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Ground State of Antiferromagnetic Quantum Spin Systems on the Triangular Lattice

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The spin-1/2 antiferromagnetic Heisenberg and XY models are investigated on the triangular lattice by numerically diagonalizing finite size systems (up to 21 spins). The ground state energies of both models are found to be close to other theoretical predictions except those of Marland and Betts who performed similar analysis to ours. We have numerically constructed a resonating valence bond state of Anderson. It has a relatively large projection on the ground state of the finite Heisenberg model, although the absolute value of the projection decreases linearly with the system size. The trial function proposed by Miyashita for the XY model does not have an appreciable projection on the numerically exact state. We present evidence for the absence of a sublattice long range order in the Heisenberg model and the existence of the same quantity in the XY model.

§1. Introduction

Frustration is known to cause unusual cooperative effects in spin systems. Typical examples are found in spin glasses,[1] but nonrandom systems also exhibit unexpected behavior[1] when frustration is introduced in a macroscopic scale. In particular, when the problem is quantum mechanical, we often do not have agreement even on what to choose to characterize the ground state. Antiferromagnetic systems on the triangular lattice are probably the best studied case of such, and we restrict our argument to this lattice. A classical picture is established on the ground state of the antiferromagnetic Heisenberg and XY models on the triangular lattice. The system has three sublattices and neighboring spins are oriented 120° relative to each other. A double degeneracy exists because of the degree of freedom in the relative angle of 120° or $-120°$. This classical state forms the basis of spin wave analyses.[2-4] A fully quantum-mechanical version of the three-sublattice picture has also been proposed.[5] Anderson,[6] on the other hand, suggests a quite different wave function as an approximate ground state of the Heisenberg model. His resonating valence bond (RVB) state consists of products of singlet pairs distributed on the whole lattice. This state apparently does not have a sublattice long range order in contrast to the classical ground state. Numerical analysis of finite size systems is another possible approach to the present problem and has been carried out by using the renormalization group method[7] and by numerical diagonalization of finite size systems.[8] From the numerical work, information on the energy and degeneracy of the ground state has been obtained. The ground state energy obtained by the numerical study showed marked discrepancy from spin wave and variational estimates.

The problems are thus summarized as follows. First we should determine the ground state energy. Concerning the spin state, the principal outstanding question is whether a sublattice long range order exists in relation to the validity of the classical picture. It is also important to acquire a knowledge on whether the proposed trial functions[5,6] represent essential features of the correct ground state. We present here the results of numerical investigation to answer these questions. Our approach is basically the same one as that of Marland and Betts[8] (referred to as MB) who have numerically diagonalized finite size systems. The only difference lies in the amount of calculations: MB have diagonalized lattices of size $N=7$, 9 and 12 with specific lattice shapes and with periodic boundary conditions. We have analyzed systems of $N=4$ to 21 with both periodic and free boundaries. For most N, a few different lattice shapes have been investigated. By the railroad trestle extrapolation[6] we have then estimated the ground state energy and spin state of the infinite system. The resulting values of the ground state energy place those of MB out of any reasonable extrapolation errors. We have found strong evidence that the Heisenberg model does not have a sublattice long range order while the XY model possibly has one. We have numerically generated an RVB state and have found that it has a relatively large projection on the exact finite size eigenstate of the Heisenberg model. But the absolute value of the projection turns out to decrease linearly with the system size up to the lattice size we have investigated. The most plausible conclusion is that our RVB state does not have a finite fraction in the exact ground state of the infinite Heisenberg model. The trial function of Miyashita[5] for the XY model has a much smaller projection (than the RVB state does) in finite size systems and the value of projection decays roughly exponentially with the system size.

The framework of this paper is as follows. In §2 some technical details of our numerical calculations are explained. The ground state energy is evaluated in §3 and the appropriateness of the trial wave functions is discussed in §4. The sublattice long range order is examined in §5 followed by general discussions in §6.

§2. Numerical Techniques

In a numerical diagonalization of a spin Hamiltonian it is customary[9] to reduce the size of matrices by using symmetries of the Hamiltonian. The present Hamiltonian

$$H=2J\sum_{\langle ij\rangle}(S_i^xS_j^x+S_i^yS_j^y+\Delta S_i^zS_j^z), \qquad (1)$$

with $J>0$ ($\Delta=1$ represents the Heisenberg model while $\Delta=0$ corresponds to the XY model) commutes with the z-component S_{tot}^z of the total spin. Hence we have chosen the space of $S_{\text{tot}}^z=0$ (or 1/2 if N is odd) to look for the ground state. For the Heisenberg model the Hamiltonian (1) commutes with all components of the total spin and thus the space of minimal S_{tot}^z always includes the ground state. As for the XY model, however, no such symmetry argument justifies our restriction of $S_{\text{tot}}^z=0$ (or 1/2). Nevertheless we do not expect that the XY model magnetizes spontaneously along the z-axis, which physically supports our choice. Time reversal and space symmetries are often used[9] to further reduce the size of matrices. We did not follow this step because our computer program runs faster if the bases of matrices are simpler (we generate matrix elements as we diagonalize a matrix). The largest dimensions of matrices we have diagonalized are 352716 (which is the number of combinations to choose 11 up spins out of $N=21$ sites) for the ground state energy and 184756 (corresponding to $N=20$) for the wave function. The ground state energy was calculated by the Lanczos method and the eigenstate was obtained by the inverse iteration method. Each step of the inverse iteration was carried out by the conjugate gradient method. All the necessary variables (in double precision) were stored in the core memory to save I/O time. For each initial vector of the Lanczos or inverse iteration process, the CPU time to reach convergence on HITAC S-810/20 at the Computer Centre of the University of Tokyo was about two minutes for the largest matrices we have diagonalized.

§3. Ground State Energy

We have estimated the ground state energies of the Heisenberg and XY models by the railroad trestle extrapolation.[6] As seen in Figs. 1 and 2, first the width of the railroad trestle is fixed and the length is increased. The boundary is free. The lowest energy is plotted as a function of the inverse of the length to estimate the infinite-length energy which is then plotted as a function of the inverse of the width. The final extrapolation to the infinite width gives the ground state energy of the two-dimensional infinite system. Both for the Heisenberg and XY models each step of extrapolation has been successfully performed because of excellent linear dependence of the energy on the inverse of the length or width of the railroad trestle. Slight deviations are found from linearity in the series of width

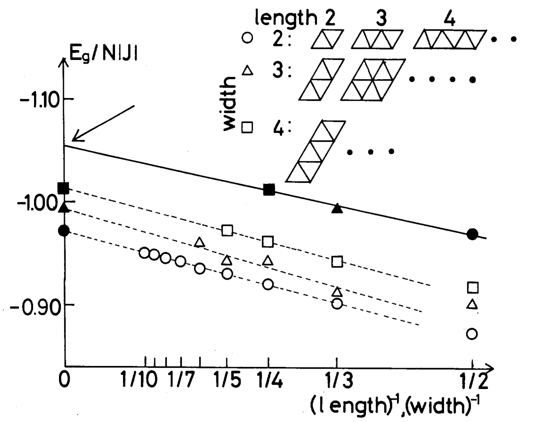

Fig. 1. The ground state energy of the Heisenberg model on railroad-trestle lattices is given as a function of the inverse of the length and width of railroad trestles. The width of the railroad trestle is first fixed to estimate the infinite-length energy (● is for width two, ▲ for width three and ■ for four). The next step is to increase the width to estimate the infinite-width energy as marked by an arrow.

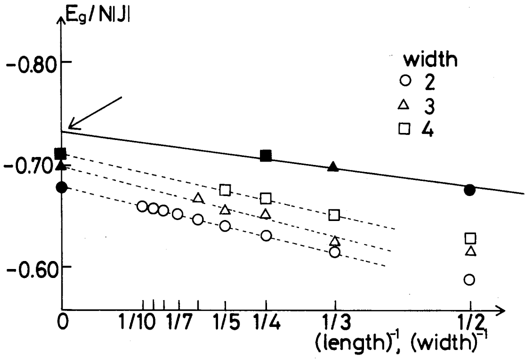

Fig. 2. The same analysis as in Fig. 1 is carried out for the *XY* model.

three in both models. These discrepancies probably originate in the difference of S^z_{tot}: If the width and length are both odd numbers, the total spin assumes only half odd integers (represented by $S^z_{tot}=1/2$ in our calculations). We have thus tried two different extrapolations in the case of width three. One is a simple least square fit of all the data and the other is to average extrapolated values from even- and odd-length railroad trestles. The results were independent of the way of extrapolation within the significant figures we have pursued.

The final ground state energies are -1.05 and -0.74 (in units of NJ) for the Heisenberg and XY models respectively. These values

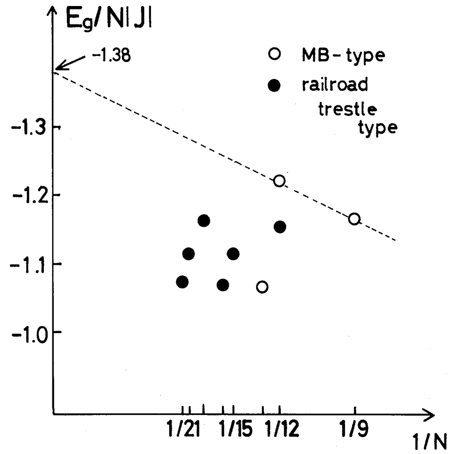

Fig. 3. The ground state energy of finite Heisenberg models with periodic boundary conditions. The sizes and shapes of lattices we have investigated are $N=9$, 12, 13 (MB-type diamond shaped lattices marked ○), $N=3\times4$, 3×5, 3×6, 3×7, 4×4, 4×5 (railroad-trestle type lattices marked ●: $N=\text{width}\times\text{length}$). The extrapolation by MB is given in a dashed line.

show marked discrepancies from those of MB. They predict -1.38 and -0.89 respectively by extrapolating from data on diamond-shaped lattices of $N=9$ and 12 with periodic boundaries. For comparison we have calculated the energies of various lattices from $N=9$ to 21 with periodic boundary condition. The result is given in Figs. 3 and 4 as a function of the inverse of the system size. We conclude that the extrapolation of MB does not lead to quan-

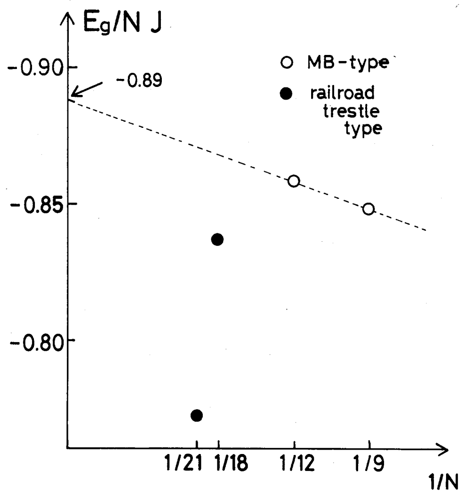

Fig. 4. The same plot as in Fig. 3 for the XY model. The lattices are $N=9$, 12 (MB type ○) and $N=3\times6$, 3×7 (railroad trestles ●).

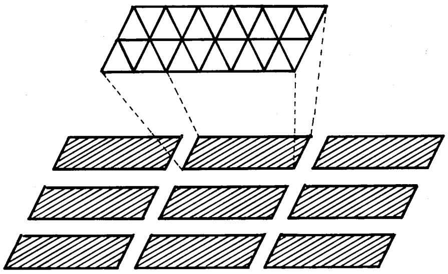

Fig. 5. The whole lattice is covered with 3×7 lattices with boundaries shared with neighboring ones. The lowest energy of a 3×7 lattice yields a lower bound on the ground state energy of the infinite lattice (times a trivial topological factor).

titatively reliable values. We have also obtained numerically-exact lower bounds of the ground state energy by a variational method originally due to Anderson.[10] The infinite system is tiled with finite lattices of width 3 and length 7 with boundary exchange J (interior $2J$) as shown in Fig. 5. Then the ground state energy of a 3×7 lattice provides a lower bound on the infinite-lattice energy (times a trivial topological factor). It is not essential to work with a 3×7 lattice, but this case has given the best bounds. The results are -1.26 (Heisenberg) and -0.87 (XY), which unambiguously exclude the values of MB. Other predictions on the ground state energy are listed in Table I. Our estimates are in fair agreement with most results except those by MB and by the renormalization group.[7] The renormalization group method accepts uncontrollable approximations which should prevent one from drawing quantitative conclusions on non-universal variables such as the energy. Our ground state energy -0.74 of the XY model is higher than that of Miyashita[5] -0.79 by a variational method. The latter is an upper bound on the exact value. Hence the difference between -0.74 and -0.79 (6.8%) gives the limit of confidence of our extrapolation in the case of the XY model. A remark is made here on the ground state degeneracy. We have found no degeneracy in the ground states of free-boundary lattices in the series of railroad trestles. Even with periodic boundaries most instances did not show ground state degeneracies. Therefore we conclude, in agreement with MB, that frustration in quantum spin systems does not lead to degenerate ground states.

§4. Trial Wave Functions

The RVB state proposed by Anderson[6] as an approximate ground state of the Heisenberg model consists of products of singlet pairs distributed on the whole lattice (Fig. 6). A linear combination of these products (summed over all possible ways to distribute singlet pairs) is the trial wave function:

$$|\psi_{\mathrm{RVB}}\rangle=\sum_{\mu} c_{\mu} \prod_{\langle ij\rangle} (\alpha_i\beta_j-\beta_i\alpha_j), \tag{2}$$

where α denotes an up spin and β a down spin. A finite temperature version of (2) has also been proposed by Suzuki.[11] Anderson[6] states that the coefficients c_μ can be determined by starting from an arbitrary configuration μ and successively operating the off-diagonal part of the Hamiltonian (1). However he did not prove the consistency of this process (see also ref. 12). Therefore we have instead determined the coefficients c_μ in (2) according to the following rule. The absolute value of c_μ is simply a constant (independent of μ). The sign is fixed positive by ordering the product over the pairs $\langle ij\rangle$ such that $i>j$. The site index (i, j, etc) was assigned by the rule displayed in Fig. 7. This is the RVB state we have constructed. It should be recognized that a change in the sign of c_μ could yield quite different results on physical quantities (which we have observed in several instances). However, for small lattices ($N\leqq10$) we have learned by trying out a few different choices of the sign that the above-mentioned assignment gives the best results. The correct determination of the phase factor

Table I. Ground state energy of the Heisenberg (H) and XY models on the triangular lattice.

	Our estimate (Railroad trestle)	Our estimate (Lower bound)	Spin wave[4] to $O(1/S^2)$	Variational[5]	RG[7]	MB[8]
H	-1.05	-1.26	-1.09	-1.01	—	-1.38
XY	-0.74	-0.87	-0.81	-0.79	-0.88	-0.89

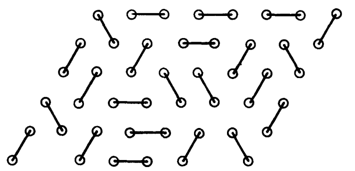

Fig. 6. Singlet pairs are distributed on the whole lattice covering all sites. A linear combination of the products of these singlet pairs is the RVB state.

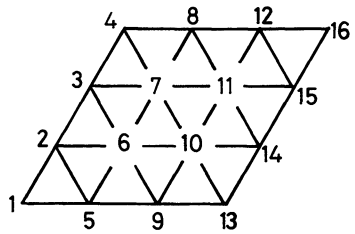

Fig. 7. For a given finite lattice in the series of railroad trestle (with free boundary), we assign the site index starting from the left-bottom corner. The index is increased as the site location proceeds upward until the left-top site is reached. Then we return to the bottom line and choose the second-leftmost site as the next site. This process is repeated until all sites are exhausted.

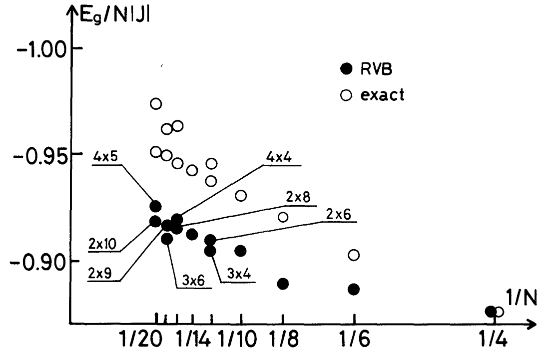

Fig. 8. The variational energy of the RVB state (●) is given for various finite lattices. For comparison the exact energy (○) of the corresponding lattices are also exhibited. The width-length combinations are indicated if there are two of them for a given N.

is the essential part in constructing a meaningful trial function, and this problem will be discussed in §6. The variational energy thus obtained is shown in Fig. 8. The largest error is 5% ($N=4\times5$), a rather impressive value. Next we have taken projection of our RVB state on the exact ground state of a finite system: $f=|\langle\psi_{\rm exact}|\psi_{\rm RVB}\rangle|^2$. f represents the fraction of the RVB state in the correct ground state. To estimate the value of f in the infinite system we have plotted f for each width of railroad trestles as in Fig. 9. It is seen that f decreases linearly with the length for a fixed width, suggesting a vanishing fraction for large enough systems. Of course it could happen that f saturates to a finite value if larger systems are investigated, particularly because the smallest f we have found was 0.728 ($N=3\times6$; not shown in Fig. 9) which is not a negligible number.

The trial wave function of Miyashita[5] for the XY model on the triangular lattice starts with the variational state first proposed by Suzuki and Miyashita[13] for the ferromagnetic

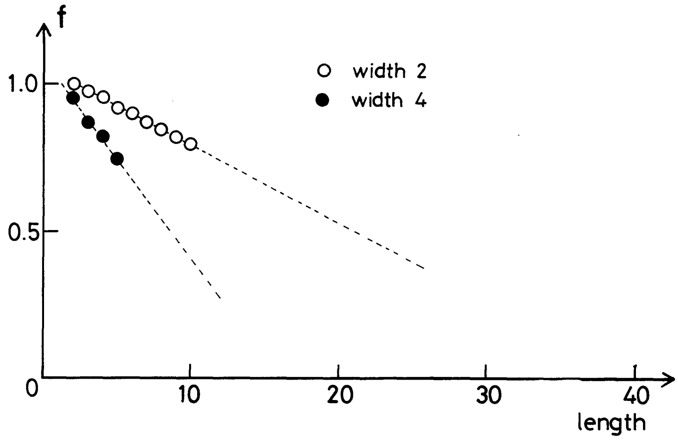

Fig. 9. The fraction f of the RVB state included in the exact ground state of finite Heisenberg systems is given as a function of the length of railroad trestles. The symbol ○ denotes the lattices with width two and ● is for width-four series. Both series are seen to give linearly decaying f.

XY model:

$$|\psi\rangle=\exp\,(-a\sum_{\langle ij\rangle}S_i^zS_j^z)\,|\psi_0\rangle, \qquad (3)$$

where a is a variational parameter and $|\psi_0\rangle$ is the eigenstate of total spin with $S_{\rm tot}=N/2$ (the maximum value) and $S_{\rm tot}^z=0$ (assuming N is even). To take account of the possible three-sublattice structure of the antiferromagnetic triangular system, the state (3) is rotated $\pm120°$ (or $0°$) in each sublattice:

$$|\tilde{\psi}\rangle=\exp\,(2\pi i\sum_A S_j^z/3-2\pi i\sum_B S_j^z/3)\,|\psi\rangle, \quad (4)$$

where the first sum is over one of the sublattices A and the second runs over another sublattice B. This trial function yields excellent ground state energy by optimizing a (see Table I). However the fraction f of (4) in the exact ground state is not large as shown in Fig. 10. For each width of railroad trestles f decays roughly exponentially with the length, thus implying orthogonality of (4) to the correct ground state in the limit of the infinite system.

§5. Long Range Order

In the classical antiferromagnetic Heisenberg and XY models on the triangular lattice the ground states have a sublattice long range order. There are three sublattices and spins on a sublattice have a relative angle of 120° to spins on another sublattice. Spin wave theory[2-4] also predicts a finite sublattice long range order. The variational function of Miyashita (4) in the preceding section sustains the three-sublattice structure. On the other hand the RVB apparently does not have a conventional long range order. It is therefore an interesting test to see if an extrapolation from finite size systems predicts a sublattice long range order. We have calculated the expectation values of the operators

$$M_H=(\sum_A S_j^z+\sum_B R_yS_j^zR_y^{-1}+\sum_C R_y^{-1}S_j^zR_y)^2,$$
$$M_{XY}=(\sum_A S_j^x+\sum_B R_zS_j^xR_z^{-1}+\sum_C R_z^{-1}S_j^xR_z)^2, \quad (5)$$

for the Heisenberg and XY models respectively. Here the sums over A, B and C are defined as in (4) and R_y and R_z are rotation operators, $R_{y,z}=\exp\,(-2\pi iS_j^{y,z}/3)$. If a sublattice long range order exists, the expectation value of M_H (or M_{XY}) normalized by N^2 tends to a finite number as $N\rightarrow\infty$ (because $\langle S_j^zS_{j+r}^z\rangle$ (or $\langle S_j^xS_{j+r}^x\rangle$) approaches a finite value as $r\rightarrow\infty$).[14] We have thus plotted the logarithm of $\langle M_H\rangle/N^2$ and $\langle M_{XY}\rangle/N^2$ as functions of the logarithm of the length for each width of railroad trestles (Figs. 11 and 12). The slope of the ex-

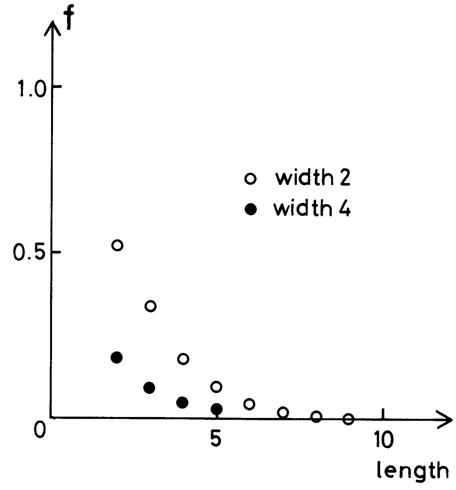

Fig. 10. The fraction f of the trial function of Miyashita for the XY model decreases rapidly as the length of the railroad trestle is increased (○ is for width-two series and ● represents the lattices of width four).

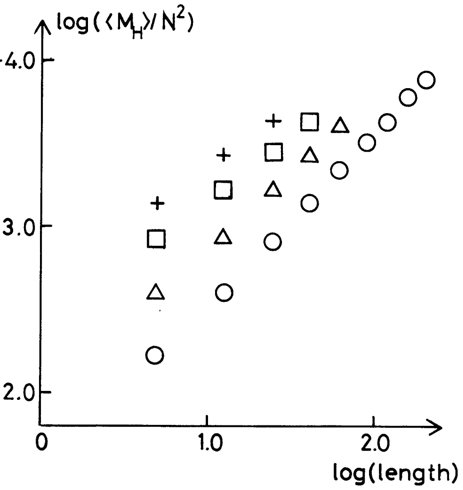

Fig. 11. A log-log plot of the order parameter (normalized by N^2) versus the length of the railroad trestle for the Heisenberg model. Different symbols are given to different-widths railroad trestles. ○, △, □, + represent width two, three, four and five respectively. The slope in this figure is the exponent k as explained in the text.

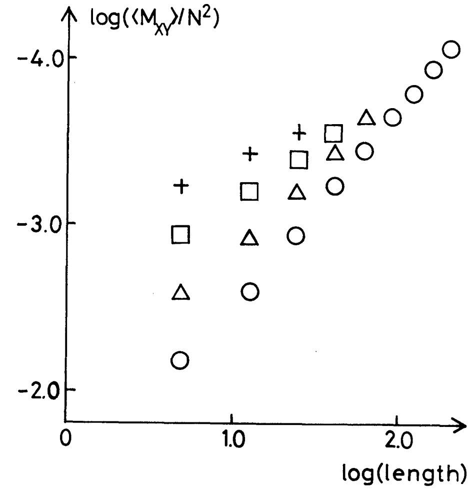

Fig. 12. The same plot as in Fig. 11 for the XY model.

trapolation line in these figures give the power k in $\langle M_{H,XY}\rangle/N^2 \sim N^{-k}$. Here N is proportional to the length of a railroad trestle (since we now fix the width). The extrapolated value of k of each width is then plotted as a function of the inverse of the width to estimate k in the limit of the infinite lattice (Fig. 13). Finiteness of k means the absence of a long range order. For the Heisenberg model the series apparently tend to a finite value, while those of the XY model may have a vanishing k. For comparison the same exponent k is given in Fig. 13 calculated from the RVB state: $\langle \psi_{RVB}|M_H|\psi_{RVB}\rangle/N^2$. It is clearly seen that

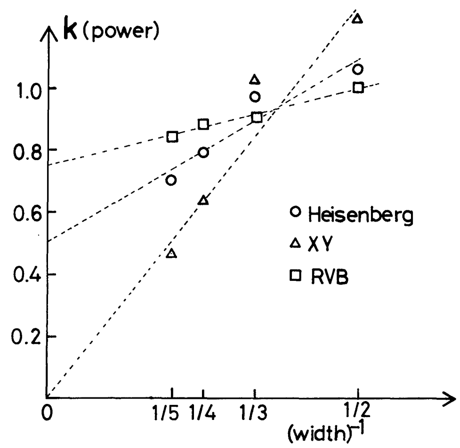

Fig. 13. The exponent k of the long range order is given as a function of the inverse of the width of the railroad trestle. Three cases (Heisenberg ○, XY △ and RVB □) show apparently distinct behavior for large systems.

the RVB state has a rapidly decaying correlation function, i.e. a large k ($\lesssim 1$). Although we cannot definitely exclude the possibility of the absence of a long range order in the XY model, the correlation function of the XY model decays, at least, much more slowly than in the Heisenberg model. It is useful to remember that similar calculations by Oitmaa and Betts[14] on the unfrustrated square lattice lead to a finite sublattice long range order for the Heisenberg (and XY) model. Thus we conclude that frustration is the principal reason for the absence of the long range order in the antiferromagnetic Heisenberg model on the triangular lattice.

§6. Discussion

We have estimated the energy and spin state of the Heisenberg and XY models on the triangular lattice. Our extrapolation by the railroad trestle method (with free boundary) gives satisfactory values of the ground state energy for the Heisenberg model. The estimated lowest energy of the XY model has been higher than the upper bound[5] by 6.8%. Periodic boundary conditions as employed by MB do not allow us to predict the energy of the infinite system in a systematic way. As for the spin state of the Heisenberg model, the ground state does not have a conventional sublattice long range order, which invalidates the classical picture. Recent experiments on $NaTiO_2$[15] (which is considered as a realization of the present Heisenberg model) are consistent with our conclusion. The absence of ground state degeneracy in most finite systems is another support for the breakdown of the classical ground state. The RVB state is one of the plausible alternatives. But the fraction of our RVB state in the correct ground state has been found to decrease linearly with the system size at least up to $N=20$. The XY model has a correlation function decaying much more slowly than in the Heisenberg model. One may reasonably expect a finite sublattice long range order in the XY model. We nonetheless stress that the absence of degeneracy is sufficient to upset the classical predictions on the ground state properties. The trial function of Miyashita with a sublattice long range order does not have an appreciable projection of the exact ground state.

In relation to the appropriateness of a trial wave function we point out here the importance of the phase factor. If the system is unfrustrated, all coefficient in an expansion of the ground state can be chosen positive after a trivial canonical transformation.[16-18] Accordingly a trial function with positive coefficients would necessarily have a nonvanishing projection on the exact ground state even for large enough systems. (This should be the case, for instance, in the variational state (3) of Suzuki and Miyashita[13] for the XY model.) For frustrated cases, however, no general statement has been proved on the phase factor. The concept of phase coherence[6,12] is a plausible candidate, but no rigorous argument for its validity is available as far as the present authors know. Our phase-assignment rule as explained in §4 has been designed as a step towards a practical prescription to construct the phase coherent state, but the result has not been fully successful. As for the trial function of Miyashita on the XY model, the phase factor in his function (4) would be too simple to warrant a non-negligible fraction f in large systems. Both in the Heisenberg and XY models, the essential information on the structure of the frustrated ground state should be found in the phase factor. A successful trial function is the one which correctly reflects the effects of frustration on phases.

After the completion of the work we learned that S. Fujiki and D. D. Betts were also carrying out numerical analysis of the present problem.

Acknowledgement

Stimulating discussions with Dr. Seiji Miyashita and Professor Masuo Suzuki are gratefully acknowledged. We also would like to express our gratitude to Professors Satoru J. Miyake, Komajiro Niizeki and Dr. Hikaru Kawamura for useful comments and to Professor Kinshiro Hirakawa for informing us of his experiments prior to the publication of his paper.

References

1) J. L. van Hemmen and I. Morgenstern (ed.): *Heidelberg Colloquium on Spin Glasses, Lecture Notes in Physics No. 192* (Springer, 1984).
2) T. Oguchi: *Proc. Int. Meet. Transition to "New Type of Ordered Phase", Kyoto, 1982*, J. Phys. Soc. Jpn. **52** (1983) Suppl. p. 183.
3) H. Nishimori and S. J. Miyake: Prog. Theor. Phys. **73** (1985) 18.
4) S. J. Miyake: Prog. Theor. Phys. **74** (1985) No. 3.
5) S. Miyashita: J. Phys. Soc. Jpn. **53** (1984) 44.
6) P. W. Anderson: Mater. Res. Bull. **8** (1973) 153.
7) R. Jullien, K. A. Penson, P. Pfeuty and K. Uzelac: Phys. Rev. Lett. **44** (1980) 1551.
8) L. G. Marland and D. D. Betts: Phys. Rev. Lett. **43** (1979) 1618.
9) J. C. Bonner and H. W. J. Blöte: Phys. Rev. B**25** (1982) 6959.
10) P. W. Anderson: Phys. Rev. **83** (1951) 1260.
11) M. Suzuki: J. Stat. Phys. **42** (1986) March.
12) P. Fazekas and P. W. Anderson: Philos. Mag. **30** (1974) 423.
13) M. Suzuki and S. Mjiyashita: Can. J. Phys. **56** (1978) 902.
14) J. Oitmaa and D. D. Betts: Can. J. Phys. **56** (1978) 897.
15) K. Hirakawa, H. Kadowaki and K. Ubukoshi: J. Phys. Soc. Jpn. **54** (1985) 3526.
16) E. Lieb and D. Mattis: J. Math. Phys. **3** (1962) 749.
17) D. Mattis: Phys. Rev. Lett. **42** (1979) 1503.
18) H. Nishimori: J. Stat. Phys. **26** (1981) 839.