Competition between the Néel and the Effective Singlet States in Spin-1/2 Alternating Heisenberg-Ising Antiferromagnet in One Dimension

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The ground-state properties of the one-dimensional spin-1/2 alternating Heisenberg-Ising antiferromagnet are studied both analytically and numerically. The phase diagram is qualitatively predicted by the use of the phase Hamiltonian: The ground state is either the Néel or the effective singlet state according to the values of the alternation and anisotropy parameters. The phase transition between these states is of the first order. The ground state energy, its derivative with respect to the alternation parameter, excitation gap, and the various long-range order parameters are exactly calculated by the numerical method for finite systems (N≤20). The numerical calculations support the analytically predicted phase diagram. The correlation length is also numerically calculated at finite temperatures by the use of the quantum transfer-matrix method.

§1. Introduction

There has been increasing attention focused on the low temperature properties of one-dimensional spin-1/2 alternating antiferromagnetic Heisenberg model, especially in relation to the spin-Peierls transition. The Hamiltonian of this system is

\[
H = J \sum_i \left[ 1 + (-1)^i \delta \right] (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y)
+ \Delta S_i^z S_{i+1}^z, \quad J > 0,
\]

(1.1)

where \(0 \leq \delta \leq 1\) represents the degree of the bond alternation and \(\Delta\) the anisotropy. This Hamiltonian represents the spin degree of freedom of the organic compounds which fall into the spin-Peierls state at low temperatures.

This model can be transformed into the interacting spinless Fermion model through the Jordan-Wigner transformation. The earlier investigators employed the Hartree-Fock approximation for the resulting spinless Fermion model. An important progress was given by Cross and Fisher\(^4\) who used the bosonization method to treat the quantum fluctuations more completely. They showed \(\Delta E \propto \delta^{4/3}\) and \(\varepsilon_0 \propto \delta^{2/3}\) for the \(\Delta = 1\) case, where \(\Delta E\) is the decrement of the ground state energy due to the alternation and \(\varepsilon_0\) the energy gap between the ground state and the first excited state. This result should be compared with that of the Hartree-Fock approximation, \(\Delta E \propto \delta^2 |\log \delta|^{2/3}\) and \(\varepsilon_0 \propto \delta |\log \delta|^{1/3}\). Numerical studies\(^5-8\) supported the results of Cross and Fisher. The exponent 4/3 (with logarithmic correction) of \(\Delta E\) was also obtained by the renormalization group method.\(^9\) Nakano and Fukuyama\(^10,11\) refined the method of Cross and Fisher to transform the Hamiltonian (1.1) into the phase Hamiltonian and calculated \(\Delta E\) and \(\varepsilon_0\), taking the XY-like anisotropy \((0 < \Delta \leq 1)\) into consideration. Using this phase Hamiltonian, one of the present authors\(^8,12\) (K.O.) calculated the spin correlation \(\langle S_i^z S_j^z \rangle\) to find its exponential decay. These results in the \(0 < \Delta \leq 1\) case were also confirmed by the numerical calculations.\(^8,13\) Very recently, the behavior of the correlation length of \(\langle S_i^z S_j^z \rangle\) as a function of \(T\) was discussed.\(^14\) When \(\Delta = 0\), the quantities \(\Delta E\), \(\varepsilon_0\) and \(\langle S_i^z S_j^z \rangle\) (even at \(T \neq 0\)) were exactly calculated.\(^15\)

Owing to the above mentioned works, the properties of this model in the \(0 \leq \Delta \leq 1\) case have been fairly clarified as far as the ground state is concerned. The ground state of this
model in the $\delta=0$ and $0 \leq \Delta \leq 1$ case is complicated and may be called the spin fluid state. It contains the singlet-like components, triplet-like components, Néel-like components and so on. This fact is reflected in the gaplessness of the excitation spectrum, the power-law decay of $\langle S_i^+ S_j^- \rangle$ and the finite value of the susceptibility at $T=0$. Once the bond alternation is introduced ($\delta>0$), however small it may be, the singlet-like components are semi-exclusively selected for the ground state (i.e., spins $S_{2i}$ and $S_{2i+1}$ form an effective singlet pair), which brings about the excitation gap, the exponential decay of $\langle S_i^+ S_j^- \rangle$ and the vanishing of the susceptibility. As $\delta \to 0$, the quantities $\Delta E$, $e_g$ and $1/\xi$ ($\xi$ is the correlation length of $\langle S_i^+ S_j^- \rangle$) continuously tend to 0. Therefore the transition from the $\delta>0$ state to the $\delta=0$ state is of the second order.

On the other hand, the $\Delta>1$ case is quite different from the $0 \leq \Delta \leq 1$ case. When $\delta=0$, the system is in the Néel state, which leads to the existence of the energy gap between the $S^z=0$ and $S^z=1$ branches of the excitation spectrum, and the finite value of $\langle S_i^+ S_j^- \rangle$ as $|i-j| \to \infty$. Consequently, when $\delta>0$, the subtle competition between the Néel state and the effective singlet (ES) state will be observed. Inagaki and Fukuyama investigated this problem for the case of small Ising anisotropy ($0<\Delta-1<1$) using the method of Nakano and Fukuyama and showed that the ground state is either the Néel state or the ES state, depending on $\Delta$ and $\delta$. Very recently, Kuboki and Fukuyama investigated this problem by applying the renormalization group method to the phase Hamiltonian, including the next nearest interaction between spins. However, no numerical work for the $\Delta>1$ case has been reported in the literature. Therefore, it is interesting to investigate this problem by the numerical methods. We note that there is a slight difference between our model and that of Inagaki and Fukuyama. The quantity $\delta$ is a given parameter in the present work, while in their paper it was determined so as to minimize the sum of the spin energy and the lattice distortion energy (proportional to $\delta^2$).

In this paper, we show the existence of two states (Néel and ES) for the $\Delta>1$ case and clarify the various properties of these states. In §2, the phase Hamiltonian approach is described to draw a sketch of the phase diagram on the $\Delta-\delta$ plane. The results of the exact numerical calculations for finite systems using a computer program developed by Nishimori and Taguchi are given in §3. The last section §4 is devoted to conclusion.

§2. Phase Hamiltonian Approach

Nakano and Fukuyama[10,11] transformed the spin Hamiltonian (1.1) into the following phase Hamiltonian:

$$H = \int dx [A(\nabla \theta)^2 + CP^2 - B \cos \theta + D \cos 2\theta],$$

(2.1)

$$[\theta(x), P(x')] = i\delta(x-x').$$

(2.2)

The constants $A$, $C$, $B$ and $D$ are

$$A = \frac{Jd}{8\pi} \left( 1 + \frac{3\Delta}{\pi} \right), \quad C = 2\pi Jd \left( 1 - \frac{\Delta}{\pi} \right),$$

(2.3)

$$B = \frac{J \delta}{d}, \quad D = \frac{\pi^2 \Delta J}{8d},$$

(2.4)

where $d$ is the spin spacing. The phase variable $\theta(x)$ is related to $S^z(x)$ as

$$S^z(x) = \frac{1}{d} \sin \left( \frac{\pi x}{d} + \theta(x) \right) + \frac{1}{2\pi} \nabla \theta(x),$$

(2.5)

where the first term varies in space much faster than the second term.

When $\delta=0$ (i.e., $B=0$), eq. (2.1) is the sine-Gordon Hamiltonian and $D \cos 2\theta$ term is known to be irrelevant in the sense of renormalization group as far as $\eta>1$, where $\eta$ is defined by

$$\eta = (2\pi)^{-1} \sqrt{C/A}.$$  

(2.6)

In this case ($B=0$ and effectively $D=0$), the behavior of the spin correlation function $\langle S_i^+ S_j^- \rangle$ and the excitation spectrum are exactly calculated as

$$\langle S_i^+ S_j^- \rangle \sim |i-j|^{-\eta},$$

(2.7)

$$\omega(q) = v_s q,$$

(2.8)

respectively, where $v_s$ is the phason velocity (i.e., the spin wave velocity) given by

$$v_s = 2\sqrt{AC}.$$  

(2.9)

However, the values of $\eta$ and $v_s$ are different
from the exact values$^{22,23}$ obtained directly from the spin Hamiltonian (1.1), which seems to be due to the approximations in transforming the spin Hamiltonian into the phase Hamiltonian. The important approximations are the linearization of the cosine band and the neglect of the wave vector dependence of the strength of the mutual interaction between Fermions. This effect becomes serious as we move away from $\Delta = 0$ to larger $\Delta$. In fact, the expansions of eqs. (2.6) and (2.9) with respect to $\Delta$ agrees with those of the exact values to the order of $\Delta^2$. For the $\Delta = 1$ case, Cross and Fisher$^{20}$ proposed a neat way to remove this difficulty: Their proposal was, in the language of the phase Hamiltonian, to adjust the coefficients $A$ and $C$ so that eqs. (2.6) and (2.9) yield the exact values. This procedure was applied by Nakano and Fukuyama$^{11}$ for the $0 < \Delta \leq 1$ case. By the use of the phase Hamiltonian with adjusted $A$ and $C$, the exact value of the susceptibility$^{24,25}$ at $T = 0$ is reproduced, which suggests the justifiability of the above procedure. We note that $\eta = 2$ and $1$ for $\Delta = 0$ and $1$, respectively, after the adjustment of $A$ and $C$. Consequently, the term $D \cos 2\theta$ is irrelevant for the $0 < \Delta < 1$ (i.e., $XY$-like) case.

On the other hand, the $\Delta > 1$ (i.e., Ising-like) case is more complicated, because the term $D \cos 2\theta$ is relevant and it is necessary to adjust three coefficients ($A$, $C$ and $D$). However, the adjusting procedure is not clear, since $\langle S_i^z S_j^z \rangle$ has long range order and the exponent $\eta$ in eq. (2.7) is no longer available. For $0 < \Delta - 1 < 1$, Inagaki and Fukuyama$^{16}$ tried to adjust the coefficients so that the low-lying excitation derived from the phase Hamiltonian agrees with the exact one.$^{22,23}$ But some numerical parameters were left undetermined.

When $\Delta > 1$, the validity of the bosonization method is not readily guaranteed, because the interaction energy is larger than the kinetic energy in the spinless Fermion picture. Nevertheless, here we assume the validity of the phase Hamiltonian if the coefficients are suitably chosen, since the phase Hamiltonian representation yields the intuitive description for the states which are expected to exist. In the following, we consider the behavior of each coefficient as $\Delta$ and $\delta$ vary, instead of the adjustment.

The parameter $\eta$ defined by eq. (2.6) represents the degree of quantum nature, and $\eta = 1$ for $\Delta = 1$ (isotropic Heisenberg case) and $\eta = 2$ for $\Delta = 0$ ($XY$ case). Since the present system becomes classical in the Ising limit ($\Delta = \infty$), the quantum parameter should be $\eta = 0$ when $\Delta = \infty$. Therefore we may conclude that $\eta$ monotonically decreases from $1$ to $0$ as $\Delta$ varies from $0$ to $\infty$. From eq. (2.4) we may consider that coefficients $B$ and $D$ will monotonically increase with the increase of $\delta$ and $\Delta$, respectively. The values of $B(\delta = 1)$ and $D(\Delta = \infty)$ are unknown in the present stage and will be predicted later. If we substitute $x_i = i\delta$ (the location of the $i$-th spin) for $x$ in eq. (2.5), we see that the two possible states are distinguished by the value of $\theta_i$, where $\theta$ is the equilibrium value of $\theta$. Namely, the system is in the ES state or the Néel state according as $\theta_i = 0$ or $\theta_i \neq 0$. This fact is more clearly seen by calculating the spin correlation function$^{12}$ $\langle S_i^z S_j^z \rangle$. As $|i-j| \to \infty$, it decays exponentially for $\theta_i = 0$, whereas it has a finite value for $\theta_i \neq 0$. Figure 1 shows the possible shapes of the phase locking potential $V(\theta)$ defined by

$$V(\theta) = -B \cos \theta + D \cos 2\theta. \quad (2.10)$$

Of course, the minimum of $V(\theta)$ does not always correspond to $\theta_i$, because the quantum fluctuations effectively reduce the phase locking potential. In fact, when $\Delta < 1$ and $\delta = 0$, as we already stated, the phase locking potential is completely destroyed by the quantum fluctuations and becomes irrelevant. In the self-consistent harmonic approximation$^{11}$ (SCHA), the coefficients $B$ and $D$ are renormalized by the quantum fluctuations as

$$B \to \bar{B} = B \exp (-\langle \tilde{\theta}^2 \rangle / 2), \quad (2.11)$$
$$D \to \bar{D} = D \exp (-2\langle \tilde{\theta}^2 \rangle),$$

respectively, where

$$\bar{\theta} = \theta - \theta_i. \quad (2.12)$$

The value of $\theta_i$ is zero or non-zero according as the ratio

$$\bar{B} / 4\bar{D} = (B / 4D) \exp (3\langle \tilde{\theta}^2 \rangle / 2), \quad (2.13)$$

is larger or smaller than unity. Therefore, the condition $B > 4D$ is sufficient for $\theta_i = 0$. This
suggested that the system is in the ES state as far as $\delta$ is larger enough. In fact, when $\delta=1$, the singlet state is clearly realized because the Hamiltonian (1.1) is reduced to the two-spin Hamiltonian. In the following, we fix $\delta$ ($0<\delta<1$) and move $\Delta$ from 1 to $\infty$. As already shown by the previous investigators, the system is in the ES state when $\Delta=1$. The increase of $\Delta$ is advantageous to the Néel state ($\theta_1 \neq 0$) in a twofold manner. First, the minimum of $V(\theta)$ is possibly moved from 0 to $\theta_1$ or the minimum of $V(\theta)$ at $\theta_1$ becomes deeper. Second, the factor exp($3\langle \hat{\theta}^2 \rangle /2$) in the ratio (2.13) becomes smaller as expected from the behavior of the quantum parameter $\eta$ as a function of $\Delta$. In the Ising case ($\Delta = \infty$) the value of $\theta_1$ is given by the minimum of the bare phase locking potential $V(\theta)$ and it should be $\pi/2$ as can be seen from eq. (2.5).

This suggests $D(\Delta = \infty) = \infty$. Therefore the Néel state will be realized for sufficiently large $\Delta$. Even when $\Delta \gg 1$, as already mentioned, the ES state appears for $\delta=1$, from which $B(\delta=1) = \infty$ is suggested.

From the above argument, the phase diagram in the $\Delta-\delta$ plane is predicted as Fig. 2. In the SCHA, the phase transition between the ES state and the Néel state is of the first order. At the end of this section, we summarize the limiting behavior of the coefficients of the phase Hamiltonian.

$$\eta(\Delta = 1) = 1, \quad \eta(\Delta = \infty) = 0,$$

$$B \propto \delta, \quad (\delta < 1), \quad B(\delta = 1) = \infty,$$

$$D \propto \text{const.} + O(\sqrt{\Delta - 1}), \quad (0 < \Delta - 1 < 1)$$

$$D(\Delta = \infty) = \infty.$$

Equation (2.18) is taken from the paper of Inagaki and Fukuyama. Of course, eqs. (2.15), (2.17) and (2.19) were obtained under the assumption that the phase Hamiltonian description is valid for the whole range of $0 \leq \delta \leq 1$ and $1 \leq \Delta < \infty$ if the coefficients are suitably chosen.

§3. Numerical Analysis

In order to verify the predictions in §2, we numerically investigated the properties of the spin Hamiltonian (1.1). We first diagonalized the Hamiltonian of finite-size systems (number of sites $N=10, 12, \ldots, 20$) by the method described in ref. 18, and evaluated the
ground-state energy per site $E_0$, the excitation gap, the derivative of the energy $\partial E_0 / \partial \delta$, and various long-range order parameters. We also employed the quantum transfer-matrix method\textsuperscript{20} to estimate the correlation length $\xi(T)$ at finite temperatures. As a typical example, our data in the case of $\Delta=5$ are given in Figs. 3(a)–3(f). Figure 3(a) shows the energy derivative $\partial E_0 / \partial \delta$ as a function of the anisotropy parameter $\Delta$. This quantity $\partial E_0 / \partial \delta$ is the ground-state expectation value of the operator

$$J \sum_i ( -1 ) ( S_i^z S_{i+1}^z + S_i^x S_{i+1}^y + \Delta S_i^z S_{i+1}^z ) / N.$$  

(3.1)

A comparison of eqs. (3.1) and (1.1) readily reveals that $\partial E_0 / \partial \delta$ represents the energy difference between stronger bonds with exchange interaction $J(1+\delta)$ and weaker ones with $J(1-\delta)$. Thus the ES state would have a larger absolute values of the derivative $\partial E_0 / \partial \delta$ than the Néel state does. A phase transition of the first order between one state to the other results in a jump in this quantity. In fact, the system size dependence clearly changes at

$$\delta_1 = 0.43 \pm 0.01.$$  

Extrapolation to the infinite system ($N \to \infty$) by the alternating $\varepsilon$-algorithm\textsuperscript{27} yields the line marked $\infty$ which has a finite jump at the above mentioned $\delta_1$. Since the free energy is identical to the internal energy at $T=0$, this jump in the first derivative of $E_0$ marks a transition of the first order, thus confirming a phase Hamiltonian prediction in the preceding section. The energy gap between the ground state (which is in the subspace $S^z=0$) and the first excited state in the same subspace is depicted in Fig. 3(b). Similar extrapolation as in the energy derivative gives the bold curve, implying a vanishing gap for $\delta < \delta_1$ and a finite gap when $\delta > \delta_1$. The analytical calculation of excitation spectrum\textsuperscript{23} of the uniform system ($\delta=0$) tells us the existence of gapless spin-wave mode in the subspace $S^z=0$ in the Ising-like region ($\Delta > 1$). The gaplessness in $\delta < \delta_1$ in Fig. 3(b) is consistent with this result at $\delta=0$ and suggests that the system is in the Néel state for $\delta < \delta_1$. We may therefore conclude that the excitation spectrum is not seriously modified in the presence of bond alternation as far as $\delta < \delta_1(\Delta)$. Of course, excitation to a state with $S^z=1$ requires finite energy irrespective of the value of $\delta$ ($0$ or finite). In the range $\delta > \delta_1$,
energy gap opens up even within the subspace $S^z=0$. This corresponds to effective formation 
of singlet pairs (the ES state) as discussed in §2.

The first order transition from the Néel state $(\delta<\delta_c)$ to the ES state $(\delta>\delta_c)$ should further be confirmed by directly calculating the staggered long-range order

$$\sum_{i<j} (-1)^{i+j} \langle S_i^z S_j^z \rangle \sim N^x. \quad (3.2)$$

Here the expectation value is taken with respect to the ground state wave function. If the antiferromagnetic long-range order exists, the double sum on the left hand side of eq. (3.2) will yield contribution of order $N^2$ and the exponent $x$ is 2. When only short-range order exists, $x$ is equal to 1. Power-law decay of correlation function would result in an intermediate value of $x$ between 1 and 2. Our result is depicted in Fig. 3(c). The dashed line comes from the fit of data at $N=10, 12, \cdots, 20$ to eq. (3.2) while the analysis of the largest two $N$'s (18 and 20) yields the solid curve. Figure 3(c) suggests an asymptotic approach to a step function $x=2$ for $\delta<\delta_c$ and $x=1$ for $\delta>\delta_c$ with $\delta_c$ between 0.4 and 0.5. This value is consistent with $\delta_c(\Delta=5)=0.43\pm0.01$ obtained from the energy derivative given above. One may not be able to convincingly exclude the possibility of an intermediate state $(1<x<2)$ in the range $\delta_c<\delta<1$ only from Fig. 3(c). For further confirmation of the absence of such a state at $\delta_c(\Delta)<\delta$, we calculated the finite-temperature correlation length $\xi$ defined by

$$\langle S_i^z S_j^z \rangle \sim \exp\left(-|i-j|/\xi\right). \quad (3.3)$$

If the correlation function decays algebraically (i.e. $1<x<2$) in the ground state, the correlation length $\xi(T)$ should diverge as $T\to0$. The ratio of the two largest eigenvalues of the quantum transfer-matrix\textsuperscript{26} readily yields $\xi$ as a function both of the temperature $T$ and of the degree of approximation $m$ in the quantum-classical mapping used in the quantum transfer-matrix method. The original system (1.1) is recovered in the limit $m\to\infty$. The calculated $\xi$ is shown in Fig. 3(d). The parameters $\Delta=2$ and $\delta=0.6$ are chosen so that the system is definitely away from the Néel state (i.e., $0<\delta_c<0.6$ when $\Delta=2$, see Fig. 4). We find no evidence for a divergent correlation length as the temperature approaches 0. Thus the phase in the region $\delta_c<\delta<1$ is in the ES state with exponential decay of the correlation function.

The absence of long-range order in the $XY$-plane is verified by calculating the order parameters

$$M_2^z / N^2 = \left(\langle (\sum S_j^y)^2 + (\sum S_j^z)^2 \rangle / N \right) / N^2. \quad (3.4)$$

Fig. 3(c). Exponent of the long-range order defined in eq. (3.2). The dashed curve is obtained by a least-squares fit of data for $N=10, 12, \cdots, 20$ to eq. (3.2). If only the largest two $N$'s (18 and 20) are used to evaluate $x$, one obtains the solid curve.

Fig. 3(d). Correlation length at finite temperatures when $\Delta=2$ and $\delta=0.6$. Here $m$ denotes the degree of approximation used in the quantum-classical mapping in the evaluation of the quantum transfer-matrix method.\textsuperscript{26} Fully quantum-mechanical system is recovered in the limit $m\to\infty$. At each $T$ extrapolation to $m\to\infty$ is reliably performed if the data do not depend on $m$ very much, which corresponds to the temperature range $T \geq 0.15$ in this figure.
Fig. 3(e). Long-range order in the $XY$ plane depends almost linearly on $1/N$ at a fixed $T$. Thus in the limit $N \to \infty$ we expect no long range order of this type.

Fig. 4. Phase diagram obtained by the numerical method. The boundary curve is drawn to interpolate data points. One finds qualitative agreement with the prediction of the method of phase Hamiltonian given in Fig. 2.

Fig. 3(f). Staggered long-range order in the $XY$ plane has behavior similar to the uniform order in Fig. 3(e).

$$\bar{M}_1^2 / N^2 \equiv \left\{ \langle (\sum (-)^j S_j^x) \rangle^2 + \langle (\sum (-)^j S_j^y) \rangle^2 - \frac{N}{2} \right\} / N^2.$$  \hspace{1cm} (3.5)

The constant $N/2$ in these equations represents autocorrelation which is irrelevant to long-range order and should be subtracted off. If a uniform (ferromagnetic) $XY$-ordering exists, the expectation value (3.4) will approach a finite limit as $N \to \infty$, which is denied in Fig. 3(e). A staggered $XY$-ordering corresponds to a finite limit of eq. (3.5), also disproved in Fig. 3(f).

These analyses are carried out at various points in the $\Delta$-$\delta$ plane. The result is summarized in Fig. 4 and Table I. The large error bars around $\Delta \approx 2$ are a consequence of insufficient accuracy in the numerical evaluation of the energy derivative $\partial E_0 / \partial \delta$.

§4. Conclusion

We have presented both analytical and numerical results on the alternating spin system (1.1) in the region $\Delta > 1$. The method of phase Hamiltonian predicts two states, the Néel and ES states, as depicted in Fig. 2. These two states are separated by a first order transition. Numerical investigation confirmed those predictions and we could draw the precise phase boundary $\delta_c(\Delta)$ as shown in Fig.

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<th>$\Delta$</th>
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<tr>
<td>2.0</td>
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<tr>
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<tr>
<td>8.0</td>
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4. In summary, the Néel state of the uniform system ($\delta = 0$) persists in the presence of bond alternation $\delta$ below some critical value $\delta_c(\Delta)$. Beyond $\delta_c$, the ES state takes over. This ES state is essentially the same one as found in the XY-like region $\Delta \leq 1$, $0 < \delta < 1$, discussed previously.\textsuperscript{10-14}

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References