Hidden long-range order in kagomé Heisenberg antiferromagnets

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We give a physical picture of the low-energy sector of the spin-$\frac{1}{2}$ kagomé Heisenberg antiferromagnet (KAF). It is shown that the kagomé lattice can be presented as a set of stars which contain 12 spins and are arranged in a triangular lattice. Each of these stars has two degenerate singlet ground states which can be considered in terms of pseudospin. As a result of the interaction between stars we get the Hamiltonian of the Ising ferromagnet in a magnetic field. So in contrast to the common view there is a long-range order in KAF consisting of definite singlet states of stars.

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In spite of numerous theoretical and experimental studies in the last decade, some magnetic properties of kagomé antiferromagnets (KAF's) remain open problems. Experiments revealed an unusual low-temperature behavior of the specific heat and magnetic susceptibility in kagomé-like compounds. For example specific heat measurements in $SrCr_{9p}Ga_{12-9p}O_{19}$ ($S=3/2$ kagomé material) have shown that there is a peak at $T\approx 5$ K, $C\propto T^2$ at $T\lesssim 5$ K, and it appears to be practically independent of magnetic field up to 12 T.[1]

Some of the experimental findings are in agreement with the results of numerical finite-cluster investigations.[2–6] They reveal a gap separating the ground state from the upper triplet levels and a band of nonmagnetic singlet excitations with a very small or zero gap inside the triplet gap. The number of states in the band increases with the number of sites N as α^N. For samples with up to 36 sites $\alpha=1.15$ and 1.18 for even and odd N, respectively.[2,5] This wealth of low-lying singlet excitations explains the peak of specific heat at low temperature and its independence of the magnetic field.[1,7]

However, the origin of this band as well as the nature of the ground state has been unclear until now. Previous exact diagonalization studies[4,8] reveal an exponential decay of the spin-spin and dimer-dimer correlation functions. So the point of view that KAF is a spin liquid is widely accepted now.[2,4–12] It seems the best candidate for a description of KAF low-energy properties is a quantum dimer model.[6,9,13] It should be mentioned that there has been a certain recent success in this field. In Ref. 14 an approach first pioneered by Subrahmanyam in Ref. 15 was developed in which a spin-$\frac{1}{2}$ kagomé lattice is considered as a set of interactive triangles with a spin in each apex. It was suggested there to work in the subspace where the total spin of each triangle is 1/2 [short-range resonant valance bond states (SRRVB)] investigating low-lying excitations. It was shown that the low-energy spectrum obtained with SRRVB on the samples with up to 36 cites and the number of singlet excitations in the band coincide with the results of exact diagonalization. Meanwhile, a further development of this approach is required to get a full physical description of KAF.

Other types of frustrated magnets which possess a similar behavior as KAF and have many singlet states inside the triplet gap are pyrochlore[16] and CaV_4O_9.[17] Recently a model of frustrated antiferromagnets was suggested, the low-energy properties of which can be generic for these compounds as well as for KAF.[18] Weakly interactive plaquettes in the square lattice were considered there. Each plaquette has two almost degenerate singlet ground states, so a band of singlet excitations arises if the interplaquette interaction is taken into account. It is shown that there is a quantum phase transition in the model at a critical value of frustration separating a disorder plaquette phase and a columnar dimer one. In the proximity of this transition the specific heat has a low-temperature peak below which it possesses a power low-temperature dependence.

In this paper we show that such a picture is relevant for spin-$\frac{1}{2}$ KAF. It is proposed to consider a kagomé lattice as a set of stars with 12 spins arranged in a triangular lattice (see Fig. 1). A star has two degenerate singlet ground states. Interaction between stars leads to a band of low-lying excitations the number of which increases as $2^{N/12}\approx 1.06^N$. It is demonstrated that this interaction can be considered as a perturbation in the low-energy sector. As a result we get a model of the Ising ferromagnet in an effective magnetic field where degenerate states of the stars are described in terms of two projections of pseudospins 1/2. So it is shown that in contrast to the common view there is a hidden long-range order in KAF which consists of definite singlet states of stars.

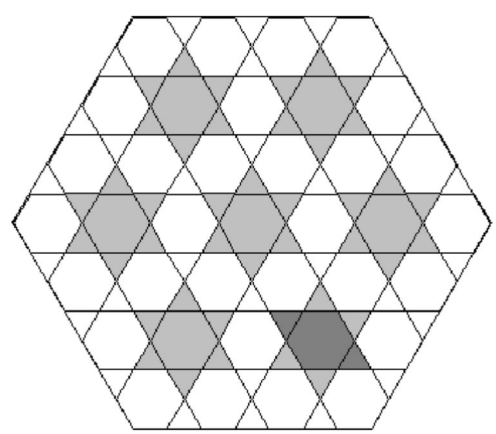

FIG. 1. Kagomé lattice (KL). There is a spin in each lattice site. The KL can be considered as a set of stars arranged in a triangular lattice. Each star contains 12 spins. A unit cell is also presented (dark region). There are four unit cells per star.

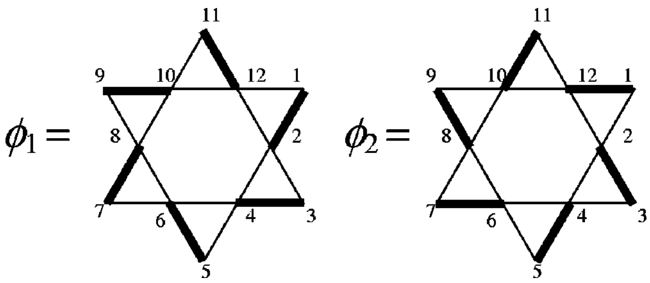

FIG. 2. Schematic representation of a star's two singlet ground-state wave functions ϕ_1 and ϕ_2. A bold line denotes the singlet state of two neighboring spins, i.e., $(|\uparrow\rangle_i|\downarrow\rangle_j - |\downarrow\rangle_i|\uparrow\rangle_j)/\sqrt{2}$.

In fact our approach is close in spirit to that proposed in Refs. 14 and 15 above discussed. The main advantage of a star over a triangle as a starting point to consider KAF is that it explicitly leads to an effective Hamiltonian which describes the singlet low-temperature dynamics.

We start with the Hamiltonian of the spin-$\frac{1}{2}$ kagomé Heisenberg antiferromagnet:

$$\mathcal{H}_0 = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j + J_2 \sum_{(i,j)} \mathbf{S}_i \mathbf{S}_j , \qquad (1)$$

where $\langle i,j \rangle$ and (i,j) denote nearest and next-nearest neighbors on the kagomé lattice, respectively, shown in Fig. 1. The case of $|J_2| \ll J_1$ is considered in this paper. We discuss a possibility of both signs of next-nearest-neighbor interactions—a ferromagnetic and an antiferromagnetic one. As shown below, in spite of the smallness the second term in Eq. (1) can be of importance for the low-energy properties.

A kagomé lattice can be presented as a set of stars arranged in a triangular lattice (see Fig. 1). To begin with we neglect the interaction between stars and put $J_2 = 0$ in Eq. (1). A star is a system of 12 spins. Let us consider its properties in detail. The symmetry group contains six rotations and reflections with respect to six lines passing through the center. There are two degenerate singlet ground states ϕ_1 and ϕ_2 which differ each other by symmetry. Their wave functions are shown schematically in Fig. 2 where a bold line represents the singlet state of the corresponding two spins, i.e., $(|\uparrow\rangle_i|\downarrow\rangle_j - |\downarrow\rangle_i|\uparrow\rangle_j)/\sqrt{2}$. Evidently ϕ_1 and ϕ_2 are invariant with respect to rotations of the star and they transform into each other under reflections. They contain six singlets each having energy $-S(S+1) = -3/4 J_1$. One can show that the interaction between singlets does not contribute to the energy of the ground states which is consequently equal to $-4.5 J_1$. We have obtained numerically that there is a gap of the value of approximately $0.26 J_1$ which separates the ground states and the lower triplet levels in the star.

It should be pointed out that ϕ_1 and ϕ_2 are not orthogonal: the scalar product of these two functions is $(\phi_1 \phi_2) = 1/32$. So in the following we will use two orthonormalized combinations

$$\Psi_1 = \frac{1}{\sqrt{2+1/16}} (\phi_1 + \phi_2), \qquad (2)$$

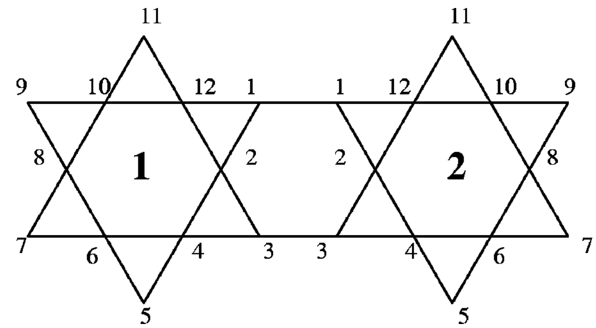

FIG. 3. There are the following interactions between each two stars: $V = J_1(\mathbf{S}_1^{(1)}\mathbf{S}_1^{(2)} + \mathbf{S}_3^{(1)}\mathbf{S}_3^{(2)})$ and $\tilde{V}_1 = J_2(\mathbf{S}_1^{(1)}\mathbf{S}_2^{(2)} + \mathbf{S}_2^{(1)}\mathbf{S}_1^{(2)} + \mathbf{S}_2^{(1)}\mathbf{S}_3^{(2)} + \mathbf{S}_3^{(1)}\mathbf{S}_2^{(2)})$, where upper indexes label the stars.

$$\Psi_2 = \frac{1}{\sqrt{2-1/16}} (\phi_1 - \phi_2). \qquad (3)$$

Because ϕ_1 and ϕ_2 are invariant under rotations and transform into each other with reflections, Ψ_1 is invariant under all symmetry group transformations and Ψ_2 is invariant under rotations and it changes sign with reflections.

We consider now the interaction between two nearest stars, still neglecting the second term in Eq. (1). Initially there are four degenerate ground states with energy $E_0 = -9 J_1$ and wave functions $\{\Psi_{n_1}^{(1)} \Psi_{n_2}^{(2)}\}$ $(n_i = 1,2)$, where the upper index labels the stars. As is seen from Fig. 3, the interaction energy has the form

$$V = J_1(\mathbf{S}_1^{(1)}\mathbf{S}_1^{(2)} + \mathbf{S}_3^{(1)}\mathbf{S}_3^{(2)}). \qquad (4)$$

Let us consider V as a perturbation. According to the standard theory[19] one has to solve a secular equation to find the first correction to the energy. In our case there are four equations and the corresponding matrix elements are given by

$$H_{n_1 n_2; k_1 k_2} = V_{n_1 n_2; k_1 k_2} + \sum_{m_1, m_2} \frac{V_{n_1 n_2; m_1 m_2} V_{m_1 m_2; k_1 k_2}}{E_0 - E_{m_1} - E_{m_2}}, \qquad (5)$$

where $V_{n_1 n_2; k_1 k_2} = \langle \Psi_{n_1}^{(1)} \Psi_{n_2}^{(2)} | V | \Psi_{k_1}^{(1)} \Psi_{k_2}^{(2)} \rangle$, $n_i, k_i = 1,2$, and indexes m_1 and m_2 denote excited levels of the first and second stars, respectively. Obviously the first term in Eq. (5) is zero and the second one can be represented as follows:

$$H_{n_1 n_2; k_1 k_2} = -i \int_0^\infty dt e^{-\delta t + i E_0 t} \times \langle \Psi_{n_1}^{(1)} \Psi_{n_2}^{(2)} | V e^{-it(H_{01}+H_{02})} V | \Psi_{k_1}^{(1)} \Psi_{k_2}^{(2)} \rangle, \qquad (6)$$

where H_{0i} are Hamiltonians of the corresponding stars. Using the symmetry of the functions Ψ_1 and Ψ_2 discussed above one can show that only elements of the first and second diagonals (i.e., with $n_1 = k_1$, $n_2 = k_2$ and with $n_1 \neq k_1$, $n_2 \neq k_2$) in Eq. (6) are nonzero. We have calculated them with a very high precision by expansion of the operator exponent up to a power of 150. The results for the elements of the first diagonal can be represented in the following form:

$$H_{11;11}=-a_1+a_2-a_3, \tag{7a}$$

$$H_{12;12}=-a_1+a_3, \tag{7b}$$

$$H_{21;21}=-a_1+a_3, \tag{7c}$$

$$H_{22;22}=-a_1-a_2-a_3, \tag{7d}$$

where $a_1=0.256J_1$, $a_2=0.015J_1$, and $a_3=0.0017J_1$. Terms of the second diagonal are much more less than a_2 and a_3 and can be neglected. So the interaction shifts all the levels on the value $-a_1$ and lifts their degeneracy. The constants a_2 and a_3 in Eqs. (7) determine the level splittings. All corrections are small enough and one can consider the interaction, Eq. (4), between stars as a perturbation at low energies. We restrict ourself to this precision here and do not consider triplet states.

So KAF appears to be a set of two-level interacting systems and one can naturally represent the low-energy singlet sector of Hilbert space in terms of pseudospins: $|\uparrow\rangle=\Psi_2$ and $|\downarrow\rangle=\Psi_1$. It is seen from Eqs. (7) that in these terms the interaction between stars is described by the Hamiltonian of Ising ferromagnet in an external magnetic field:

$$\mathcal{H}=-\mathcal{J}\sum_{\langle i,j\rangle} s_i^z s_j^z - h\sum_i s_i^z, \tag{8}$$

where $\langle i,j\rangle$ labels now nearest-neighbor pseudospins, arranged in a triangular lattice formed by the stars, $\mathcal{J}=4a_3=0.007J_1$, and $h=6a_2=0.092J_1$. We also omit a constant in Eq. (8) which is equal to $-0.439J_1N$. It should be stressed that within our precision the Hamiltonian, Eq. (8), is an exact mapping of the original Heisenberg model in the low-energy sector (excitation energy $\omega\sim\mathcal{J}$). So one can see from Eq. (8) that the ground state of KAF is that with all stars in Ψ_2 states. The energy of the ground state, $-0.443J_1N$, is close to the value $-0.438J_1N$ obtained in Ref. 5.

In fact we show the existence of a long-range order in KAF generated by singlets. This hidden order settles on the triangular star lattice and can be checked by inelastic neutron scattering: the corresponding intensity for the singlet-triplet transitions should have a periodicity in reciprocal space corresponding to the star lattice. This picture is similar to the one observed in the case of the dimerized spin-Peirls compound $CuGeO_3$.[20]

We proceed with a discussion of the number of low-energy states in KAF. As each star has two singlet ground states and contains 12 spins the number of singlet excitations in the band is given by $2^{N/12}\approx 1.06^N$. Unfortunately there is no point to compare this result with that of the previous numerical works[2,5] discussed above because a very small samples ($N\leqslant 36$) were considered there. Moreover, our scaling of the singlet states inside the gap could change because of singlets of the higher energies not discussed here.

Let us take into account the next-nearest-neighbor interactions. As seen from Fig. 3 they can be divided into three parts $\tilde{V}_1$, $\tilde{V}_2^{(1)}$, and $\tilde{V}_2^{(2)}$, where $\tilde{V}_1=J_2(\mathbf{S}_1^{(1)}\mathbf{S}_2^{(2)}+\mathbf{S}_2^{(1)}\mathbf{S}_1^{(2)}+\mathbf{S}_2^{(1)}\mathbf{S}_3^{(2)}+\mathbf{S}_3^{(1)}\mathbf{S}_2^{(2)})$ contributes to the interstar interaction and $\tilde{V}_2^{(1)}$ and $\tilde{V}_2^{(2)}$ contain 12 intrinsic next-nearest-neighbor interactions of the first and second stars, respectively. Considering now perturbation theory according to a sum of these three operators and that given by Eq. (4) we find that in addition to the corrections presented in Eqs. (7) there are new ones proportional to J_2 from the first and second terms in Eq. (5) given by $\tilde{V}_2^{(1)}$, $\tilde{V}_2^{(2)}$, and $\tilde{V}_1$, respectively. Using the symmetry of functions Ψ_1 and Ψ_2 it can be shown that the secular matrix, Eq. (6), does not change its two-diagonal structure in this case. Calculations give that terms of the second diagonal can be neglected at $|J_2/J_1|\ll 1$ and we have for the values of "exchange" and "magnetic field" in the effective Hamiltonian, Eq. (8),

$$\mathcal{J}=0.007J_1-0.002J_2, \tag{9}$$

$$h=0.092J_1+0.781J_2. \tag{10}$$

One can see from Eqs. (9) and (10) that the next-nearest interactions give a correction of the order of $|J_2|/J_1\ll 1$ to the value $\mathcal{J}$ and their contribution to the "magnetic field" is significant if $|J_2|\gtrsim 0.01J_1$. If $J_2<0$ (ferromagnet interaction), they could even change the sign of h. In the case of $h>0$ the ground state of the kagomé lattice is that with all stars in Ψ_2 states and if $h<0$ all stars are in Ψ_1 states.

The effect of the next-nearest ferromagnetic coupling was previously studied numerically on finite clusters ($N\leqslant 27$) in Ref. 2. It was shown there that at $|J_2|/J_1\sim 1$ the ground state has $\sqrt{3}\times\sqrt{3}$ magnetic structure. At $|J_2|/J_1\ll 1$ the ground state was found to be disorder and there is a band of singlet excitations inside the triplet gap. So the singlet long-range order appears to be unstable with respect to large $J_2<0$.

We could expect a logarithmic singularity of the specific heat in the point $h=0$ at the critical temperature T_c which is of the order of $\mathcal{J}$ and there should be a peak at $T\sim\mathcal{J}$ if $h\neq 0$. The specific heat decreases at $T\to 0$ as $e^{-(3\mathcal{J}+|h|)/T}$. So we do not get the low-temperature behavior $C\propto T^2$ obtained in experiments.[1] It should be noted that such a law should exist if the energy of the low-lying excitations $\epsilon_{\mathbf{q}}$ with wave vector $\mathbf{q}$ at $q\ll 1$ has the form $\epsilon_{\mathbf{q}}=\sqrt{(cq)^2+\Delta^2}$. Within the perturbation theory considered in this paper the interaction between stars is described by the Hamiltonian, Eq. (8), of an Ising ferromagnet in a magnetic field which does not imply

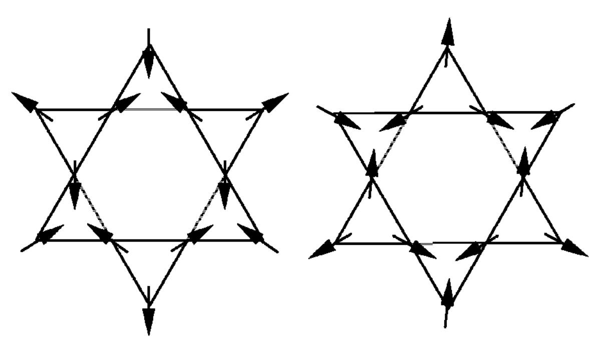

FIG. 4. Ground states of a star in the case of classical spins.

such a behavior of the low-energy spectrum. But the further orders could give some kind of anisotropy in Eq. (8) which leads to the necessary picture. This point will be considered in detail elsewhere.

It is appropriate to mention here a recent experiment on $Cu_3V_2O_7(OH)_2 \cdot 2H_2O$ (Ref. 21) which is the only candidate for spin-$\frac{1}{2}$ kagomé material by now. In spite of a strong exchange $J_1 \sim 100$ K in this compound, specific heat anomalies were not observed up to the temperature 1.8 K. At the same time for KAF's with larger spin value they were seen at larger T.[1] This apparent discrepancy can be explained if we take into account that according to our results for $S=1/2$ the low-temperature scale is defined by the energy $0.01J_1$. Hence we wait for specific heat anomalies in the considered case or some kind of transition connected with the weak anisotropic interaction to be observed at lower T.

Let us discuss the case of large S. As is obtained in Ref. 22, there is a long-range singlet order in KAF with classical spins, whereas a precise picture of the ground state has not been obtained. The ground states of a star in this case are presented in Fig. 4, so our method cannot be expanded explicitly on large S. Meanwhile, because the low-energy physics in KAF is determined by singlets, most likely our consideration is relevant for small spins only. This point will be considered in detail elsewhere.

In conclusion, we presented an insight into the low-energy physics of spin-$\frac{1}{2}$ kagomé antiferromagnets. The lattice can be presented as a set of stars which are arranged in a triangular lattice and contain 12 spins (see Fig. 1). Each star has two degenerate singlet ground states with different symmetry. It is shown that the interaction between the stars leads to a band of singlet excitations and can be considered as a perturbation in the low-energy sector. We demonstrate the existence of a long-range order in KAF on the triangular star lattice which is generated by singlets and can be detected in particular in experiments on inelastic neutron scattering.

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