Zigzag spin chains with antiferromagnetic-ferromagnetic interactions: Transfer-matrix renormalization group study

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(Dated:)

Properties of the zigzag spin chains with various nearest-neighbor and next-nearest-neighbor interactions are studied by making use of the transfer-matrix renormalization group method. Thermodynamic quantities of the systems (temperature dependence of the susceptibility and the specific heat), as well as the field dependence of the magnetization are analyzed numerically with a high accuracy in the thermodynamic limit. The results have been compared with the recent experimental data on Rb$_2$Cu$_2$Mo$_3$O$_{12}$.

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I. INTRODUCTION

Spin models with charge degrees of freedom frozen, as an effective low-energy description for insulating systems, have been proved to be a fruitful resource of fundamental concepts and principles in condensed matter physics and other related fields. Besides theoretical explorations, advanced probe tools and measurements, combined with multifarious natural and artificial materials, such as ceramic or organic compounds, and optical lattices provide remarkable practical platforms for theoretical investigations. The simplest one-dimensional (1D) quantum spin model is to include the next-nearest-neighbor (NNN) interactions: 

\[ H = \sum_{i=1}^{N} (J_1 S_i \cdot S_{i+1} + J_2 S_i \cdot S_{i+2}) , \]  

(1)

where $S_i$ is the quantum spin operator. From Bethe’s seminal paper and successive works, we know that the ground state of a spin-$1/2$ antiferromagnetic (AF) ($J > 0$) chain is a singlet ($S_{\text{tot}} = 0$) and has quasi-long-range order with algebraically decaying spin correlations. The gapless spectrum contains no single-particle excitations and is instead a continuum of states. The elementary excitations are called spinons, which carry spin $1/2$. Furthermore, the spin system is gapped where $J_1$ or $J_2$ is NN or NNN exchange interaction constant, and periodic boundary conditions are implied. This is usually called the $J_1$-$J_2$ model, which can be also considered as a model for a zigzag spin chain, as shown in Fig. 1.

The $J_1$-$J_2$ model has been investigated theoretically over the decades. With $J_2 > 0$, it is a frustrated (competing) system, irrespective of the sign of $J_1$. When $J_1 > 0$ and $J_2 > 0$ (AF-AF), the ground state is a spin liquid. The increase of the ratio of the coupling constants $\alpha (\equiv J_2/J_1)$ induces an infinite-order phase transition from a gapless state to a gapful dimerized state. The critical point $\alpha_c$ is numerically estimated to be $\approx 0.241$. When $\alpha$ is further increased to the so-called Majumdar-Ghosh (MG) point at $1/2$, the ground state is the products of singlet pairs formed by nearest neighboring spins. It is two-fold degenerate since the $Z_2$ symmetry of translations by one site is spontaneously broken. When $J_1 < 0$ and $J_2 > 0$ (F-AF) with $-1/4 < \alpha < 0$, the ground state is fully ferromagnetic (FM), and becomes an ($S = 0$) incommensurate state for $\alpha < -1/4$. It is suggested that in this incommensurate state the gap is strongly suppressed. For $\alpha = -1/4$, the exact ground state can be shown to have a $(N + 2)$-fold degeneracy, comprising by $S = 0$ and $S = N/2$ states (with $N$ the lattice size). When $J_1 > 0$ and $J_2 < 0$ (AF-F), the system...
is believed to be in a gapless antiferromagnetic phase for any permissible values of $J_1$ and $J_2$.

There are many papers contributing to the AF-AF case and the related extended models. The other two cases (F-AF and AF-F), although having caught relatively less attention, also show very interesting phenomena, which we will mainly deal with in this paper. Aside from the general aspect of theoretical interest, especially for understanding the roles played by frustration and incommensurability, an additional motivation to study this system lies in the fact that physically, it is believed that a large class of copper oxides can be essentially described by the $J_1$-$J_2$ model.

Copper oxides are excellent model systems for low dimensional spin-$\frac{1}{2}$ quantum magnets, where magnetic Cu$^{2+}$ ions carry 1/2 spins. The basic building blocks are CuO$_2$ plaquettes and there are three ways of linking these fundamental units. One is adjacent squares sharing their corners, as shown in (a) of Fig. 2. A typical example is Sr$_2$CuO$_3$. These corner-sharing chains can expand in the plane to form a CuO$_2$ sheet, constituting the basic structure in cuprates. The dominant interaction in a corner-sharing chain is the NN superexchange. Linear Cu-O-Cu bonds along the spin chains give rise to a large antiferromagnetic NN exchange coupling. As in Sr$_2$CuO$_3$, the NN coupling constant is estimated to be 2100 ± 200 K. The second kind is edge-sharing, as shown in Fig. 2 (b). Because of the nearly 90° Cu-O-Cu bond in the edge-sharing squares, an O2$p_x$ orbital hybridizing with a 3$d$ orbital of Cu ion is almost orthogonal to that of the next Cu ion. The NN interaction $J_1$ can vary from antiferromagnetic to ferromagnetic, as the angle $\theta$ of the Cu-O-Cu bond approaches 90° from a larger value. This nearly orthogonality makes the NN coupling in the edge-sharing case more than an order of magnitude smaller than the corner-sharing. The sign and the absolute value of the NN interaction depend sensitively on the bond angle $\theta$ and the distance between copper and oxygen ions. We refer readers to Table I of Ref. [14] for details. The third configuration is the combination of corner-sharing and edge-sharing in one spin chain simultaneously, such as in SrCuO$_3$, as shown in Fig. 2 (d). In SrCuO$_2$, the NNN coupling, which is the superexchange interaction through the linear Cu-O-Cu, is almost ten times greater than the NN one through sharing the edges.

In corner-sharing chains, the NNN interaction can often be neglected safely due to its small magnitude relative to the NN interaction. For the edge-sharing case, the situation is quite different. The NNN interaction $J_2$ through the Cu-O-O-Cu path is generally antiferromagnetic, usually with a magnitude of a few tens Kelvin. Despite its smallness, $J_2$ has a pronounced effect on the physical properties of these systems since the NN coupling is also small. Therefore, edge-sharing copper oxides provide abundant experimental materials for studying the zigzag spin chain model, which can cover a large region of the parameter space.

On the other hand, at low temperatures, besides intrachain couplings $J_1$ and $J_2$, other forms of interactions often become relevant, driving a system to various phases with the decrease of temperature. For example, spin-phonon interactions can induce a spin-Peierls instability, as in CuGeO$_3$. If interchain interactions are strong enough, an antiferromagnetically long-ranged order will appear below the Néel temperature $T_N$, as in Ca$_2$Y$_2$Cu$_5$O$_{10}$, La$_6$Cu$_8$Cu$_2$O$_{14}$, and Li$_2$CuO$_2$. Another interesting case is LiCu$_2$O$_2$ which undergoes a transition to a magnetic helix state at low temperatures. By a comparative study on the similar oxide NaCu$_2$O$_2$, it has been shown that interchain interactions over a few chains should be incorporated to explain the experimental results. These facts reveal the complexity of the interactions underlying edge-sharing copper oxides at low temperatures.

Since the effect of NNN interaction is important for edge-sharing copper oxide chains, the study on the $J_1$-$J_2$ model not only has its own theoretical meaning, but also obtains a connection with practical materials. In this paper, in the thermodynamic limit and extending to the low-temperature region, we use the transfer-matrix renormalization group (TMRG) method to study the thermodynamic properties of the $J_1$-$J_2$ model with antiferromagnetic-ferromagnetic interactions.

The TMRG is a powerful numerical tool for studying the thermodynamic properties of 1D quantum systems. It starts by expressing the partition function as a trace on the product of the transfer matrix $T_M$ using the Trotter-Suzuki decomposition.

$$Z = \text{Tr} e^{-\beta H} = \lim_{M \to \infty} \text{Tr} T_M^{N/2},$$

where $M$ is the Trotter number, and $N$ is the total cell number in the lattice, $\tau = \beta/M$. The definition of $T_M$ can be found from Refs. [22, 23, 24]. For the $J_1$-$J_2$ model
considered here, each cell consists of two adjoining spins. In the thermodynamic limit $N \to \infty$, the free energy $f$, internal energy $u$, and uniform magnetization $m_z$ can be expressed by the maximum eigenvalue $\lambda_{\text{max}}$ and the corresponding left $|\psi^L\rangle$ and right $|\psi^R\rangle$ eigenvectors of the transfer matrix $T_M$:

$$f = -\lim_{N \to \infty} \frac{1}{N\beta} \ln Z = -\frac{1}{2\beta} \lim_{\lambda \to \infty} \ln \lambda_{\text{max}},$$

$$u = \frac{\langle \psi^L | \tilde{T}_U | \psi^R \rangle}{\lambda_{\text{max}}},$$

$$m_z = \frac{\langle \psi^L | \tilde{T}_M | \psi^R \rangle}{\lambda_{\text{max}}},$$

where the definition of the transfer matrices $\tilde{T}_U$ and $\tilde{T}_M$, which are similar to $T_M$, can also be found in Refs. 22–24. The specific heat and magnetic susceptibility can then be calculated by numerical derivatives of $u$ and $m_z$, respectively,

$$C = \frac{\partial u}{\partial T},$$

$$\chi = \frac{\partial m_z}{\partial H}.$$
can not detect it. 

Another closely related topic worth mentioning is the incommensurability in the zigzag spin chains. For simple antiferromagnetic Heisenberg spin-1/2 chains and ladders, the well known mechanisms for generating incommensurabilities are via external magnetic fields or Dzyaloshinskii-Moria interaction. While in the zigzag spin chain, it has been found that the frustrated interaction \( J_2 > 0 \) can also produce incommensurability\[32,33\].

In classical picture, by regarding spin operators \( \mathbf{S}_i \) as classical vectors, the energy per site can be expressed as

\[
E(\theta) = J_1 \cos \theta + J_2 \cos 2\theta
\]

for Hamiltonian \[2\]. The pitch angle is given by

\[
\cos \theta = -J_1/4J_2
\]

for \(|\alpha| = |J_2/J_1| \geq 1/4\). As \( J_1 > 4J_2 \geq 0, \theta = \pi; -J_1 > 4J_2 \geq 0, \theta = 0 \). At the special point \( J_1 = 0, \theta = \pi/2 \), and the deviations from the normal values (\( \pi \) for the AF-AF case and \( 0 \) for the F-AF) begin to occur at \(|\alpha| = 1/4\).

In quantum level, the characteristic momentum \( Q \) of the spin-spin correlation function which maximizes the static structure factor is either \( \pi \) or \( 0 \), corresponding to the antiferromagnetic or ferromagnetic spin chain with \( J_2 = 0 \), respectively. With the increase of \( J_2, Q \) departs from its usual value. Numerical simulation\[32,33\] display that for the AF-AF case, \( Q \) deviates from \( \pi \) with the increase of \( J_2 \) after crossing the MG point \( (J_2 = J_1/2) \) where the dimerization has already taken place. On the other side, the departure of \( Q \) from \( 0 \) sets in at the critical point \( J_2 = -J_1/4 \) for the F-AF case. This asymmetry may be understood on account of the effects of the current-current interaction and the twist term.

It was shown that in the presence of exchange anisotropic, the twist term induces incommensurabilities in the spin correlations\[22\]. We have reason to expect this to hold true even in the SU(2) symmetric (i.e. isotropic) case\[22,32\]. In the AF-AF isotropic case, both the twist term and the current-current interaction diverge simultaneously to reach the strong coupling phase as the RG flows to \( J_2 \gg J_1 \). The pure current-current interaction induces massive spinons which can be regarded as quantum dimerization kinks, driving the zigzag spin chain into the dimerized phase. The twist term appearing in the zigzag case is merely to shift the minimum of the two-spinon continua to incommensurate wave numbers, which does not alter the massive spinon picture qualitatively\[32\].

If these two terms become relevant at different points, we may observe the emergence of dimerization and incommensurability one after the other, as in the AF-AF case. For the F-AF case, the current-current interaction is marginally irrelevant since it renormalizes logarithmically to zero\[22\]. The twist term becomes dominant. We can observe the incommensurability only, and hardly detect the gap following with the dimerization. But there is not a simple way to separate the effects of the current-current and twist interactions in the isotropic zigzag spin chains as in the anisotropic ones. There is still much theoretical work to be accomplished.

Since the TMRG gives results for observable quantities in the thermodynamic limit, we are able to exploit it to study the bulk properties of the system without worrying the finite size effect. The complicated size dependence of the ground-state energy and correlation function found in the region \( \alpha < -1/4 \) (Ref.\[32,33\]) may be avoided.

Figures\[3,4\] show the TMRG results on the temperature dependence of the susceptibility \( \chi \), specific heat \( C \), and heat coefficient \( C/T \) at various \( \alpha \) for the F-AF case. At \( \alpha = 0 \), the model reduces to the case of ferromagnetic spin chain. The curves of \( \alpha = 0 \) describe the properties of \( \chi \) and \( C \) for a ferromagnetic spin chain, which behave in the low temperature limit:

\[
\chi \sim T^{-2}, \quad C \sim \sqrt{T}.
\]

At \( \alpha > -1/4 \), the temperature dependence of \( \chi \) always diverges, indicating that the system lies in a ferromagnetic state. At the critical point \( \alpha = -1/4 \), a phase transition from the ferromagnetic state to the singlet state takes place. In the region \( \alpha < -1/4 \), remarkable suppressions of the susceptibility can be observed, and with the decrease of \( \alpha \), the peaks of \( \chi \) move to higher temperatures with its heights decreased rapidly.

For the temperature dependence of the specific heat, the most remarkable feature is the development of a double-peak structure. With the decrease of \( \alpha \), at intermediate temperatures, a broad maximum is maintained and its height lowered. A relatively sharp peak at low temperatures, induced by the NNN AF interaction \( J_2 \), appears and develops, and its position moves to higher temperatures. We expect that this peak would approach the maximum of \( C(T) \) for a pure AF Heisenberg chain\[34\]. The drastic change on the shape of the curves between \( \alpha = -0.125 \) and \( \alpha = -0.28 \) implies a phase transition.
III. THE AF-F CASE

In the AF-F case with \( J_1 > 0 \) and \( J_2 < 0 \), the behavior of the zigzag spin chain is believed to be antiferromagnetic with no gap for any permissible values of \( J_1 \) and \( J_2 \). The arguments are based on a simple spin-wave analysis. The spectrum of spin-wave excitations is given by

\[
\epsilon_k = 2S \sqrt{\lambda_k^2 - \gamma_k^2},
\]

where \( S = 1/2 \), \( \alpha = J_2/J_1 < 0 \), \( \lambda_k = -2\alpha \sin^2 k + 1 \), \( \gamma_k = \cos k \). \( \epsilon_k \) is linear as \( k \to 0 \):

\[
\epsilon_k \sim \nu k
\]

with \( \nu = 2S\sqrt{4\alpha + 1} \). Although the spin-wave theory is not quite correct for antiferromagnetic spin chains, it is still insightful in shedding light on the qualitative picture of the excitations around the characteristic momentum \( k = 0, \pi \). We see that for the AF-F case, there also exist gapless excitations at \( k = 0, \pi \).

According to Lieb’s discussion, Hamiltonian with \( J_1 > 0, J_2 < 0 \) describes spin systems at a bipartite lattice. The absolute ground state of the AF-F spin chain should lie in the \( S = 0 \) sector. On the other hand, the relative ground state in each subspace \( V(M) \) of the total magnetic quantum number \( M \) is unique. Thus we deduce that the absolute ground state in the AF-F case is nondegenerate. The Lieb-Schultz-Mattis theorem asserts that for a half-integer spin chain with reasonably local Hamiltonian respecting translational and rotational symmetries, either the parity is spontaneously broken in the ground state or else there are gapless excitations of odd parity, under the condition that the ground state is rotational invariant. Combining these two statements, we expect the system should be gapless.

It is not trivial to make the above statement completely rigorous, considering the relative ground state in the subspaces \( V(M) \) may not be unique in the thermodynamic limit \( N \to \infty \). In order to confirm the conclusion, we calculate the temperature dependence of the susceptibility and specific heat for various \( \alpha \) by using the TMRG method.

Figures 5 and 6 show the TMRG results on the temperature dependence of the susceptibility \( \chi \), specific heat \( C \), and heat coefficient \( C/T \) at various \( \alpha \) for the AF-F case. With the decrease of \( \alpha \), the behaviors of \( \chi \) and \( C \) do not change qualitatively. \( \chi \to \text{const} \) (nonzero) and \( C \sim T \) as \( T \to 0 \). The Bonner-Fisher peak in \( \chi(T) \) (Ref. 30) moves to higher \( T \) with the decrease of \( \alpha \). Phase transition is not observed. The slopes of \( C \), or the intersections of \( C/T \) at zero temperature (shown in Fig. 5), decrease with the decrease of \( \alpha \). This indicates that the density of low-excitation states becomes smaller and the spin fluctuations are suppressed by the NNN FM interaction.
 Recently, the magnetic susceptibility and magnetization of the edge-sharing copper oxide Rb$_2$Cu$_2$Mo$_2$O$_7$ (Ref. 14) have been measured. The most interesting finding is that no magnetic phase transition was observed down to 2 K. Therefore, the compound is suitable for studying the properties of the ground state of the J$_{1}$-J$_{2}$ model. It was proposed that at first approximation, it could be described by a F-FA chain, the unusually long correlation length, which exist even at high-frustrated region, can lead to prominent finite-size effect for the usual cluster simulations. Therefore, as a primary step, reliable numerical results on thermodynamic quantities free of finite-size effect on the pure J$_{1}$-J$_{2}$ model may be essential in making a quantitative comparison between experimental and theoretical studies. In this section, we use the TMRG method to simulate the experimental results based on model (2).

The strategy is as follows: For a fixed $\alpha$, we first use the TMRG to calculate the temperature dependence of the susceptibility in the units of J$_1$. Then, according to the position of the peak in the numerical result and the actual value obtained in the experiment, we can determined the value J$_1$ uniquely, on the condition that the calculated peak's position coincides with the experiment. The g value is taken to be 2.03 as in Ref. 14. The following is what we observed.

For the AF-AF case (Fig. 7), the height of the peak decreases with the increase of $\alpha$. The possibility for this type of interaction can be excluded.

For the F-AF case (Fig. 8), we see that with $\alpha$ approaching the critical point $-1/4$, to keep the peak position unchanged, the magnitude of J$_1$ acquires a value of thousands of Kelvin. Furthermore, the deviation from the experiment data at high temperatures becomes more conspicuous. While taking the parameters J$_1$ = −138 K and $\alpha = −0.37$ as in Ref [14] the temperature for the peak is less than $T_{\text{max}} = 14.3$ K found in the experiment. Besides, for $\alpha < -0.5$, a broad peak located at $T \sim J_2$ in the temperature dependence of susceptibility emerges, which is a characteristic feature of one-dimensional Heisenberg
antiferromagnetic chain. This suggests that the antiferromagnetic component becomes significant for larger $\alpha$. However, we failed to obtain a satisfactorily stable result below the peak temperature when $\alpha < -1$.

For the AF-F case (Fig. 9), at $J_1 = 12.31$ K and $\alpha = -3.5$, the numerical result seems to fit the experiment data rather well, except for a little deviation in the left side of the peak at low temperatures. But there are several questionable points remained. According to the analysis of Mizuno et al., the NNN interaction through Cu-O-O-Cu should be antiferromagnetic, i.e. $J_2 > 0$. However, the interactions of AF-F type may also be a candidate. In Ref. [40], Matsuda and coworkers proposed a model to explain the anomalous magnetic excitations in the edge-sharing CuO$_2$ chains of La$_5$Ca$_9$Cu$_{24}$O$_{41}$. The intra- and interchain interactions are of AF-F type. (See also Fig. 10)

We also calculated the field dependence of the magnetization for the two cases: $J_1 = -138$ K, $\alpha = -0.37$; and $J_1 = 12.3$ K, $\alpha = -3.5$, to verify further if the only $J_1$-$J_2$ model is sufficient to describe the behaviors of the compound. We find the anomalous slow saturation of the magnetization cannot be reproduced either, and the result for the AF-F case seems worse.

From the TMRG numerical results on the $J_1$-$J_2$ model and the comparison made with the experiment, we feel that only the NN and NNN interactions can not describe the properties of Rb$_2$Cu$_2$Mo$_3$O$_{12}$ satisfactorily, although a model based on the F-AF interactions seems to be an appropriate starting point.

The above facts reveal the complexity of the interac-
tions underlying the edge-sharing copper oxides. On one hand, due to the strong electron correlations in these so-called Mott insulators, very limited information on the electronic structure can be obtained reliably. This makes it difficult to calculate accurately the superexchange interaction. For instance, based on a three-band Hubbard Hamiltonian and cluster calculation, the NN and NNN interaction $J_1$, $J_2$ for Li$_2$CuO$_2$ were obtained as $J_1 = -100$ K, $J_2 = 62$ K ($\alpha = -0.62$). The results of the quantum chemical calculation were given as $J_1 = -142$ K, $J_2 = 22$ K ($\alpha = -0.15$). On the other hand, because the NN coupling in edge-sharing copper oxides is extremely small, other interactions, such as quantum frustrations, weak interchain correlations, and anisotropies can all have a chance to play an unnegligible role in determining the phase and behavior of the system. They are also closely related to the lattice structures and chemical compositions. These combined effects make any reasonable analysis intricate. In order to quantitatively recover the experimental data and various magnetic orders at low temperatures, more parameters are needed. This brings some “flexibility” to the theory. For instance, the broadening of the magnetic excitations found in Ca$_2$Y$_2$Cu$_3$O$_{10}$ requires the introduction of the antiferromagnetic interchain interactions and anisotropies for superexchange interactions. In order to understand the helicoidal magnetic order in NaCu$_2$O$_2$, four parameters including frustrated longer-range exchange interactions are needed. However, we wish to emphasize here that accurate knowledge on the behavior of model especially at low-temperature region, is indispensable in understanding the properties of these materials.

In conclusion, we go back to the compound Rb$_2$Cu$_2$Mo$_3$O$_{12}$ and take a closer look at the lattice structure of Rb$_2$Cu$_2$Mo$_3$O$_{12}$. Since the nearest neighbor Cu-Cu bond has two slightly alternating configurations by turns, the chain is distorted into a zigzag shape (See Fig. 1 in Ref. 1). Additional antisymmetric exchange interactions, such as Dzyaloshinskii-Moriya (DM) or anisotropic interactions together with an alternating $g$ tensor, should become more important than in a straight-line chain. As discussed by Dzyaloshinskii and Moriya for the magnetic crystals with lower symmetries, the effect of this antisymmetric exchange should become more manifest than the exchange anisotropy. The magnitude of this interaction is estimated as

$$D \sim (\Delta g/g) J,$$  \hspace{1cm} (13)

and the usual term of DM interaction can be expressed as

$$H_{DM} = \sum_j D_j \cdot (S_j \times S_{j+1}).$$  \hspace{1cm} (14)

Because of the alternating $g$ tensor and the DM interaction, an external uniform magnetic field can induce an effective staggered field. If $D_j$ takes the form $(-1)^j D$, as it should be in the present case, the transverse component of the staggered field, which is perpendicular to the uniformly applied field, becomes dominant. For the $S = 1/2$ Heisenberg antiferromagnetic chain, a gap is generated by a staggered field. The magnetization becomes gradually saturated for large fields. The similar mechanism may also work for the F-AF case. A combination with the frustration effect caused by the NNN $J_2$ make the situation more interesting.

Recently, it was suggested that for $\alpha < -0.38$, there exists an incommensurate-commensurate transition at some critical field in the magnetization process. The sharp increase up to $M \approx 0.4$ at $B \approx 14$ T and the following gradual saturation found in the experiment is argued to be connected to this phase transition.

V. SUMMARY

In this paper, we explored the properties of the zigzag spin chain with different combinations of ferro- and antiferromagnetic interactions between the NN and NNN sites. The existence of the gap in the F-AF case and the nonexistence of the gap in the AF-F case were discussed. Thermodynamic properties of the zigzag spin chain in various phases were studied by using the TMRG method. The obtained results were used to compare with the experimental data from Rb$_2$Cu$_2$Mo$_3$O$_{12}$. We pointed out that for such edge-sharing copper oxide chains, besides the NN and NNN couplings, more ingredients, such as the interchain or DM exchange interactions, may be important in these materials.

Acknowledgments

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