Hidden long range order in Heisenberg Kagomé antiferromagnets

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

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In spite of numerous theoretical and experimental studies in the last decade, some magnetic properties of Kagomé antiferromagnets (KAFs) remain open problems. Experiments revealed unusual low-temperature behavior of the specific heat and magnetic susceptibility in Kagomé-like compounds. For example specific heat behavior of the specific heat and magnetic susceptibility in Kagomé antiferromagnets (KAFs) remain open problems. Experiments revealed unusual low-temperature behavior of the specific heat and magnetic susceptibility in Kagomé-like compounds. For example specific heat $C$ measurements in $\text{SrCr}_9\text{Ga}_{12-9p}\text{O}_{19}$ ($S = 3/2$ 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.

Some of the experimental findings are in agreement with the results of numerical finite cluster investigations. 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 $\alpha^N$. For samples with up to 36 sites $\alpha = 1.15$ and 1.18 for the even and odd $N$, respectively. This wealth of low-lying singlet excitations explains the peak of specific heat at low temperature and its independence of the magnetic field.

However the origin of this band as well as the nature of the ground state were unclear until now. Previous exact diagonalization studies reveal 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. It seems the best candidate for description of KAF low-energy properties is a quantum dimer model. It should be mentioned a certain recent success in this field. In the paper a spin 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 RVB states (SR-RVB)) investigating low-lying excitations. It was shown that 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.

Another type of frustrated magnets which possess a similar behavior as KAF and have many singlet states inside the triplet gap are pyrochlore and $\text{CuV}_2\text{O}_9$. Recently it was suggested a model of frustrated antiferromagnet which low-energy properties can be generic for these compounds as well as for KAF. 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 inter-plaquette 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 behavior is do relevant for spin 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 the band of low-lying excitations which number 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 effective magnetic field where these degenerate states 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 the stars. This picture should be relevant also for KAFs with larger values of spin.

We start with the Hamiltonian of the spin 1/2 Kagomé Heisenberg antiferromagnet:

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

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

Kagomé lattice can be presented as a set of stars arranged in a triangular lattice (see Fig. 1). To begin with the low-energy properties. The symmetry group contains 6 rotations and reflections with respect to 6 lines passing through the center. There are two degenerate singlet ground states \( \phi_1 \) and \( \phi_2 \) 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 second term in Eq. (1). Initially there are four degenerate ground states with energy \( E_0 = -9J_1 \) and wave functions \( \{ \Psi^{(1)}_n \Psi^{(2)}_n \} \) (\( n_i = 1, 2 \)), where upper index labels the stars. As it is seen from Fig. 2, the interaction energy has the form:

\[
V = J_1 (S^{(1)}_1 S^{(2)}_1 + S^{(1)}_3 S^{(2)}_3). \tag{4}
\]

Let us consider \( V \) as a perturbation. According to the standard theory [18] one have 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

\[
H_{n_1,n_2;k_1,k_2} = V_{n_1,n_2;k_1,k_2} + \sum_{m_1,m_2} V_{n_1,n_2;m_1,m_2} V_{m_1,m_2;k_1,k_2}, \tag{5}
\]

where \( V_{n_1,n_2;k_1,k_2} = \langle \Psi^{(1)}_n | V | \Psi^{(2)}_k \rangle, \ n_i, k_i = 1, 2 \) and indexes \( m_1 \) and \( m_2 \) denote excited levels of the first and the second star, respectively. Obviously the first term in Eq. (3) is zero and the second one can be represented as follows:

\[
H_{n_1,n_2;k_1,k_2} = -t \int_0^\infty dt e^{-it} e^{it H_{01} + H_{02}} V \langle \Psi^{(1)}_n | \Psi^{(2)}_k \rangle, \tag{6}
\]

where \( H_{0i} \) are Hamiltonians of the corresponding stars. Using the symmetry of the functions \( \phi_1, \phi_2, \phi_1 \) and \( \phi_2 \) discussed above one can show that only diagonal elements (i.e. \( n_1 = k_1, n_2 = k_2 \)) in Eq. (6) are nonzero. We have calculated them with a very high precision by expansion of the operator exponent up to the power 130. The results 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 \(a_3 = 0.0027J_1\). So the interaction shifts all the levels on the value \(-a_1\) and lifts their degeneracy. Constants \(a_2\) and \(a_3\) in Eqs. (8) determine the levels splitting. All corrections are small enough and one can consider interaction Eq. (8) between stars as a perturbation at low energies. We restrict ourselves with this precision here and don’t consider triplet states.

So KAF appears to be a set of two-levels 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. (8) that in these terms the interaction between stars is described by the Hamiltonian of Ising ferromagnet in the external magnetic field:

\[
\mathcal{H} = -\mathcal{J} \sum_{(i,j)} s_i^z s_j^z - h \sum_i s_i^z, \tag{8}
\]

where \((i,j)\) labels nearest-neighbor pseudospins, arranged in a triangular lattice formed by the stars, \(\mathcal{J} = 4a_3 = 0.011J_1\) 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 Hamiltonian Eq. (8) is an exact mapping of the original Heisenberg model in the low-energy sector (excitation energy \(\omega \approx \mathcal{J}\)). So one can see from Eq. (8) that the ground state of KAF is that with all the stars in \(\Psi_2\) states.

In fact we show 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: corresponding intensity for the singlet-triplet transitions should have periodicity in the reciprocal space corresponding to the original star lattice. This picture is similar to observed one in the case of the dimerised spin-Pairls compound \(\text{CuGeO}_3\) [17]. Because low-energy physics in KAF is determined by singlets, our consideration should be relevant qualitatively also for KAFs with the larger values of spin.

We proceed with the discussion of the number of low-energy states in KAF. As each star has two singlet ground states, the Hamiltonian (8) gives for the values of “exchange” and “magnetic field” the effective Hamiltonian Eq. (8):

\[
\mathcal{J} = 0.011J_1 - 0.005J_2, \tag{9}
\]

\[
h = 0.092J_1 + 3.635J_2. \tag{10}
\]

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 considerable 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 \(\Psi_2\) states and if \(h < 0\) all stars are in \(\Psi_1\) states.

We could expect a logarithmic singularity of the specific heat \(C\) 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 \(h \neq 0\). Specific heat decreases at \(T \rightarrow 0\) as \(e^{-|\mathcal{J}|/\hbar T}\). So we don’t get 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 low-lying excitations \(\epsilon_q\) with wave vector \(q\) at \(q \ll 1\) has the form \(\epsilon_q = cq^2 + \Delta\). Within the first order of perturbation theory considered in this paper there are contributions to the inter-stars interaction and \(\tilde{V}_2(1)\) and \(\tilde{V}_2(2)\) contain 12 intrinsic next-nearest-neighbor interactions of the first and the second star, respectively. Considering now perturbation theory according to a sum of these three operators and that given by Eq. (8) we find that in addition to corrections presented in Eqs. (8) there are new ones proportional to \(J_2\) from the first and the second terms in Eq. (8), given by \(\tilde{V}_1(1), \tilde{V}_2(2)\) and \(\tilde{V}_1\), respectively. Using symmetry of functions \(\phi_1\) and \(\phi_2\) it can be shown that secular matrix Eq. (8) remains diagonal in this case. As a result calculations give for the values of “exchange” and “magnetic field” in the effective Hamiltonian Eq. (8):

\[
\mathcal{J} = 0.011J_1 - 0.005J_2, \tag{9}
\]

\[
h = 0.092J_1 + 3.635J_2. \tag{10}
\]

It is appropriate to mention here a recent experiment on \(\text{Cu}_3\text{V}_2\text{O}_7(\text{OH})_2 \cdot 2\text{H}_2\text{O}\) which is the only candidate for spin-1/2 Kagomé material by now. In spite of strong exchange \(J_1 \sim 100\text{ K}\) in this compound, there is no regime obtained for KAF with the larger values of spins has been achieved up to the temperature 1.8 K. In this respect we point here on a small scale of the dynamics in spin-1/2 KAF. According to our results the representa-
tive temperature is of the order of $0.01J_1$, so the region $T \lesssim 1$ K should be attained for this material.

In conclusion, we present a new insight of low-energy physics of spin $1/2$ Kagomé antiferromagnet (KAF). 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 interaction between the stars leads to the 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. This physical picture should be relevant also for KAFs with larger spin values.

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