Random Magnetism in $S = 1/2$ Heisenberg Chains with Bond Alternation and Randomness on the Strong Bonds

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The $S = 1/2$ Heisenberg chains with bond alternation and randomness on the strong bonds are studied by the density matrix renormalization group method. It is assumed that the odd-th bond is antiferromagnetic with strength J and even-th bond can take the values $J_{\rm A}$ and $J_{\rm F}$ ($J_{\rm A} > J > 0 > J_{\rm F}$) randomly. The ground state of this model interpolates between the Haldane and dimer phases via a randomness dominated intermediate phase. Based on the scaling of the low energy spectrum and mean field treatment of the interchain coupling, it is found that the magnetic long range order is induced by randomness in the intermediate regime. In the magnetization curves, there appears a plateau at the fractional value of the saturated magnetization. The fine structures of the magnetization curves and low energy spectrum are understood based on the cluster picture. The relation with the recent experiment for $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$ is discussed.

KEYWORDS: random quantum spin chain, bond alternation, DMRG, disorder induced order, magnetization plateau

1. Introduction

In the recent studies of quantum many body problem, the one-dimensional random quantum spin systems have been attracting a renewed interest from theoretical and experimental viewpoints.[1–15)]

Among them, the phenomenon of the disorder induced order have been widely investigated. Experimentally, Uchinokura and coworkers[3,4)] have found the antiferromagnetic ordered phase in Zn, Mg, Si-doped spin-Peierls compound $CuGeO_4$ as one of the earliest examples of this type of phenomenon. The theoretical explanation is given by Fukuyama and coworkers[5)] using the bosonization approach.

Similar phenomena are observed in Zn-doped $SrCu_2O_3$[6)] which is the quasi-1-dimensional $S = 1/2$ ladder system and Mg-doped $PbNi_2V_2O_8$[4,7)] which is the quasi-1-dimensional $S = 1$ Haldane gap system. Correspondingly, the effect of the bond and site randomness on the spin gapped quasi-1-dimensional $S = 1/2$ and $S = 1$ antiferromagnets are studied by the quantum Monte Carlo method by Yasuda and coworkers.[8)] They have also found the randomness induced long range order for appropriate range of randomness.

Recently, Manaka and coworkers[1,2)] studied the magnetic and thermal properties of the quasi-one-dimensional compound $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$ (IPACu(Cl_xBr_{1-x})). For $x = 0$, this material is the $S = 1/2$ antiferromagnetic-antiferromagnetic alternating Heisenberg chain (AF-AF chain) whose ground state is the dimer phase[16)] while for $x = 1$ it is the $S = 1/2$ ferromagnetic-dominant ferromagnetic-antiferromagnetic alternating Heisenberg chain (F-AF chain) with Haldane ground state.[17,18)] In the intermediate concentration regime, however, both the Haldane and dimer phases are destroyed by randomness. Remarkably, Manaka and coworkers[1)] experimentally found the magnetically ordered phase for $0.44 < x < 0.87$. It should be also remarked that the energy gap estimated from the temperature dependence of the specific heat and susceptibilty remains finite even in the close neighbourhood of the critical concentration.

Modivated by this experiment, we investigate the random $S = 1/2$ Heisenberg chain whose Hamiltonian is given by,

$$H = \sum_{i=1}^{N} J\boldsymbol{S}_{2i-1}\boldsymbol{S}_{2i} + \sum_{i=1}^{N} J_i\boldsymbol{S}_{2i}\boldsymbol{S}_{2i+1}, \qquad (1)$$

where $J > 0$ and $J_i = J_{\rm F}(< 0)$ with probability $1-p$ and $J_i = J_{\rm A}(> 0)$ with probability p. The ground state of this model interpolates between the Haldane phase ($p = 0$) and dimer phase ($p = 1$).

As discussed by Manaka and coworkers,[2)] the relation between x and p is not trivial because the exchange paths between two Cu ions are bibridged bonds. For $x = 1$, the ferromagnetic bonds are Cu $<^{\rm Cl}_{\rm Cl}>$ Cu bonds, and for $x = 0$, the strong antiferromagnetic bonds are Cu $<^{\rm Br}_{\rm Br}>$ Cu bonds. The addition of the Br ions into the $x = 1$ chain induces the Cu $<^{\rm Br}_{\rm Cl}>$ Cu bonds which are absent in the pure chains. We assume, however, that the Cu $<^{\rm Br}_{\rm Cl}>$ Cu bonds are strongly antiferromagnetic because otherwise the Haldane phase cannot be destroyed and no magnetic order can take place for relatively small Br concentration ($1 - x \simeq 0.13$) as observed in the experiment. In this case, the concentration of the ferromagnetic bond $1-p$ is equal to x^2. Further, we may safely ignore the Cu $<^{\rm Br}_{\rm Br}>$ Cu bonds in the concentration regime $1 - x << 1$ which we are mainly concerned in this paper. Actually at the critical concentration $x = 0.87$, the concentration of Cu $<^{\rm Br}_{\rm Br}>$ Cu bonds is $(1 - x)^2 \sim 0.017$

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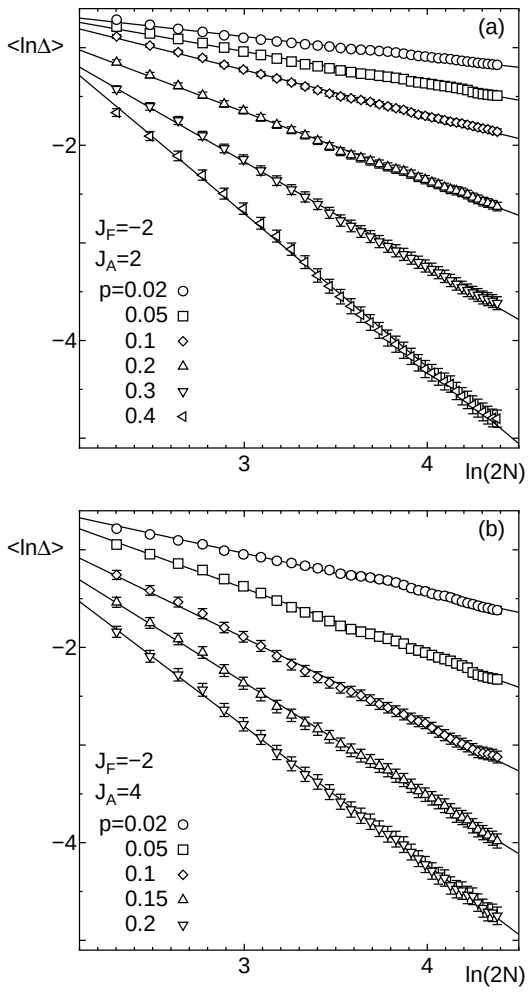

Fig. 1. The system size-dependence of $< \ln\Delta >$ for (a) $J_{\rm A} = 2$ and (b) $J_{\rm A} = 4$ with $J_{\rm F} = -2$. The solid line is the fit to the relation $\ln\Delta = \text{const.} - z\ln N$.

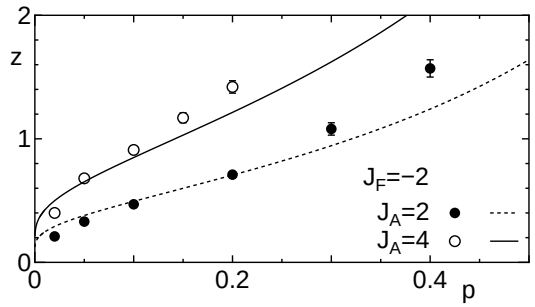

Fig. 2. The p-dependence of the dynamical exponent z. The circles are the estimation from the system size dependence of $< \ln\Delta >$. The lines are $z = \alpha/ \mid \ln p \mid$ where α is determined from the q dependence of Δ_q.

which is sufficiently small. We can therefore set $p = 1-x^2$ in this regime. In the following, we take $J_{\rm F} = -2J$ following the experimental estimation[1] and $J = 1$ to fix the energy unit.

In the present system, the spin gap state in the pure system is intrinsic. Therefore the mechanism of the disorder induced order is not related with the lattice degrees of freedom as in the spin-Peierls systems.[3–5] The situation is somewhat similar to the site depleted 2-dimensional $S = 1$ antiferromagnet studied by Yasuda and coworkers[8] if $J_{\rm F}$ and $J_{\rm A}$ bonds are infinitely strong. However, as explained later, the finiteness of these bonds introduces interesting fine structures in the single chain properties which should be observable in experiments.

This paper is organized as follows. In the next section, the single chain properties of the present model are discussed. In subsection 2.1, the low energy spectrum of the single chain is calculated and shown to have quantum Griffiths singularity. The physical origin of this behavior is also explained in the cluster picture. In subsection 2.2, the magnetization curve of this model is presented and the randomness induced plateau is shown to appear. The fine structures of the magnetization curve are also explained in terms of the cluster picture. In the third section, the effect of interchain coupling is studied by means of the interchain mean field approximation. It is shown that the disorder induced magnetic order takes place in appropriate concentration regime. The final section is devoted to summary and discussion. The brief report on the main results of this work is published in ref. 11.

2. Single Chain Properties

2.1 Low Energy Excitation Spectrum

To support the long range order in the coupled chain systems, enough number of low energy states are required in the spectrum of the constituent single chains. We therefore calculate the size dependence of the energy gap Δ of the random ensemble of spin chains described by Eq. (1). Due to computational reason, we concentrate on the regime of small p (small $1-x$) in the following. Figure 1 shows the system size dependence of the average of the logarithm of the energy gaps Δ with $J_{\rm F} = -2$ and (a) $J_{\rm A} = 2$ and (b) 4 for $10 \le 2N \le 80$ calculated by the density matrix renormalization group (DMRG) method.[10,19] The average is taken over 400 samples. In this parameter range, these curves are fitted well by $< \ln\Delta > \sim$ const. $- z\ln N$ where z defines the dynamical exponent. This implies that the energy gap is scaled by N^{-z} which is a typical behavior of the quantum Griffiths phase.[9,10] If we assume such scaling holds for all low energy excitation spectrum, the distribution function of the low lying excitation energies scales as

$$P(\Delta)d\Delta = N^z f(N^z\Delta)d\Delta \qquad (2)$$

where $P(\Delta)d\Delta$ is the number of states with excitation energy in the range $[\Delta, \Delta + d\Delta]$. For large N, $P(\Delta)$ should be proportional to the system size N, so that $f(x) \to x^{1/z-1}$ as $x \to \infty$. Thus, in the thermodynamic limit, we have

$$P(\Delta)d\Delta \propto N\Delta^{1/z-1}d\Delta. \qquad (3)$$

Therefore the number of the low energy states diverges if $z > 1$. As shown in Fig. 2, z increases with p and becomes larger than unity above a critical value $p_{\rm c}$ where $p_c \simeq 0.3$ for $J_{\rm A} = 2$ and $p_c \simeq 0.1$ for $J_{\rm A} = 4$. We can expect that the long range order would be stabilized for $p > p_{\rm c}$ if the interchain coupling is switched on.

These features of the low energy spectrum can be un-

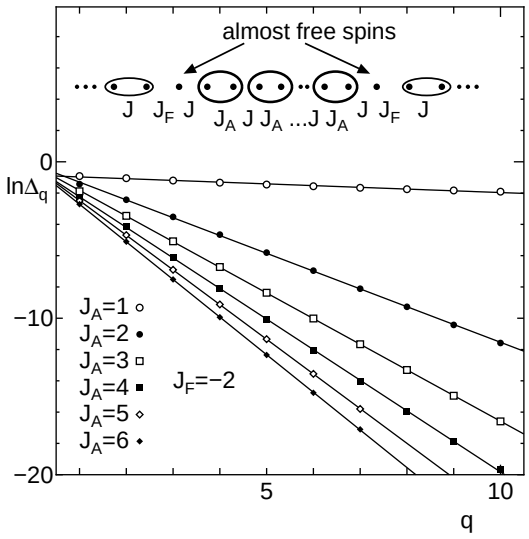

Fig. 3. The q-dependence of Δ_q. The inset shows the q-cluster.

derstood in the following way. Let us consider a cluster consisting of q $J_{\rm A}$-bonds and $q-1$ J-bonds embedded in the F-AF chain as depicted in the inset of Fig. 3. This is called 'q-cluster' in the following. The middle $2q$ spins form a tightly bound singlet cluster. The two spins connected to both ends of this cluster by J-bonds are almost free but weakly coupled with each other mediated by the quantum fluctuation within the strongly coupled cluster. For small p, we can regard the whole system as a random assembly of q-clusters. The q-dependence of the singlet triplet gaps Δ_q of q-clusters calculated by the DMRG method is well fitted by $\Delta_q \simeq \Delta_0 {\rm e}^{-\alpha {\rm q}}$ as shown in Fig. 3. On the other hand, the number of q-clusters in a chain is proportional to $Np^q(1-p)^2$. Eliminating q, the number $P(\Delta)d\Delta$ of the q-clusters with energy gap between Δ and $\Delta + d\Delta$ behaves as Eq. (3) with

$$z = \alpha / \mid \ln p \mid . \tag{4}$$

This value of z is plotted by the solid and dotted lines in Fig. 2. The agreement with the values obtained by fitting the numerical data of energy gap in Fig. 1 is good for small p. As p becomes larger, the interference between the clusters would prevent the above simple-minded interpretation.

Using the formula (4), we can estimate the parameter regime in which the low energy spectrum has divergent singularity as $p > p_c \equiv \exp(-\alpha)$. The critical value p_c is plotted against $J_{\rm A}$ in Fig. 4 using the values of α obtained from Fig. 3. For $p > p_c$, it is possible that the long range order is stabilized in the presence of interchain coupling.

The above picture explicitly demonstrates that the low energy excitation of the present model is dominated by the large size clusters and the characteristic size of the clusters increases by the power law with the decrease of the energy scale. This is a typical feature of the Griffiths phase. This singular excitation spectrum is reflected in the low temperature magnetic susceptibility χ and magnetic specific heat C as $\chi \sim T^{1/z-1}$ and $C \sim T^{1/z}$.

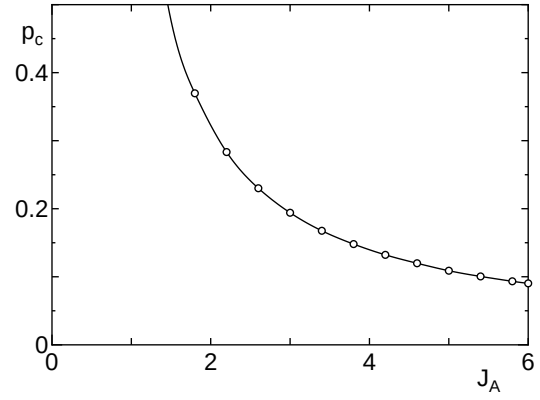

Fig. 4. The parameter regime in which the low energy spectrum has divergent singularity.

2.2 Magnetization Curve

The magnetization curves at $T = 0$ is calculated by the finite size DMRG method for $2N = 100$ for $J_{\rm A} = 2$ and 4 with $J_{\rm F} = -2$ as shown in Fig. 5(a) and (b), respectively. The average is taken over 100 samples. A big plateau appears at $M = M_s(1-p)$ where M_s is the saturated magnetization and small jumps appear below and above the main plateau. Again, the physical interpretation of this structure can be given in terms of the q-clusters. On the main plateau the spins connected by $J_{\rm A}$-bonds form singlet clusters and remaining spins are polarized along the direction of the magnetic field. These plateaux are well quantized so that it must be useful to determine p directly from experimental data. Similar fractional plateaux has been also found in random polymerized XXZ chains.[12)]

Recently, Totsuka[13)] analytically discussed the effect of randomness on the magnetization plateaux. At first sight, our fractional plateau appears to contradict with his criterion (Eq. (36) of ref. 13). However, his argument concerns the fate of the plateaux which already exist in the regular system and does not rule out the plateaux induced by the bond randomness in our model as well as in the model in ref. 12 as commented by Totsuka himself at the end of his paper.[13)]

The small jumps above the plateau are the contribution from the spins connected by the $J_{\rm A}$ bond and those below the plateau are the contribution from the pairs of almost free spins which support the low energy magnetic exciations. The position of these jumps are identified by the DMRG calculation for the q-clusters as indicated by the up and down directed triangles in Fig. 5(a) and (b) for each q. The position of the main jumps above the plateau $H_{\rm aq}$ and those below the plateau $H_{\rm bq}$ due to the q-cluster are depicted in Fig. 6 against $J_{\rm A}$ for $q = 1$ and 2. Because $H_{\rm a1}$ is sensitive to $J_{\rm A}$, we could determine $J_{\rm A}$ from experimental value of $H_{\rm a1}$. However, considering that the saturation field of the $x = 1$ compound ($p = 0$) is already above 40T,[18)] the observation of this jump would be rather difficult within the presently available pulse magnetic field $\sim$ 80T unless $J_{\rm A}$ is relatively small. Although less precise, $J_{\rm A}$ could be also determined from experimental data for $H_{\rm b1}$ which should be observ-

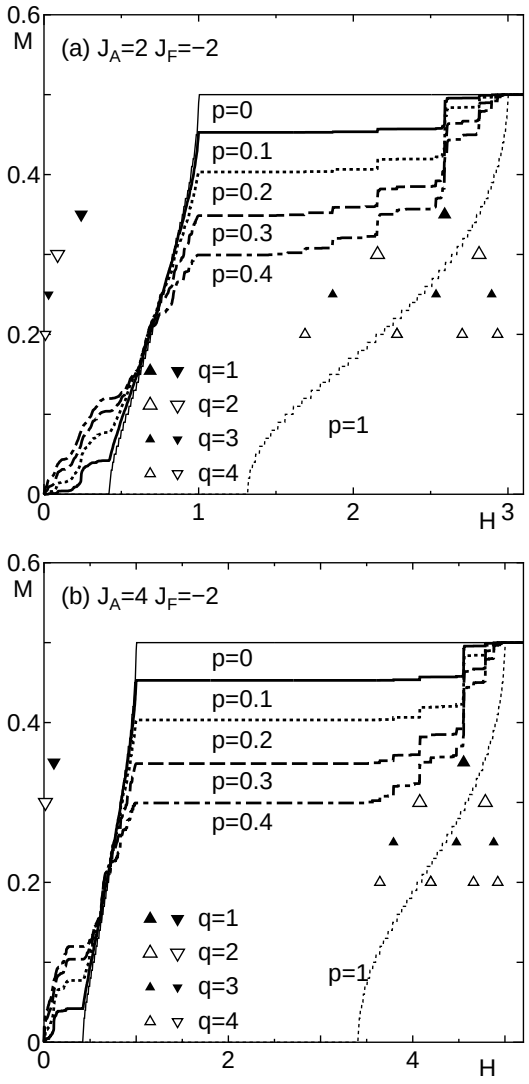

Fig. 5. Magnetization curves for (a) $J_{\rm A}=4$ and (b) $J_{\rm A}=2$ with $J_{\rm F}=-2$ and $2N=100$ for various values of p. The magnetic field and the magnetization per site are plotted in units of $J/g\mu_{\rm B}$ and $g\mu_{\rm B}$, respectively.

able within the presently available magnetic field. In recent magnetization measurement,[2] however, no prominant structure is observed in real IPACu(Cl_xBr_{1-x}) for $x \geq 0.87$ in the low field regime. The origin of this discrepancy is unclear at present.

3. Effect of Three Dimensionality - Disorder Induced Magnetic Order

As demonstrated above, the low energy sectors of the present model is dominated by the almost free spins in q-clusters with large q. In the presence of the interchain coupling, we therefore expect that the 3 dimensional network of these spins sustain the magnetic order observed in the experiment mediated by the interaction with spins in neighbouring chains. Let us assume the Hamiltonian with the interchain coupling as follows,

$$H = \sum_j [\sum_{i=1}^{N} J\boldsymbol{S}_{2i-1,j}\boldsymbol{S}_{2i,j} + \sum_{i=1}^{N} J_i \boldsymbol{S}_{2i,j}\boldsymbol{S}_{2i+1,j}]$$

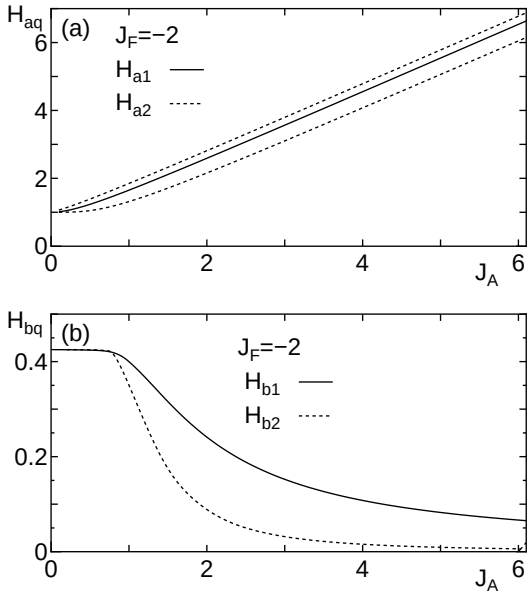

Fig. 6. The $J_{\rm A}$-dependence of the magnetic fields where the jumps due to $q=1,2$ clusters take place (a) above the plateau (solid line: $H_{\rm a1}$, two dotted lines: $H_{\rm a2}$) and (b) below the plateau (solid line: $H_{\rm b1}$, dotted line: $H_{\rm b2}$). $J_{\rm F}=-2$.

$$+ \sum_{i=1}^{2N} \sum_{<j,j'>} J_\perp \boldsymbol{S}_{i,j}\boldsymbol{S}_{i,j'}, \qquad (5)$$

where j and j' distinguish the chains and $\sum_{<j,j'>}$ denotes the summation over nearest neighbour chains. We assume that the correlation between two almost free spins separated by the $J_{\rm A}$ bonds is mainly mediated by the F-AF part of the neighbouring chains, because the correlation length of the AF-AF part is small compared to that of the F-AF part. Furthermore, for small p, the probability to find the $J_{\rm A}$ bonds on the neighbouring chains between two almost free spins is small. As a result, the interchain interaction and the short range correlation within the neighbouring chains support the $\uparrow\uparrow\downarrow\downarrow$-type long range order as far as p is small.

For quasi-one-dimensional systems, the mean field approximation for the interchain coupling gives reliable results,[20] because a large number of spins are envolved in the low energy long wave length fluctuations in each chain. These fluctuations are frozen by interchain interaction resulting in the three dimensional ordering. We therefore employ the interchain mean field approximation assuming the $\uparrow\uparrow\downarrow\downarrow$ order to obtain the single chain mean field Hamiltonian given by,

$$H^{MF} = \sum_{i=1}^{N} J\boldsymbol{S}_{2i-1}\boldsymbol{S}_{2i} + \sum_{i=1}^{N} J_i \boldsymbol{S}_{2i}\boldsymbol{S}_{2i+1} - \sum_{i=1}^{2N} h_i S_i^z \quad (6)$$

with $h_i = -z_{\rm c}J_\perp < S_i^z >$ and $z_{\rm c}$ is the number of nearest neighbour chains. Reflecting the $\uparrow\uparrow\downarrow\downarrow$ order, we take $h_{4i} = h_{4i+1} = h$ and $h_{4i+2} = h_{4i+3} = -h$. In Fig. 7, the staggered magnetization $m_{\rm st} \equiv < \frac{1}{2N}\sum_{i=1}^{N}(-1)^i(S_{2i}^z + S_{2i+1}^z) >$ is plotted against $\lambda \equiv z_{\rm c}J_\perp$ for (a) $J_{\rm A}=2$ and (b) $J_{\rm A}=4$ with $J_{\rm F}=-2$. The chain length is $2N=200$ and finite size DMRG method is used. The average is taken over 200 samples. For $p=0$, $m_{\rm st}$ vanishes unless

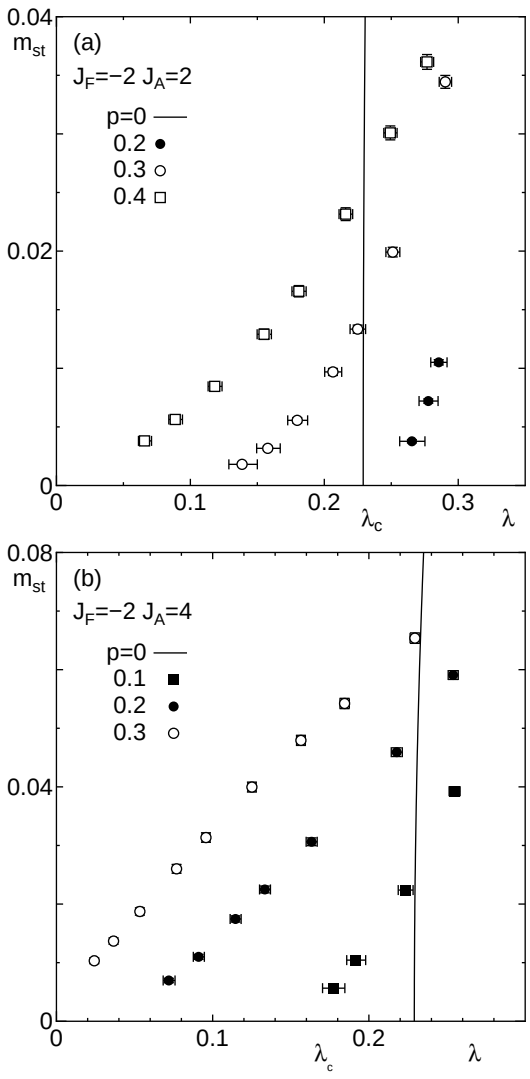

Fig. 7. The λ-dependence of staggered magnetization for (a)$J_{\rm A} = 2$ and (b)$J_{\rm A} = 4$ with $J_{\rm F} = -2$. The solid line is the staggered magnetization for $p = 0$.

λ is larger than a critical value λ_c. For finite p, however, $m_{\rm st}$ takes a finite value even for small $\lambda < \lambda_c$. The magnitude of $m_{\rm st}$ increases with p and $J_{\rm A}$, namely with the increase of randomness. It should be noted that the long range order for $\lambda < \lambda_c$ starts to appear around $p \sim 0.3$ for $J_{\rm A} = 2$ and around $p \sim 0.1$ for $J_{\rm A} = 4$. Theses values are approximately consistent with the estimation of p_c from the energy gap scaling. Therefore the $\uparrow\uparrow\downarrow\downarrow$-type long range order is stabilized for appropriate strength of the interchain coupling if the low energy density of state has divergent singularity as expected.

In the experiment, the long range order is observed for $x < 0.87$. If we assume $p = 1 - x^2$, the regime $x < 0.87$ corresponds to $p \gtrsim 0.24$. Although the exact value of $J_{\rm A}$ is unknown, we find from Fig 7 that the long range order is stabilized in this concentration range even for $\lambda < \lambda_c$ both for $J_{\rm A} = 2$ and 4. These results explain the qualitative features of the experimental observation of the magnetic ordered state. For the quantitative comparison with experiments, the strength of the Cu $<^{\rm Br}_{\rm Cl}>$ Cu bonds need to be determined. Similar analysis is made for the random $S = 1$ Heisenberg antiferromagnet by Villar *et al.*[14]

4. Summary and Discussion

The $S = 1/2$ Heisenberg chains with bond alternation and randomness on the strong bonds are studied by the DMRG method. The low energy spectrum is shown to have the Griffiths type singularity and the physical origin of this behavior is explained based on the q-cluster picture. In the magnetization curves, there appears a randomness induced plateau at the fractional value of the saturated magnetization. This plateau and the fine sructures of the magnetization curves are also understood based on the q-cluster picture. By the mean field treatment of the interchain coupling, the magnetic long range order is shown to be stabilized by randomness in the intermediate concentration regime. The results are discussed in relation with the recent experiment for $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$.

We carried out the interchain mean field calculation assuming the $\uparrow\uparrow\downarrow\downarrow$ type long range order. However, the possibility of different type of ordering such as spin glass ordering cannot be excluded. From this viewpoint, the experimental determination of the magnetic structure by neutron scattering experiment is also hoped to elucidate the nature of the long range order.

Recently, Nakamura[15] has shown that the $\uparrow\downarrow\uparrow\downarrow$ correlation becomes most critical in the intermediate concentration regime in the present model from the nonequilibrium relaxation method analysis of the quantum Monte Carlo data. We have also checked the possibility of the $\uparrow\downarrow\uparrow\downarrow$ order within the DMRG and interchain mean field approximation. According to our calculation, the staggered magnetization for $\uparrow\downarrow\uparrow\downarrow$ order is much lower than that for the $\uparrow\uparrow\downarrow\downarrow$ order. For example, for $J_{\rm A} = -J_{\rm F} = 2$ and $p = 0.4$, the staggered magnetization for the $\uparrow\downarrow\uparrow\downarrow$ order is approximately one order of magnitude smaller than that for the $\uparrow\uparrow\downarrow\downarrow$ order. For other values of parameters $J_{\rm A}, J_{\rm F}$ and p, the ratio of $\uparrow\downarrow\uparrow\downarrow$ order to $\uparrow\uparrow\downarrow\downarrow$ order is even smaller. Especially, the $\uparrow\downarrow\uparrow\downarrow$ order decreases rapidly with the decrease of p and becomes less than 10^{-3} for $p \leq 0.3$ with $J_{\rm A} = -J_{\rm F} = 2$ and for $p \leq 0.2$ with $J_{\rm A} = 4$, $J_{\rm F} = -2$ within the regime $\lambda < \lambda_c$. Therefore we may conclude that the dominant order is of the $\uparrow\uparrow\downarrow\downarrow$ type although it is possible that the weak $\uparrow\downarrow\uparrow\downarrow$ order also coexist. It should be noted that Nakamura's calculation does not explicitly include the effect of interchain coupling. In the random systems, the criticality of the correlation in a single chain does not always imply the corresponding long range order in its quasi-1-dimensional counterpart.

The absence of the fine structure in the low field magnetization curve of the real IPACu(Cl_xBr_{1-x}) is not understood in the present calculation. In the presence of interchain coupling, however, it is possible that the almost free spins form localized singlet clusters mediated by interchain coupling. This can suppress the low field structure in the magnetization curve. However, such local correlation effect is not properly described within the present interchain mean field approximation. This problem is left for future studies.

In this work, we concentrated on the ground state

properties due to the limitation of the numerical method (DMRG). The finite temperature effects must be important for the direct comparison with experiments. This is left for future studies.

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