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Quantum Monte Carlo simulation of diluted Heisenberg antiferromagnets ($S = \frac{1}{2}$) on the square lattice

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Abstract. The effects of inhomogeneity in a system with strong quantum fluctuations are investigated by a quantum Monte Carlo simulation (QMC). The properties of an antiferromagnetic Heisenberg model with $S = \frac{1}{2}$ on the square lattice diluted by non-magnetic impurities are investigated. The spatial distribution of the spin correlations near impurities are found to be largely enhanced. On the other hand, from the size dependence of the square of the staggered magnetization per spin, the correlations between largely separated spins are found to be smaller than in the non-diluted case. It has been found that the existence of non-magnetic sites causes the ergodicity problem in QMC. In order to avoid this difficulty we introduce a new type of global flip. With this type of flip QMC gives good agreement with results obtained by exact diagonalization methods even for systems with highly concentrated impurities. The methodological points are also discussed in detail.

1. Introduction

The nature of the order of the Heisenberg antiferromagnet ($S = \frac{1}{2}$) on the square lattice has been studied extensively [1–3]

$$\mathcal{H} = 2J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1.1}$$

where $\boldsymbol{S} = \frac{1}{2}\boldsymbol{\sigma}$; $\boldsymbol{\sigma}$ are the Pauli matrices. It is generally believed that this model has long-range staggered order in the ground state. But the value of the order parameter is much reduced by the quantum fluctuations due to the non-commutativity between the order parameter

$$N_z \equiv \sum_{i \in A} \sigma_i^z - \sum_{i \in B} \sigma_i^z \tag{1.2}$$

and the Hamiltonian. In such systems, the effect of inhomogeneity on the correlation function has been one of recent interest [4–8]. We have studied the effect of non-magnetic impurities by using quantum Monte Carlo simulations (QMC) [7]. We are interested in phenomena due to the combination of the effect of the impurities and the quantum effects. It has been found that correlation functions are not necessarily reduced but sometimes enhanced by impurities [4–8]. On the other hand, we may also

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expect the quantum fluctuations to cause a reduction in the long-range correlations and thus a change in the percolation threshold of impurity concentration for long-range order in the ground state. In order to study these effects, we investigate the microscopic distribution of the spin correlation functions. For the local effects of the impurities, we studied $L \times L = 4 \times 4$ lattices by an exact diagonalization method. We find the enhancements near impurities very clearly. In the present model the nearest-neighbour (NN) correlation is proportional to the energy and the enhancement can be understood generally [9] from the variational principle that

$$|\langle G_p | \mathcal{H}_{imp} | G_p \rangle| \leqslant |\langle G_i | \mathcal{H}_{imp} | G_i \rangle| \tag{1.3}$$

where $| G_p \rangle$ is the ground-state wavefunction for the pure system and $| G_i \rangle$ the ground-state wavefunction of $\mathcal{H}_{imp}$. Because the Hamiltonian is the summation of NN correlations, the average value of NN correlations should be enhanced. Some of the further correlations are, however, also found to be enhanced. Then, it is another interesting problem to investigate how far the correlations are enhanced.

For larger lattices, we have performed QMC simulations based on the Suzuki–Trotter decomposition. We find similar enhancements near impurities. But the enhancement is found to be localized around the impurity and the correlation between largely separated spins are found to be reduced. In order to study the nature of long-distance correlations, we investigate the square of the staggered magnetization, $\langle N_z^2 \rangle / N_s^2$ where N_s is the number of spins. In our previous report [7], large enhancements of the square staggered magnetization were reported. But it has been found that the existence of non-magnetic sites causes difficulty for the ergodicity in QMC. In particular at low temperatures, the antiferromagnetic order has a tendency to be enhanced too much by a type of freezing effect due to a lack of ergodicity in QMC. We have concluded that the previous data at low temperatures were frozen in some metastable states. In order to avoid this difficulty we introduced a new type of global flip. The methodological points are given in the appendix. QMC with the new algorithm gives good agreement with results by an exact diagonalization method even for systems with highly concentrated impurities (non-magnetic sites). $\langle N_z^2 \rangle / N_s^2$ is now found to be reduced for larger lattices. From the size dependence of this quantity, we conclude that the correlation between large separated spins are smaller than in the pure case.

In section 2, the model and the methods are given and in section 3.1, the local enhancement of correlations and the spatial distribution of correlations obtained by diagonalization method and QMC are shown. In section 3.2 the size dependence of the square of the staggered magnetization is investigated. Discussion and a summary are given in section 4.

2. Model and methods

The Hamiltonian of the model studied here is

$$\mathcal{H} = 2J \sum_{\langle i,j \rangle} \epsilon_i \epsilon_j \boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{2.1}$$

where $\epsilon_i = 1$ or 0. We choose the positions where $\epsilon = 0$ randomly. These positions will be called 'impurities'. The concentration of impurities is defined by δ,

$$\delta = 1 - \sum_i \epsilon_i / L^2. \tag{2.2}$$

We used the QMC method based on the Suzuki–Trotter decomposition [10] using the chequer-board decomposition [11]:

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 \tag{2.3}$$

where

$$\mathcal{H}_1 = \frac{J}{2}\sum_{i,j} \sigma_{2i-1,2j-1}\sigma_{2i,2j-1} + \sigma_{2i,2j-1}\sigma_{2i,2j} + \sigma_{2i,2j}\sigma_{2i-1,2j} + \sigma_{2i-1,2j}\sigma_{2i-1,2j-1}$$

$$\mathcal{H}_2 = \frac{J}{2}\sum_{i,j} \sigma_{2i,2j}\sigma_{2i+1,2j} + \sigma_{2i+1,2j}\sigma_{2i+1,2j+1} + \sigma_{2i+1,2j+1}\sigma_{2i,2j+1} + \sigma_{2i,2j+1}\sigma_{2i,2j}$$

for all lattice sites with spin. Here we have three states on a site i,

$$\sigma_i = 0 \text{ or } +1 \text{ or } -1 \tag{2.4}$$

corresponding to impurity, spin-up and spin-down, respectively. In the present simulation we prepared a general algorithm for three states. Partly because this provides uniform operation for the whole lattice, which has an advantage in the vectorization of the program and partly because this can be extended to more general cases where the impurities can move (hopping of hole) or the case with $S = 1$. Thus, the unit of the local Boltzmann factor

$$\rho(\sigma_1, \sigma_2, \sigma_3, \sigma_4, \sigma_1', \sigma_2', \sigma_3', \sigma_4') = \langle \sigma_1, \sigma_2, \sigma_3, \sigma_4 | \exp(-\beta/m\mathcal{H}_{(1)}) | \sigma_1', \sigma_2', \sigma_3', \sigma_4' \rangle \tag{2.5}$$

is $3^4 \times 3^4$, where m is the Trotter number. In the present case the model has the conservation law,

$$\sigma_1 + \sigma_2 + \sigma_3 + \sigma_4 = \sigma_1' + \sigma_2' + \sigma_3' + \sigma_4' \tag{2.6}$$

and the size of the table of ρ is 3^7.

Detailed points of the simulation are given in the appendix but the most important new point is how to recover the ergodicity of the Monte Carlo time evolution. A 'Néel chain', which is a sequence of up–down spins in real space in a Trotter layer, hardly flips simultaneously near impurities, if we do not include the global flip. And configurations are easily frozen at low temperatures. This also caused large enhancements of the staggered correlations [7]. In order to avoid this effect we introduced a type of global flip around the impurities (loop flip) together with a non-straight Marcu flip [12].

3. Distribution of the staggered correlation

3.1. Local enhancements

In order to see the nature of the enhancement of the staggered magnetization around the impurities, we first investigated the ground state and several excited states of 4×4 systems by a diagonalization method. Low-temperature data are obtained from low excitations using the canonical weight. The distribution of NN correlations are given for several samples in figure 1 (for comparison we used $T = 0.2J$). As we mentioned in section 1, it is natural that the NN correlations are enhanced but we find that some

further-neighbour correlations are also enhanced and these are listed in table 1. In particular figure 1(*a*) was investigated in [6]. Although in the spin wave calculations the x- and z-components are not the same, a similiar tendency is found. For this lattice $T = 0$ and $T = 0.2$ are essentially the same. Next we study larger lattices in order to investigate how well-separated correlation functions are enhanced. The 4×4 lattice is too small and even the furthest correlation function is enhanced in some configuration. Then we investigate 8×8 and 12×12 lattices by QMC. The distribution of NN correlations at $T = 0.2J$ for one configuration is given in figure 2, where we find enhancements which are qualitatively similar to those in figure 1. But we would like to point out that some correlations are reduced even in neighbours of impurities. This non-uniform enhancement will be discussed in section 4. The average of the NN correlations gives the energy. The concentration dependence of the energy $E(\delta)$ is given in figure 3, where $E(\delta)$ is normalized by the number of sites, N, and also the number of spins, N_s.

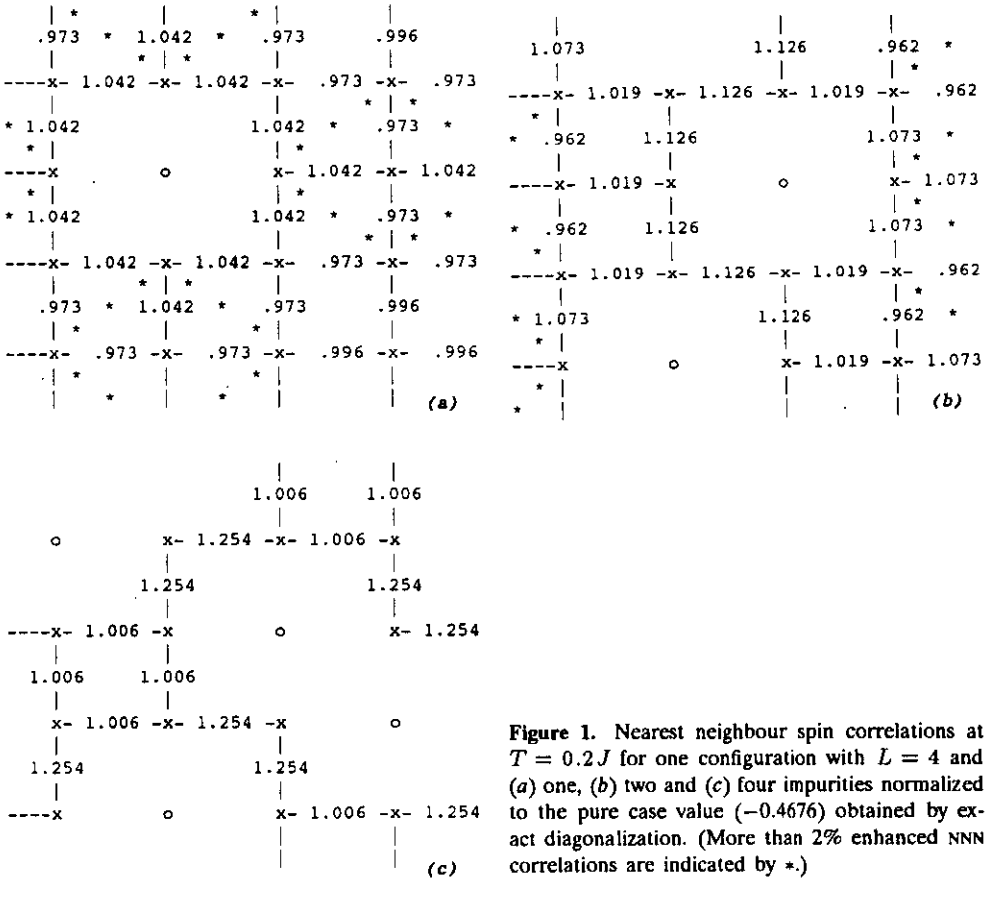

Figure 1. Nearest neighbour spin correlations at $T = 0.2J$ for one configuration with $L = 4$ and (*a*) one, (*b*) two and (*c*) four impurities normalized to the pure case value (−0.4676) obtained by exact diagonalization. (More than 2% enhanced NNN correlations are indicated by *.)

Further-neighbour correlations are listed in table 2, where correlations enhanced by more than 4% are listed. We find that the number of enhanced correlations decreases as the distance increases. The range of enhancement is rather short. In order to see the distance dependence of the correlations, we study the square of the staggered magnetization in the next section.

Table 1. Further-neighbour correlations c_r enhanced by more than 2% normalized to the pure case value c_p for the same configurations as in figures 1(*a*), (*b*), (*c*).

Configuration of figure 1(*a*)			Configuration of figure 1(*b*)			Configuration of figure 1(*c*)		
Site$_1$–site$_2$	c_r	c_p	Site$_1$–site$_2$	c_r	c_p	Site$_1$–site$_2$	c_r	c_p
(1, 1)–(2, 2)	1.029	0.285	(4, 4)–(1, 1)	1.020	0.285	(1, 1)–(3, 2)	1.079	0.269
(3, 1)–(2, 2)	1.029	0.285	(1, 1)–(4, 2)	1.020	0.285	(4, 3)–(2, 4)	1.079	0.269
(4, 2)–(1, 3)	1.029	0.285	(1, 2)–(4, 3)	1.020	0.285	(4, 3)–(1, 1)	1.079	0.269
(4, 2)–(3, 3)	1.029	0.285	(4, 3)–(1, 4)	1.020	0.285	(2, 4)–(3, 2)	1.079	0.269
(1, 3)–(4, 4)	1.029	0.285	(1, 1)–(1, 3)	1.020	0.285			
(3, 3)–(4, 4)	1.029	0.285	(4, 1)–(4, 3)	1.020	0.285			
(2, 4)–(3, 1)	1.029	0.285	(4, 3)–(1, 1)	1.160	0.269			
(2, 4)–(1, 1)	1.029	0.285	(1, 1)–(2, 3)	1.035	0.269			
(1, 1)–(1, 3)	1.029	0.285	(3, 4)–(1, 1)	1.035	0.269			
(3, 1)–(3, 3)	1.029	0.285	(1, 1)–(3, 2)	1.035	0.269			
(2, 2)–(4, 2)	1.029	0.285	(3, 1)–(4, 3)	1.035	0.269			
(2, 4)–(4, 4)	1.029	0.285	(2, 2)–(4, 3)	1.035	0.269			
(4, 1)–(2, 2)	1.038	0.269	(4, 3)–(2, 4)	1.035	0.269			
(4, 1)–(1, 3)	1.038	0.269						
(3, 3)–(4, 1)	1.038	0.269						
(2, 4)–(4, 1)	1.038	0.269						

3.2. Size dependence of the square of the staggered magnetization

The square of N_z, $\langle N_z^2 \rangle$ where $\langle \cdots \rangle$ denotes the expectation value in the ground state, is a summation of all staggered correlations in the sense

$$\langle N_z^2 \rangle = \sum_{i,j} \epsilon_i \epsilon_j \langle \sigma_i^z \sigma_j^z \rangle. \tag{4.1}$$

Thus, if we study the size dependence of this quantity per spin,

$$\mathcal{A} \equiv \langle N_z^2 / N_s^2 \rangle \tag{4.2}$$

the distance dependence of the correlation function can be known. In classical systems, $\mathcal{A}$ does not depend on N. For quantum systems, $\mathcal{A}$ may be larger than the pure case in small lattices such as 4×4 because the short-range correlations dominate. On the other hand, for large lattices the long-range correlations dominate. The size dependence of $\mathcal{A}$ is plotted in figure 4, where we find that $\mathcal{A}$ decreases as the size increases. Thus we conclude that the correlation functions of largely separated spins are reduced by the existence of impurities. In figure 5 the correlation function for $N = 12$ with $\delta = 0.125$ averaged over five samples are compared with the pure case. Here we again see the reduction. The amount of the reduction has not yet been investigated systematically but it might give a change in the percolation threshold of the impurity concentration for the long-range order in the ground state. This should be due to the quantum fluctuations and will be studied in future [15].

4. Summary and discussion

We have studied the spin correlation function $\langle S_i S_j \rangle$. Due to the quantum effects the correlation functions are modified even in the ground state. We have shown

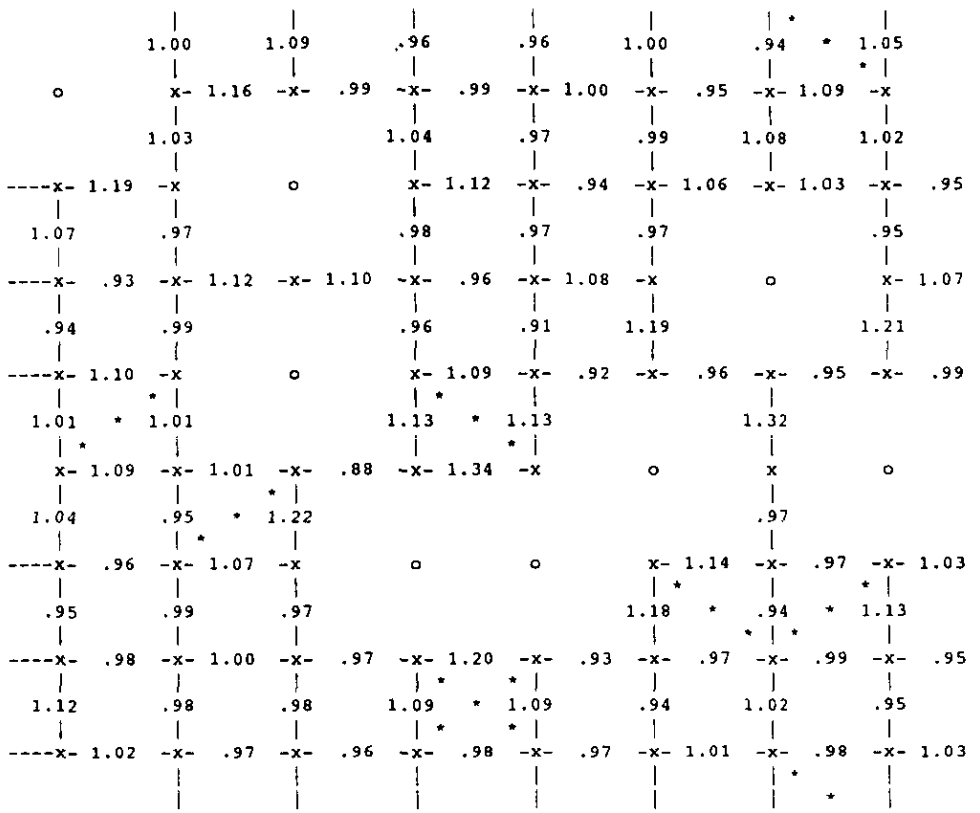

Figure 2. Nearest neighbour spin correlations at $T = 0.2J$ for one configuration with $L = 8$ and 8 impurities normalized to the pure case value (-0.462) obtained by QMC simulation. (More than 4% enhanced NNN correlations are indicated by *.)

the distribution of correlation functions obtained by QMC for several samples. We found a similiar tendency, which has been previously pointed out [6]; namely the spin correlations are enhanced near the impurity sites (non-magnetic sites). But we also found that the range of the enhancement is somewhat short. On the other hand, the correlation functions are reduced at large distance. As we pointed out in section 2, the enhancement near impurities is not necessarily uniform. There are many correlations which are reduced even in the neighbours of impurities, although the others are very strongly enhanced. The mechanism of this non-uniform enhancement should have a purely quantum origin and it remained as 'quantum interference' in spin system.

Here we would like to point out the following tendency of the enhancement. The correlations at corner positions, where a spin has only two neighbours, are strongly enhanced (e.g. $(x = 5, y = 4)$ or $(x = 6, y = 3)$ in figure 2). And the correlations of other spins which have only two neighbours also have the tendency to be enhanced. At least one of the correlations around spins with three neighbours is enhanced. Thus we see a strong relation between the number of neighbours and enhancement. On the other hand we also find an alternate enhancement, namely, if a bond is enhanced, the next ones are reduced. The distribution of enhancement seems to be determined by these tendencies. Most cases in figure 2 can be understood in this way. A more precise mechanism for this competition between order and fluctuation will be studied

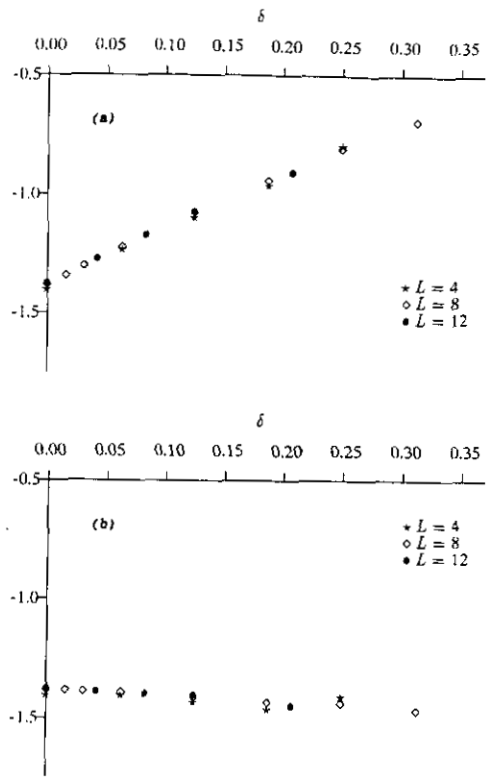

Figure 3. Energy depending on impurity concentration δ normalized (*a*) to the number of lattice sites N and (*b*) to the number of spins N_s ($T = 0.2\,J$).

Table 2. Further-neighbour correlations c_r enhanced by more than 4% normalized to the pure case value c_p for the same configuration as in figure 2.

Site$_1$–site$_2$	c_r	c_p
(5, 4)–(4, 5)	1.114	0.288
(1, 4)–(2, 5)	1.067	0.288
(4, 1)–(5, 2)	1.056	0.288
(2, 3)–(3, 4)	1.050	0.288
(7, 2)–(6, 3)	1.050	0.288
(5, 1)–(4, 2)	1.046	0.288
(8, 8)–(7, 1)	1.045	0.288
(7, 2)–(8, 3)	1.041	0.288
(7, 1)–(6, 3)	1.090	0.242
(1, 1)–(8, 3)	1.085	0.242
(8, 2)–(6, 3)	1.056	0.242
(2, 5)–(8, 6)	1.050	0.242
(2, 5)–(1, 7)	1.042	0.242

in the future [13].

We may expect that the effect of the quantum interference causes a rapid reduction in the spin correlation function at a large distance in random systems, which is an analogy of the Anderson localization in spin systems. This reduction mechanism

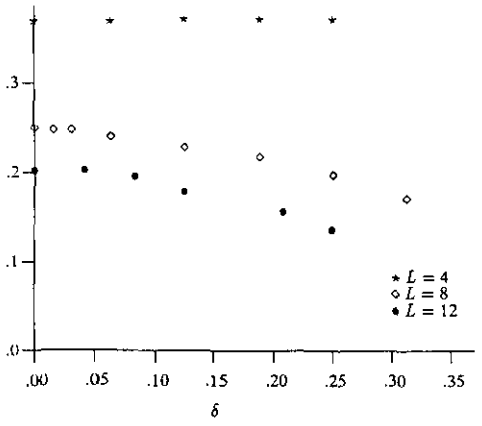

Figure 4. Sublattice magnetization depending on impurity concentration δ normalized to the number of spins N_s ($T = 0.2J$).

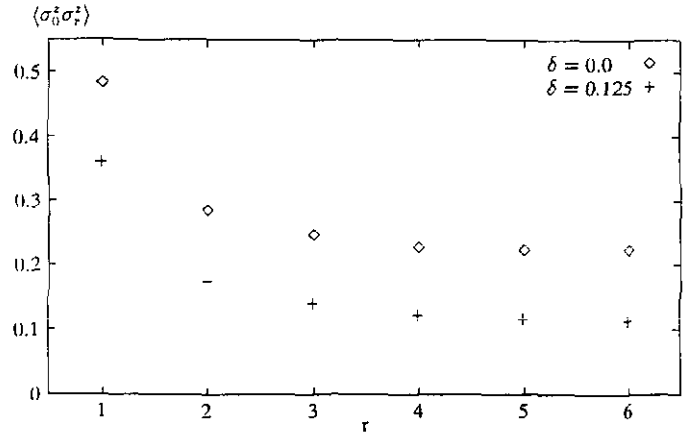

Figure 5. The correlation $\langle \sigma_0^z \sigma_i^z \rangle$ as a function of the distance r for $N = 12$ with $\delta = 0.0$ and $\delta = 0.125$ at $T = 0.2$.

might give a change in the percolation threshold of impurity concentration for the long-range order in the ground state. The point is being investigated and will be reported elsewhere [13]. Similar effects could be seen on the triangular lattice with non-magnetic impurities, although the frustration effect may have an important role for the local enhancements there [14].

In this paper we concentrate on the microscopic properties in distribution of the enhancements of correlation due to the impurities, which is one of the most interesting features of the imhomogeneous quantum system. For the macroscopic quantities such as the energy, magnetization, etc, we have to perform a sample average over randomness of impurity position. In this paper we studied $\langle N_z^2/N_s^2 \rangle$ preliminarily in order to see the distance dependence of the correlation. So far we have found small scatterings of data over a few samples. But the properly averaged data will be reported elsewhere. The problem of the percolation threshold of the long-range order in the ground state will also investigated quantatively there [15].

We would also like to point out the following. So far we have studied $\langle S_i S_j \rangle$,

Table 3. Flip acceptance ratios for one plane and the simulation of different Trotter numbers m_0 for the flip types shown in figure 7 and the simulation of figure 9.

	Flip types					
m_0	1	2	3	4	5	6
16	0.0346	0.0344	0.0345	0.0345	0.0022	0.0021
24	0.0155	0.0156	0.0158	0.0157	0.0005	0.0005
32	0.0083	0.0083	0.0082	0.0083	0.0002	0.0002
48	0.0031	0.0031	0.0031	0.0031	0.0000	0.0000

which is the direct correlation. If we see the frequency of types of flip, we find that loop flips around impurities frequently occur. This suggests that the configurations around the impurities very often change keeping the relative antiferromagnetic order. In table 3, the acceptance ratios for flips are given. Thus it will also be interesting to study the canonical correlation

$$(\sigma_i \sigma_j) = \frac{1}{\beta} \int_0^\beta \sigma_i e^{-\lambda \mathcal{H}} \sigma_j e^{(\lambda-\beta)\mathcal{H}} d\lambda \tag{5.1}$$

where β is the inverse temperature, at low temperatures. We also presented the technical details of our QMC algorithm, which are important to obtain correct data efficiently in the diluted systems and to overcome the ergodicity problem of the simulation.

Acknowledgments

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Appendix

In this appendix we describe the details of the QMC simulation. As has been mentioned in section 2, we used the chequer-board type of the Suzuki–Trotter decomposition. Thus the local Boltzmann factor contains four spins and then is a $3^4 \times 3^4$ matrix. Because an impurity does not move, the ergodicity of the simulation is much more difficult to satisfy than in the pure case. If we consider a worldline with a non-zero winding number between two impurities (figure 6), we find that the line cannot escape from the two impurities. Thus in order to satisfy the ergodicity of winding numbers we have to prepare types of global flip for all possible paths between the impurities. We prepare these in the following way. First we prepare 'a loop flip' around each cluster of impurities. Next, we prepare several horizontal global flips (Marcu flips), some of which are not straight. It is easy to see that a combination of these will give all possible paths. In figure 7 the different flip types which are used in the simulation are shown.

We have compared the data with and without the new flips. First we have compared the distribution of NN correlations of figure 1(*b*) (exact diagonalization)

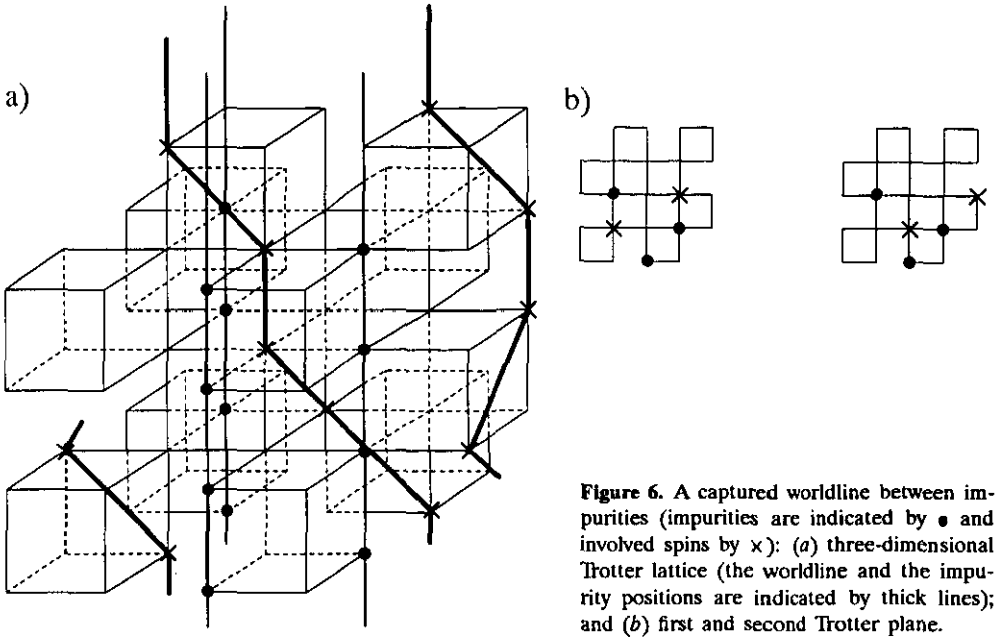

Figure 6. A captured worldline between impurities (impurities are indicated by • and involved spins by ×): (*a*) three-dimensional Trotter lattice (the worldline and the impurity positions are indicated by thick lines); and (*b*) first and second Trotter plane.

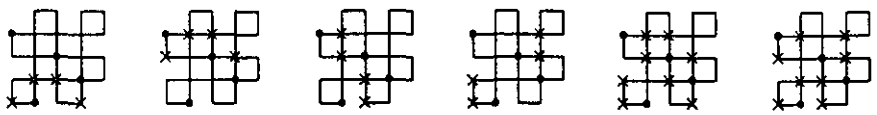

Figure 7. Used combinations of spins for different flip types for the configurations of figure 9 with $L = 4$ and four impurities. No standard straight global flip is possible. (Impurity positions are indicated by • and spins used for the flip by ×.)

with the data by QMC without the new flips but with straight Marcu flips and with those by QMC without the new flip types. We found that all of them agree within the error bars. This is so because the straight Marcu flips are sufficient to recover the ergodicity for this case. In the simulation which was previously reported, we used QMC with only the straight Marcu flips, which seemed to be sufficient. However we found that this was not enough for more complicated cases. Then, we look at the data for a lattice with four impurities. Here the straightest Marcu flip no longer works. We find a clear difference between figures 8 and 9 (see also figure 1(*c*)), and the data obtained by QMC without the new flip types (figure 8) show too much enhancement of the correlations. This is the reason why we reported too much enhancement in the previous report [7]. Now we believe that we have overcome this difficulty. In order to confirm the validity of the simulation, we compared two equivalent samples in different installations. Namely we shifted the origin site one lattice spacing to the right. This is simply a translational shift and does not change anything physically because of the periodic boundary condition. But it gives large changes in the three-dimensional Trotter lattice. We performed both simulations and we found no difference between them. Thus, we conclude that our new method is valid for imhomogeneous quantum systems such as the present model.

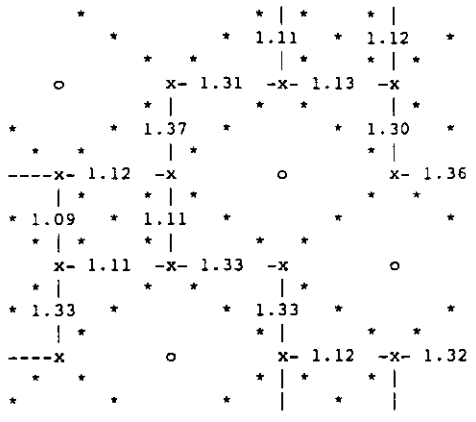

Figure 8. Nearest neighbour spin correlations at $T = 0.2J$ for the same configuration as in figure 1(*c*) normalized to the pure case value (−0.4676) obtained by QMC (more than 2% enhanced nnn correlations are indicated by *.)

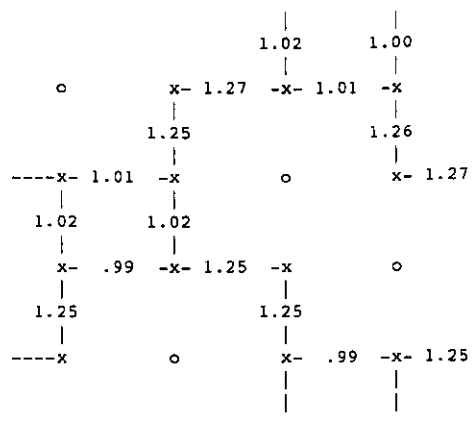

Figure 9. Nearest neighbour spin correlations at $T = 0.2J$ for the same configuration as in figure 1(*c*) normalized to the pure case value (−0.4676) obtained by QMC with additional new types of flips (figure 7).

References

[1] Reger J D and Young A P 1988 *Phys. Rev.* B **37** 5978
[2] Miyashita S 1988 *J. Phys. Soc. Japan* **57** 1934
[3] Manousakis E 1991 *Rev. Mod. Phys.* **63** 1
[4] Talbot E F 1986 *Phys. Rev.* B **33** 4906
[5] Camley R E, von der Linden W and Zevin V 1989 *Phys. Rev.* B **40** 119
[6] Nagaosa N, Hatsugai Y and Imada M 1989 *J. Phys. Soc. Japan* **58** 978
Bulut N, Hone D, Scalapino D J and Loh E Y 1989 *Phys. Rev. Lett.* **62** 2192
Lee K J B and Schlottmann P 1990 *Phys. Rev.* B **42** 4426
[7] Behre J, Miyashita S and Mikeska H J 1990 *J. Phys. A: Math. Gen.* **23** L1175
[8] Betts D D and Oitmaa J 1991 private communication
[9] Betts D D and Miyashita S 1990 *Can. J. Phys.* **68** 1410
[10] Suzuki M 1987 *Quantum Monte Carlo Methods in Equilibrium and Nonequilibrium Systems* ed M Suzuki (Berlin: Springer) p 2
[11] Loh E Y, Scalapino D J and Grant P M 1985 *Phys. Rev.* B **31** 4712
[12] Marcu M 1987 *Quantum Monte Carlo Methods in Equilibrium and Nonequilibrium Systems* ed M Suzuki (Berlin: Springer) p 64
[13] Miyashita S and Behre J 1992 *Computational Approaches in Condensed Matter Physics* ed S Miyashita, M Imada and H Takayama (Berlin: Springer) in press
[14] Deutscher R 1992 private communication
[15] Behre J and Miyashita S 1992 in preparation