Low-Energy Sector of the $S = 1/2$ Kagome Antiferromagnet

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Starting from a modified version of the $S = 1/2$ Kagome antiferromagnet to emphasize the role of elementary triangles, an effective Hamiltonian involving spin and chirality variables is derived. A mean-field decoupling that retains the quantum nature of these variables is shown to yield a Hamiltonian that can be solved exactly, leading to the following predictions: (i) The number of low-lying singlet states increases with the number of sites N like 1.15^N; (ii) a singlet-triplet gap remains in the thermodynamic limit; (iii) spinons form bound states with a small binding energy. By comparing these properties with those of the regular Kagome lattice as revealed by numerical experiments, we argue that this description captures the essential low-energy physics of that model. [S0031-9007(98)07095-1]

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Despite a very intense activity over the past ten years, the magnetic properties of the $S = 1/2$ Kagome antiferromagnet remain an open problem. If a number of facts seem to be rather firmly established by now thanks to the very extensive numerical simulations that have been performed on that system [1–8], a simple theoretical picture that accounts for the basic findings has not emerged yet. The most striking feature is probably the presence of many, low-lying singlet states [7,8]. The first indication that this might be the case was the appearance of a low temperature peak in the specific heat. While the evolution of this peak with the size of the system is not clear yet, the numerical determination of all the low-lying singlet states for systems with up to 36 sites shows that their number increases like 1.15^N, where N is the number of sites of the system. The best candidate to explain this proliferation of low-lying singlets is a short-range resonating-valence-bond description of the low-energy sector based on dimer coverings of the Kagome lattice with nearest-neighbor singlets [9–11]. The main problem with this approach is that the number of dimer states increases like 1.26^N, i.e., much too fast [12], and no convincing criterion could be found that allows one to select the relevant singlet states. The other important, although less accurately established, findings of the numerical simulations are the absence of a long-range magnetic order in the ground state and the presence of a singlet-triplet gap in the thermodynamic limit [7,8]. Finally the role of spin $1/2$ excitations, as well as the consistency of the numerical results with some exotic types of order [13–16], is still under investigation.

In this paper, we propose a simple explanation of these properties. We start from the following observation: The exponential increase of the number of these low-lying states suggests that they originate from the partial lifting of a local degeneracy that would be present if some of the couplings were set to zero. Now the natural bricks to construct the Kagome lattice are triangles, and spins $1/2$ on a triangle lead to a fourfold degenerate ground state: two doublets that differ by their chirality. So let us investigate how this degeneracy is lifted if one constructs the Kagome lattice by coupling triangles. This amounts to studying the modified Kagome lattice depicted in Fig. 1 starting from the limit $J'/J \ll 1$. This can be seen as a triangular lattice of triangles with $N_t = N/3$ sites, where N is the number of sites of the Kagome lattice.

The first step is to derive an effective Hamiltonian in the subspace of the ground states of the triangles, as in Subrahmanyam's block spin perturbation approach to the nondimerized Kagome lattice [17]. Following Schulz's approach to the problem of three coupled Heisenberg chains with periodic boundary conditions [18], we describe the four ground states of a triangle with two Pauli matrices: $\vec{\sigma}$ for the spin of the doublet, the eigenstates of σ_z being denoted $\uparrow$ and $\downarrow$, and $\vec{\tau}$ for its chirality, the eigenstates of τ_z being denoted R for right and L for left. In

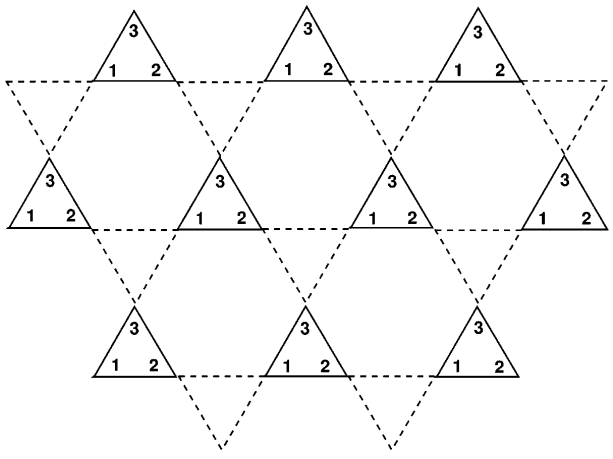

FIG. 1. Sketch of the dimerized Kagome lattice. Solid lines: J; dashed lines: J'.

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terms of the original spins $\vec{S}$, these states can be written

$$|\alpha R\rangle = \frac{1}{\sqrt{3}}\left(|-\alpha\alpha\alpha\rangle + \omega|\alpha-\alpha\alpha\rangle + \omega^2|\alpha\alpha-\alpha\rangle\right), \tag{1}$$

$$|\alpha L\rangle = \frac{1}{\sqrt{3}}\left(|-\alpha\alpha\alpha\rangle + \omega^2|\alpha-\alpha\alpha\rangle + \omega|\alpha\alpha-\alpha\rangle\right),$$

where $\omega = \exp(2\pi i/3)$ and $\alpha = \uparrow$ or $\downarrow$. $|\alpha_1\alpha_2\alpha_3\rangle$ represents a configuration of the original spins $\vec{S}$ within one triangle, the indices corresponding to the convention of Fig. 1. Note that the total spin is now given by $(\sum_i' \vec{\sigma}_i)^2$, where the prime means that the sum runs over the triangular lattice. Each triangle has an energy $-3J/4$, and energies will be measured with respect to the ground state energy $-(3J/4)N_t$ of the $J'/J = 0$ case. Then, to the first order in J', the effective Hamiltonian $\tilde{H}$ on the triangular lattice is given by

$$\tilde{H} = (J'/9)\sum_{\langle i,j\rangle}{}' \tilde{H}^{\sigma}_{ij}\tilde{H}^{\tau}_{ij}, \qquad \tilde{H}^{\sigma}_{ij} = \vec{\sigma}_i.\vec{\sigma}_j, \tag{2}$$

$$\tilde{H}^{\tau}_{ij} = [1-2(\alpha_{ij}\tau^-_i + \alpha^2_{ij}\tau^+_i)][1-2(\beta_{ij}\tau^-_j + \beta^2_{ij}\tau^+_j)],$$

where $\langle i,j\rangle$ denotes pairs of nearest neighbors. In $\tilde{H}^{\tau}_{ij}$, α_{ij} and β_{ij} are complex parameters that depend on the type of bond: α_{ij} (respectively, β_{ij}) equals 1, ω^2, or ω when the original spin in triangle i (respectively, j) involved in the bond (i,j) sits at site 1, 2, or 3 with the convention of Fig. 1. In the basis $|RR\rangle$, $|RL\rangle$, $|LR\rangle$, and $|LL\rangle$ the eigenstates of $\tilde{H}^{\tau}_{ij}$ can be easily calculated

$$\begin{aligned}
|\phi^{\tau}_1(i,j)\rangle &= \frac{1}{2}(1,-\beta_{ij},-\alpha_{ij},\alpha_{ij}\beta_{ij})\ E_1 = 9,\\
|\phi^{\tau}_2(i,j)\rangle &= \frac{1}{2}(1,\beta_{ij},\alpha_{ij},\alpha_{ij}\beta_{ij})\ E_2 = 1,\\
|\phi^{\tau}_3(i,j)\rangle &= \frac{1}{2}(1,-\beta_{ij},\alpha_{ij},-\alpha_{ij}\beta_{ij})\ E_3 = -3,\\
|\phi^{\tau}_4(i,j)\rangle &= \frac{1}{2}(1,\beta_{ij},-\alpha_{ij},-\alpha_{ij}\beta_{ij})\ E_4 = -3,
\end{aligned} \tag{3}$$

while the eigenstates of $\tilde{H}^{\sigma}_{ij}$ are denoted $|\phi^{\sigma}_{S,m}(i,j)\rangle$ with energies $-3/4$ for the singlet ($S = 0$, $m = 0$) and $1/4$ for the triplets ($S = 1$, $m = 0, \pm 1$).

It is useful to start with two triangles coupled by a single link. In the spirit of the Majumdar-Ghosh solution of the $J_1 - J_2$ chain [19], the ground state wave function in terms of the spin $\vec{S}$ is obtained as the product of three singlets involving, respectively, the link between the triangles and the remaining two spins on each triangle (see Fig. 2), and its energy is $-6J/4 - 3J'/4$. In that simple case, the effective Hamiltonian $\tilde{H}_{ij}$ is the product of a spin part $\tilde{H}^{\sigma}_{ij}$ and a chirality part $\tilde{H}^{\tau}_{ij}$, and its eigenvalues are the products of the eigenvalues of $\tilde{H}^{\sigma}_{ij}$ with those of $\tilde{H}^{\tau}_{ij}$. The ground state energy of $\tilde{H}_{ij}$ is thus $-3J'/4$: The exact ground state energy is recovered, as it should be, since it depends linearly on J'.

In the general case of Eq. (2), the Hamiltonian is not the product of a spin part and a chirality part, and its solution is, in principle, as difficult as the original problem. However, we note that the Hamiltonian of Eq. (2) is formally similar to the Kugel-Khomskii model that was introduced in the context of orbitally degenerate magnets [20], and a mean-field decoupling of the spin and orbital degrees of freedom is known to give an accurate picture of the physics when the asymmetry between spin and orbital degrees of freedom is strong enough [21]. In our case, the asymmetry between spin and chiral variables in Eq. (2) is clearly very strong, and such a decoupling amounts to the replacement of $\tilde{H}$ with the mean-field Hamiltonian defined on the triangular lattice,

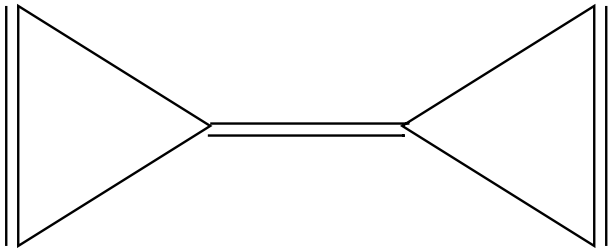

FIG. 2. Solution of the two-triangle problem. Singlets are represented as double lines.

$$H_{MF} = \sum_{\langle i,j\rangle}{}' (a^{\tau}_{ij}\tilde{H}^{\sigma}_{ij} + a^{\sigma}_{ij}\tilde{H}^{\tau}_{ij} - a^{\sigma}_{ij}a^{\tau}_{ij}), \tag{4}$$

where the parameters $a^{\tau}_{ij} \equiv \langle\tilde{H}^{\tau}_{ij}\rangle$ and $a^{\sigma}_{ij} \equiv \langle\tilde{H}^{\sigma}_{ij}\rangle$ have to be determined self-consistently. Note that this mean-field problem is still very complicated *a priori* since it involves $S = 1/2$ Heisenberg-like models on a triangular lattice.

Remarkably enough, the low-energy solutions of that problem can be determined analytically. Let us concentrate for the moment on clusters with an even number of sites and with periodic boundary conditions, and let us consider a dimer covering of the triangular lattice by nearest-neighbor dimers. Denoting by D the set of nearest-neighbor pairs that enter this covering, we can construct a wave function $|\Phi_0(D)\rangle$ in the following way:

$$|\Phi_0(D)\rangle = \prod_{\langle i,j\rangle\in D} |\phi^{\tau}_1(i,j)\rangle \otimes |\phi^{\sigma}_{0,0}(i,j)\rangle. \tag{5}$$

Clearly $|\Phi_0(D)\rangle$ will be a solution of the problem if $a^{\tau}_{ij} = a^{\sigma}_{ij} = 0$ as soon as $\langle i,j\rangle \notin D$. This turns out to be true thanks to the following properties:

$$\begin{aligned}
\langle\phi^{\tau}_1(i,j)\phi^{\tau}_1(k,l)|\tilde{H}^{\tau}_{jk}|\phi^{\tau}_1(i,j)\phi^{\tau}_1(k,l)\rangle &= 0,\\
\langle\phi^{\sigma}_{0,0}(i,j)\phi^{\sigma}_{0,0}(k,l)|\tilde{H}^{\sigma}_{jk}|\phi^{\sigma}_{0,0}(i,j)\phi^{\sigma}_{0,0}(k,l)\rangle &= 0,
\end{aligned} \tag{6}$$

which can be easily checked directly with the expressions of the wave functions. So $|\Phi_0(D)\rangle$ is a solution characterized by $a^{\tau}_{ij} = 9$, $a^{\sigma}_{ij} = -3/4$, if $\langle i,j\rangle \in D$ and 0 otherwise; its energy is given by $E_0(S=0) = -(3J'/8)N_t$, and it is a singlet [22]. To prove that it minimizes the energy of Eq. (4), we have solved the mean-field problem numerically on small clusters with up to 3×4 sites.

This has been done by iteration starting from random values of a_{ij}^{σ} and a_{ij}^{τ}, and we found that the lowest energy is always equal to $-(3J'/8)N_t$. In fact, solutions such as $\Phi_0(D)$ exist most of the time for such mean-field Hamiltonians, but they are usually significantly higher in energy than uniform solutions. In the present case, however, other solutions involving $|\phi_2^{\tau}(i,j)\rangle$ are very bad energetically because $E_2 = E_1/9$. Note that the wave function $|\phi_1^{\tau}(i,j)\rangle$ corresponds neither to ferromagnetic nor antiferromagnetic ordering of the chiral variables since it is a linear combination of the four basis states $|RR\rangle$, $|RL\rangle$, $|LR\rangle$, and $|LL\rangle$. So there is no chiral ordering of the type discussed by Baskaran [23] for the triangular lattice even locally.

Similarly, triplet solutions can be constructed for a given dimer covering of the triangular lattice. Choose two neighboring sites (i_0, j_0), and consider a dimer covering $D(i_0, j_0)$ of the remaining sites. The wave function with lowest energy is of the form

$$|\Phi_0[D(i_0,j_0)]\rangle[|\phi_n^{\tau}(i_0,j_0)\rangle \otimes |\phi_{1,m}^{\sigma}(i_0,j_0)\rangle]. \quad (7)$$

In this expression, $m = 0, \pm 1$ and n can take the values of 3 or 4 [24]. The energy of this state is $E_0(S=1) = E_0(S=0) + (2/3)J'$. Again it was checked numerically on a 3×4 cluster that this is indeed the lowest energy in the triplet sector. So this mean-field approach predicts that there is a singlet-triplet energy gap Δ equal to $(2/3)J'$.

Another class of low-lying states exists in the triplet sector. It can be constructed in the following way: Choose two sites (k,l) that are *not* nearest neighbors, denote by $D(k,l)$ a dimer covering of the remaining sites, and consider the wave function

$$|\Phi_0(D(k,l))\rangle\,|\sigma_k\tau_k\rangle\,|\sigma_l\tau_l\rangle\,, \quad (8)$$

where $|\sigma_k\tau_k\rangle$ (respectively, $|\sigma_l\tau_l\rangle$) can be any configuration at site k (respectively, l). Then similar arguments show that this is a solution with energy $E_1(S=1) = E_0(S=1) + J'/12$. Each unpaired site corresponds to a $S = 1/2$ excitation and can be interpreted as a spinon. This mean-field approach predicts that spinons form triplet bound states on neighboring sites with a binding energy equal to $J'/12$. In fact, if we consider a cluster with an odd number of sites, the ground state can be shown to consist of one unpaired site—i.e., one spinon—times $|\Phi_0(D)\rangle$, where D is a dimer covering of the remaining sites.

Now let us turn to the very interesting question of the ground state degeneracy. The energy $E_0(S=0)$ does not depend on the particular dimer covering D of the triangular lattice used to construct the wave function $|\Phi_0(D)\rangle$. So, for a given cluster, the degeneracy is controlled by the number of dimer coverings. This number can be calculated using standard techniques [25,26]. For the triangular lattice, we found that it increases with the number of sites N_t like α^{N_t} with $\ln\alpha = \frac{1}{16\pi^2}\int_0^{2\pi}\int_0^{2\pi} \ln(4 + 4\sin x \sin y + 4\sin^2 y)\,dx\,dy$. A numerical integration yields $\ln\alpha = 0.4286$, or $\alpha = 1.5351$. Translated into the language of the original Kagome lattice, this corresponds to a degeneracy that increases like $(\alpha^{1/3})^N = 1.1536^N$, where N is the number of sites of the Kagome lattice since $N_t = N/3$.

How good is this mean-field approach? One argument in its favor is that it is qualitatively correct in the case of three coupled chains with periodic boundary conditions: Schulz's analysis based on the renormalization group argument predicts a dimerized, twofold degenerate ground state with gapped spin excitations [18,27], and this is exactly the physics of the mean-field solution adapted to that case [28]. Besides, one can compare mean-field and exact results for the effective Hamiltonian on small clusters. We have done this on rings of n sites with diagonal bonds up to $n = 6$. The details will be given elsewhere, but the answer is unambiguous: The number of degenerate mean-field ground states always corresponds to the number of low-lying singlets of the exact solution. To go beyond, the mean field is expected to partially lift the degeneracy within the ground state manifold but not to change the number of low-lying singlets.

Now, let us come to the most important question: What can we learn from this approach concerning the regular (nondimerized) Kagome lattice? The first issue to address is whether the present picture, developed in the limit $J'/J \ll 1$, can remain a good one for the original model corresponding to $J'/J = 1$. A necessary and sufficient condition for this to be true is that the singlet sector of the model of Fig. 1 evolves smoothly as a function of J'/J, i.e., that the low-lying singlets of the regular Kagome model can be reached adiabatically from those of the modified one. As a first step in this direction, we have studied the evolution of the spectrum of the two-triangle problem of Fig. 2. The results are quite interesting: The hierarchy of the low-lying states is exactly the same for $J'/J = 0.1$ and $J'/J = 1$. In particular, cranking up J' does not pull down singlet states of higher energy even when $J' = J$, i.e., when the coupling between triangles is already a significant fraction of the gap of an isolated triangle $(3J/2)$. To see whether the same is true for the Kagome lattice itself is a numerical task far beyond the scope of the present paper. However, the previous result is quite encouraging, and it is the author's hope that the present paper will indeed motivate such a calculation.

Finally, let us translate our results in terms of the original spins $\vec{S}$. We know from the analysis of the two-triangle problem that the basic brick of our mean-field wave function, namely, a two-site wave function of the type $|\phi_1^{\tau}(i,j)\rangle \otimes |\phi_{0,0}^{\sigma}(i,j)\rangle$, corresponds to a dimer mapping of the two-triangle problem (see Fig. 2). So, the wave functions of Eq. (5) correspond to a certain subset of the dimerized wave functions of the Kagome lattice used by Zeng and Elser [11]. Besides, the number of states selected with this criterion increases roughly like 1.15^N, in agreement with the numerical results for even clusters [8]. This is probably the most interesting result of the present approach since it provides a simple but nevertheless quantitative explanation of the very numerous low-lying

singlet states of the $S = 1/2$ Kagome antiferromagnet: Their number scales with the number of dimer coverings of the underlying triangular lattice of triangles. The fact that one can choose triangles pointing upwards or downwards to build the wave function means that the actual low-lying states can have domains with different orientations of the triangles. Elementary energy considerations suggest that these domains are large, however, so that they can only marginally contribute to the increase of the number of singlet states with the size of the system. Interestingly enough we can also explain the apparent discrepancy between odd and even clusters in the results of Ref. [8]. For odd clusters, there is an unpaired site, and according to the present theory, the degeneracy is expected to scale like $N \times 1.15^N$. Omitting this prefactor in the fit leads to an overestimate of α (see Fig. 3).

To summarize, we have studied a dimerized version of the $S = 1/2$ Kagome antiferromagnet by performing a mean-field analysis of the effective Hamiltonian that describes its low-energy sector. This approach leads to a transparent picture of the low-energy properties which turns out to bear remarkable similarities to those reported for the regular Kagome lattice [7,8], e.g., the number of low-lying singlets or the presence of a singlet-triplet gap. Besides, these results lead to natural subspaces of dimer wave functions to describe the low-energy singlet and triplet sectors of the regular Kagome model. A variational study of this model using these wave functions is in progress to make more precise statements about the accuracy of this description of the low-energy sector of the regular Kagome antiferromagnet. It is expected to give useful information concerning the structure of the low-lying singlet sector, for instance, the low temperature specific heat, and to allow one to make more precise predictions concerning the persistence of the singlet-triplet gap and the binding of spinons in the thermodynamic limit.

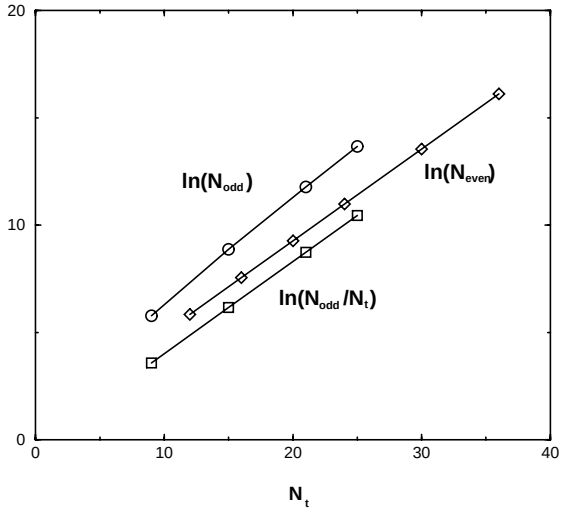

FIG. 3. Logarithm of the number of dimer coverings of the triangular lattice as a function of the number of sites N_t for small systems with periodic boundary conditions. The slope of $\ln(N_{\rm odd})$ is 1.18, while the slope of both $\ln(N_{\rm even})$ and $\ln(N_{\rm odd}/N_t)$ is 1.15.

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