Excited states and the thermodynamics of a fully frustrated quantum spin chain

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A one-dimensional fully frustrated spin system called a Δ chain is considered and its excited states and the specific heat are investigated. As in the Heisenberg antiferromagnet on the *kagomé* lattice, the classical ground state of the system has infinite continuous degeneracies. Numerical studies of finite systems show that the low-lying excited states have an almost dispersionless spectrum near a small energy gap. As a result the specific heat exhibits a two-peak structure, one of which arises from these low-lying excited states.

Recent experiments on ^{3}He adsorbed on graphite[1] and also on highly frustrated compounds such as $SrCr_8Ga_4O_{19}$ (Ref. 2) induced theoretical interests on the fully frustrated quantum spin systems.[3-10] By the term "a fully frustrated quantum spin system," I mean a system whose classical ground state has infinite continuous degeneracies. A well-known example of such a system is the antiferromagnetic Heisenberg model (AFHM) on the *kagomé* lattice. As was discussed by Harris, Kallin, and Berlinsky[6] in detail, if one considers a typical ordered ground state of classical AFHM on the *kagomé* lattice, continuous local distortions of the spin configuration from the ground state are possible without changing the total energy. As they are local, the degeneracy of the ground state is somehow proportional to C^{N_S} where N_S is the number of sites, though we cannot define a finite constant C as the spins have continuous degree of freedom in the classical limit. If we consider the corresponding quantum system, the linear spin-wave theory leads to dispersionless zero energy modes. This kind of full frustration is not restricted to two dimensions but exists also in three dimensions.[11,12] In general the AFHM on the line graphs of a bipartite graph possess this property.[13] It is a quite interesting and challenging problem to study what kind of ground state is realized in fully frustrated quantum spin systems. There have recently appeared several investigations on the ground state of the AFHM on the *kagomé* lattice. Whether its ground state has a long-range order or not is still an open question. It was argued recently that for large S system, the zero energy spin-wave modes acquire finite dispersions through the spin-wave interaction and the existence of the long-range magnetic order is plausible.[8,9] The small S system seems, however, a very good candidate for a ground state without magnetic long-range order.[10]

I consider in this paper a very simple one-dimensional quantum spin chain, called a Δ chain, which is also fully frustrated and investigate its low-lying excited states as well as its specific heat. It reveals that full frustration manifests itself in the dispersionless low-lying excitations as well as in a low-temperature peak of the specific heat.

The system considered is described by the Hamiltonian

$$H=\sum_{i=1}^{N} h_i \ , \tag{1}$$

where

$$h_i = S_{2i-1}S_{2i} + S_{2i}S_{2i+1} + S_{2i-1}S_{2i+1} \tag{2}$$

and S_i denotes the spin with size $\frac{1}{2}$ at the site i. The system is schematically shown in Fig. 1. In the classical approximation the ground state of the local Hamiltonian h_i is realized by the configuration of the three spins making the angle $2\pi/3$ to each other. If one assumes a ground state of the total system where all the spins are laid in a plane, one easily sees that three successive spins, i.e., S_{2i}, S_{2i+1}, and S_{2i+2} can be rotated simultaneously without raising the energy. The zero-temperature specific heat of the classical model is $\frac{3}{4}$ (we take as $k_B=1$ throughout this paper) per spin. This leads to the existence of two dispersionless zero modes in the linear spin-wave theory.

The ground state of the quantum system was considered by several authors previously.[14,15] It is easily obtained since the ground state of h_i is realized by pairing any two of its three spins into the singlet state. One can immediately construct the ground state of the total system from four independent ground states of h_i. Monti and Sütö showed rigorously:[15]

(1) The system with open boundary conditions ($2N+1$ spins) has $2(N+1)$-fold degenerate ground states with N singlet pairs and one free spin. Degeneracy arises from the freedom of position and the direction of the free spin.

(2) The system with periodic boundary conditions ($S_{2N+1}=S_1$) has doubly degenerate ground state with N singlet pairs.

(3) The excited states has a finite energy gap.

The ground states are essentially dimer states. The high degeneracy of the ground states for open boundary conditions already exhibits the effect of the full frustra-

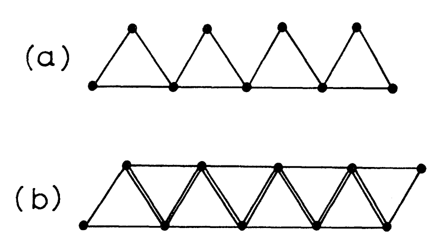

FIG. 1. The Δ chain (a) and the Majumdar-Ghosh model (b).

tion. The degeneracy is, however, proportional to the system size and does not give rise to a residual entropy at the zero temperature. The ground state of the periodic boundary systems are identical to those of Majumdar-Ghosh model (MGM)[16] whose Hamiltonian is given by

$$H=\sum_{i=1}^{N}(h_i+h_{i+1/2}) \tag{3}$$

(see Fig. 1). This model is not fully frustrated in the sense as mentioned above. We must look into excited states to understand the peculiarity of the thermodynamic property of the fully frustrated systems.

Low-lying excited states of finite systems with periodic boundary conditions have been obtained by numerical diagonalization of the Hamiltonian. The lowest excited state for each wave vector k is a triplet ($S=1$) state and its excitation energy δE_1 is shown in Fig. 2 as a function of k. The k dependence of δE_1 for finite systems is very weak and it decreases with increasing N. The minimum and the maximum excitation energies are realized at $k=\pi$ and $k=0$, respectively. The obtained spin correlation $C_{ij}=\langle S_iS_j\rangle$ in the excited states does not show systematic decrease with distance. For example, $|C_{2,2+2r}|$ in the $k=\pi$ state increases with r between $r=2$ and 4 for $N=10$ and it is 0.0135 at $r=5$. On the other hand, $|C_{1,1+2r}|$ decreases with r down to 0.00963 at $r=5$ and roughly proportional to $.75^{2r}$. In the $k=0$ state $|C_{1,1+2r}|$ has a minimum value 0.00448 at $r=3$ and increases again up to 0.00875 at $r=5$. This irregular behavior is showing that the lowest excited states are not strongly localized in a few atomic distances. Extrapolating δE_1 from $N=6$, 8, and 10 to the infinite N by a quadratic function of N^{-2}, I obtain $\delta E_1=0.219$ at $k=\pi$ and 0.218 at $k=0$. The present result implies that the lowest excitations of an infinite system are almost dispersionless with $\delta E_1\cong 0.22$. I have also calculated the second lowest excited state, which is singlet ($S=0$), as a function of k. Though the results of finite systems show a little larger dispersion compared to δE_1, it appears to converge to an almost dispersionless mode with the energy identical to the lowest excitation.

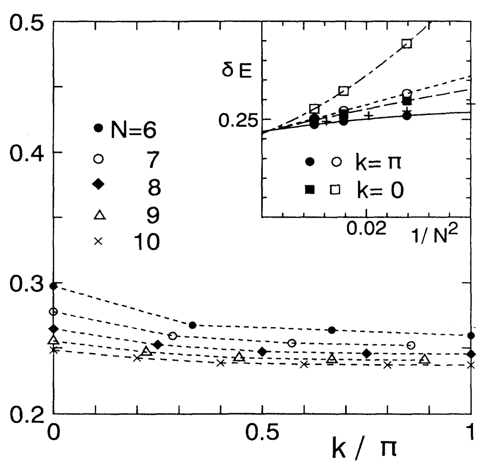

FIG. 2. The lowest excitation energy is shown as a function of the wave vector for various system sizes. System sizes are indicated in the figure. Lines are only to guide the eye. The inset displays the system size dependence of the excitation energy, where full (open) circles denotes the lowest (second lowest) excitation energies at $k=\pi$ and full (open) squares the lowest (second lowest) ones at $k=0$. Lines in the inset show quadratic fits in terms of N^{-2}. Crosses in the inset show the variational excitation energy $\delta E_{v,N}$.

One can construct a variational excited state which is defined by

$$\Psi_{n,i}=\prod_{j=1,i-1}\varphi_{s,j}\psi_{n,i}\prod_{j=i+n+1,N}\varphi_{s,j}\ ,$$

$\varphi_{s,j}$ is the singlet state of S_{2j-1} and S_{2j} and $\psi_{n,i}$ is a first excited state of the Hamiltonian of $2n$ spins, $H_{n,l}=\sum_{j=i,i+n-1}h_j+S_{2(i+n)-1}S_{2(i+n)}$. Let us denote the variational excitation energy of $\Psi_{n,i}$ by $\delta E_{v,n}$. The state for $n=1$ is nothing but the state where one of the dimers of the ground state is replaced by a triplet state and its excitation energy $\delta E_{v,1}$ is the unity. $\delta E_{v,2}$ is easily obtained as $\frac{1}{2}$. For larger n's $\delta E_{v,n}$ has been obtained numerically up to $n=10$ and is shown in Fig. 2. It is observed that $\psi_{n,i}$ is a triplet and an eigenstate of $S_{2i-1}+S_{2i}$ with $|S_{2i-1}+S_{2i}|=1$. The spin correlation $C_{i,j}$ in $\psi_{n,i}$, however, does not show simple behavior in terms of the distance. For example, $C_{1,2j}$ in $\psi_{10,1}$ is 0.02386 for $j=1$ and increases with j with a maximum 0.05052 at $j=5$ and then decreases to 0.00419 at $j=10$. $C_{2,2j}$ shows a similar behavior. So the correlation between S_1 for S_2 and those at the center of the cluster S_{10} or S_{12} is not negligible. Otherwise correlations between far apart spins are weak. It is interesting that if we construct the variational state with wave vector k, i.e., $\Psi_{n,k}=\sum_j\Psi_{n,j}e^{ikj}$, the excitation energy is $\delta E_{v,n}$ independent of k. This is easily deduced from the fact that $\psi_{n,i}$ is an triplet eigenstate of $S_{2i-1}+S_{2i}$.

Above features of excitation spectrum are quite different from those of MGM which has equivalent ground states. The lowest excitation energy of a finite MGM ($N=6$) shows a larger dispersion with band width ~ 0.9 above an energy gap 0.638 for $N=6$.[17] It should be noted that the energy gap of the Δ chain is quite small compared to the unity, the excitation energy of the local triplet. The above result implies that excited states has a spatially coherent character which lowers the energy from that of a local triplet excitation as well as a localized character which leads to the absence of dispersion.

The peculiar feature of the excitation spectrum clarified above should influence the specific heat. I have calculated the specific heat by making use of the quantum transfer matrix method.[18] The transfer matrix has been obtained by applying the usual checkerboard decomposition to the density matrix; $e^{-\beta H}$ is approximated by $[e^{-\beta H_e/m}e^{-\beta H_o/m}]^m$ where H_e (H_o) is defined by taking the summation in H over only even (odd) i. The largest eigenvalue and its eigenvector of the transfer matrix have been calculated by employing the power method. Then the energy of the system is calculated by using the eigenvector and then the entropy by subtracting the energy from the free energy. So the specific heat can be obtained in two ways, i.e., by differentiating energy and entropy numerically. Both results agree quite well except for low temperatures. At $T<0.05$ they differ by several percent due to the finiteness of the Trotter slicing m. In Fig. 3 the overall behavior of the specific heat is shown as a function of the temperature. The specific heat exhibits two peaks. The broad peak at $T \cong 0.58$ is a common feature of the one-dimensional antiferomagnetic quantum spin systems.[19] The rather sharp peak at a lower temperature is a peculiarity of this system. The calculation has been done for $m=4 \sim 8$ and the low-temperature peak grows as well as shifts to the lower temperature with increasing m. This m dependence excludes the possibility that this peak is an artifact of the finite m effect, though the present result appears to be not yet converged. The peak position is at $T \cong 0.12$ for $m=8$ and the peak height is 0.18 per site. The entropy contributed by this peak is roughly estimated to be ~ 20 percent of the total entropy. This result is consistent with the excitation spectrum with $\sim 2^{0.4N}$ states accumulated at $\delta E \sim 0.2$. It is also intriguing that this rate is roughly equal to 1/4, the deficiency of the zero-temperature specific heat of the classical model.

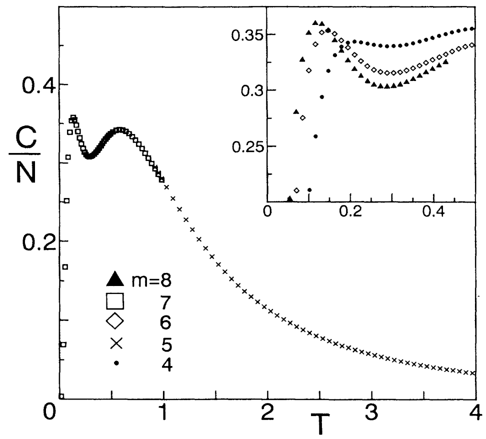

FIG. 3. The specific heat is shown as a function of the temperature. Data obtained for the number of Trotter slicing $m=5$ are used at $T<1.0$ and those for $m=7$ at $T<1.0$. The inset displays the dependence of the low-temperature peak on m. It is seen that the peak becomes sharper as m increases.

It has been shown above that the Δ chain has almost dispersionless excitation modes with a very small energy gap. From the present result one may conjecture that both the triplet and the singlet first excited states converge to the exactly dispersionless excited states with an identical energy in the thermodynamic limit. These excitations are interpreted as the zero energy spin-wave modes which have acquired a finite energy through quantum fluctuations.[9] It is interesting to study excited states with higher spins whether their energies also converge to the same limiting value as $N \rightarrow \infty$. These excitations lead to a sharp peak of the specific heat at a low temperature. The two-peak structure of the specific heat was discussed previously on the *kagomé* lattice in relation to the ^{3}He adsorbed layer.[3] Though the previously reported result of the twelve spin *kagomé* cluster now seems to be an artifact due to the finite size,[20] the full frustration can lead to a Schottky-type low-temperature peak in one dimension. Although the system considered here is only a very simple example, one might expect that the property exhibited here may realize in fully frustrated systems in higher dimensions if their ground states have no magnetic order and their excitations have energy gaps. So far, however, no fully frustrated system with exactly known (e.g., dimer) ground state has been constructed in higher dimensions than one. Whether the reported absence of the low-temperature peak in the specific heat of $S=\frac{1}{2}$ *kagomé* antiferromagnet[20] implies the existence of a magnetic order or not is still to be clarified. It will be quite interesting to investigate fully frustrated systems in two and three dimensions both in theoretical and experimental way to see whether these properties are really observed.

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