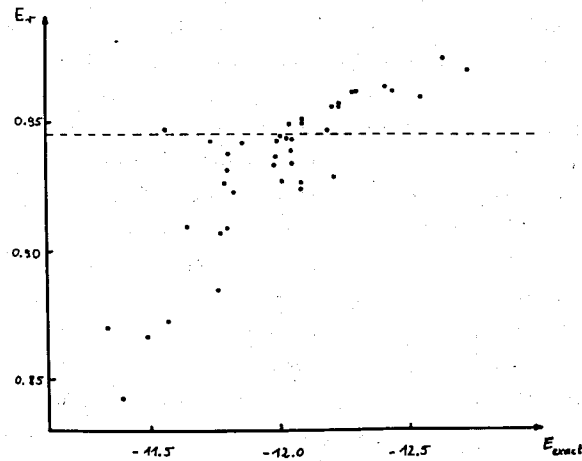


Fig. 6: $E_r = E_{RVB}/E_{exact}$ versus the energy of the spin system E_{exact} for all 41 different two-hole configurations on a 3×4 cluster with $I_{hole} = -0.5$.

holes doped in the Cu-O planes mainly occupy the oxygen sites. Because the exchange coupling between the Cu spins is dominated by a superexchange mechanism the holes modify locally the bonds. Even a local change from an antiferromagnetic to a strong ferromagnetic bond due to a hole is suggested [56]. Here we discuss one and two holes on a 3×4 cluster. The antiferromagnetic bonds not modified by

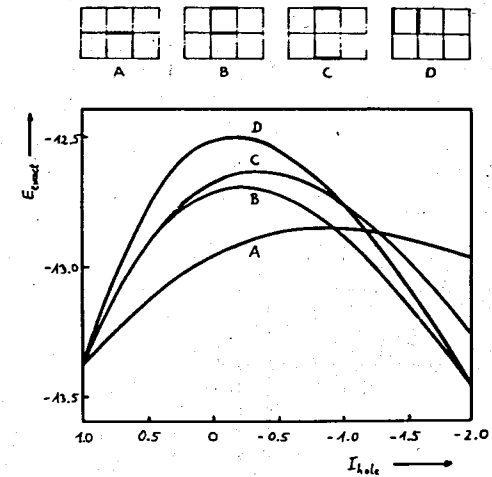


Fig. 7: Energy E_{exact} of the one-hole configuration (A) and of the three two-hole configurations (B,C,D) with the lowest energies in dependence on I_{hole} .

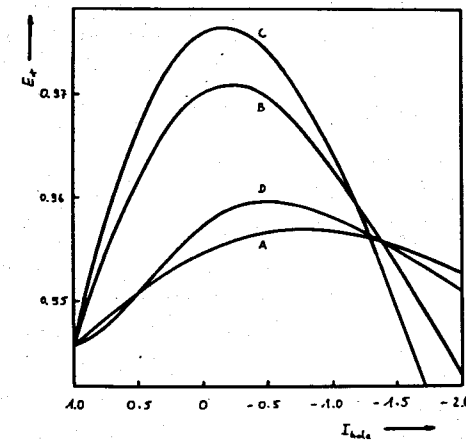


Fig. 8: $E_r = E_{RVB}/E_{exact}$ in dependence on I_{hole} for the same hole configurations as in fig. 7.

a hole are as usual assumed to $I_{ij} = 1$ and the bonds modified by a hole are $I_{ij} = I_{hole}$ where I_{hole} can vary from antiferromagnetic to ferromagnetic exchange. In the one-hole case we put the hole in the center of the cluster (cf. fig. 7). In the case of two holes there are all together 41 different arrangements of holes. In fig. 6 for these 41 two-hole configurations the ratio $E_r = E_{RVB}/E_{exact}$ is

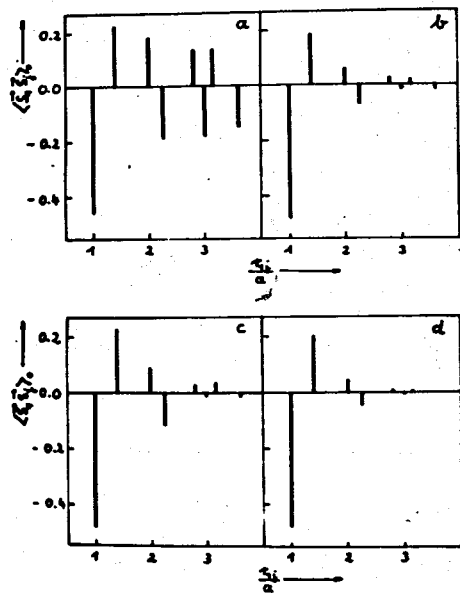


Fig. 9: Spin-spin correlation $\langle S_i^z S_{i+a}^z \rangle$ for the ordinary AFM on a 3×4 cluster and for the two-hole configuration C (cf. fig. 7) with $I_{\text{hole}} = -0.2$. Spin with number 1 sits at the corner of the cluster. a: AFM, exact; b: AFM, RVB; c: two holes, exact; d: two holes, RVB.

drawn versus the energy of the configuration E_{exact} for $I_{\text{hole}} = -0.5$. Obviously, there is a certain "correlation" between E_r and E_{exact} , i.e. the hole configurations with low energies favour the RVB state. Since the low-energy configurations are just realized in the system we study the three ones with lowest energy (cf. fig. 8) in more detail. The energy of the configuration versus I_{hole} is shown in fig. 7 and $E_{\text{RVB}}/E_{\text{exact}}$ versus I_{hole} in fig. 8. The realization of a RVB state is particularly supported by holes if the hole modifies the bond to a weak ferromagnetic one. For stronger ferromagnetic bonds the singlet pairing is not sufficient and triplet pairing has to be taken into account. In the last figure we show the spin correlation function calculated with the exact eigenstate as well as the RVB state for the pure AFM and the doped AFM. The slow decay of the exact spin correlation in comparison with the fast decay of the RVB result for the pure AFM is evident. Due to holes the decay becomes steeper and the RVB result is close to the exact one. In both cases the strong antiferromagnetic SRO is well described by the RVB state.

4. SUMMARY

The resonating-valence bond scheme is a suitable method to construct the ground state of the quantum spin Heisenberg AFM in low dimensions. The RVB state is a superposition of basis states which correspond to dimer coverings of the system. Hence the choice of relevant basis states to construct a trial wave functions can be based on the physical ingredients of the considered spin system. For short range-correlated states as observed in slightly doped high- T_c superconducting materials the restriction to nn dimer coverings is reasonable. Such a nn RVB state, however, significantly deviates from the real ground state for the pure square lattice AFM. Therefore we investigated the influence of various deviations from the pure Heisenberg AFM which could be present in the slightly doped materials. We found that exchange disorder and, particularly, frustration can stabilize a short-range correlated RVB state. Furthermore we considered the influence of holes directly, assuming that holes locally modify the exchange bonds. Also in this case we obtained a possible stabilization of the RVB state due to doping.

Acknowledgements

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