1 Quantum Monte Carlo Simulations of the Impurity-Induced Phase Transitions in Low-Dimensional Magnets

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We study the impurity-induced phase transitions in a quasi-one-dimensional Heisenberg antiferromagnet doped with magnetic spin-1/2 impurities and non-magnetic ones. The impurity-induced transition temperature determined by the quantum Monte Carlo method with the continuous-time loop algorithm is monotonically increasing as a function of the magnitude of the impurity spin. To these results, we give discussions based on the valence-bond solid-like picture for the pure system and the inspection of the local magnetic structure around the impurities.

1.1 Motivation

Impurities in the low-dimensional quantum magnets drastically change the magnetic properties of the host materials. For the low-dimensional magnets such as CuGeO\textsubscript{3} \cite{1} and PbNi\textsubscript{2}V\textsubscript{2}O\textsubscript{8} \cite{2}, pure systems have disordered ground states, while non-magnetic-impurity-doped ground states are antiferromagnetically ordered. These impurity-induced ordered states have been given a qualitative understanding based on the valence-bond solid (VBS) picture \cite{3} for low-dimensional magnets. According to the VBS picture, a spin with the magnitude $S$ on each site is treated as ferromagnetically coupled $2S$ subspins with the magnitude 1/2, and the disordered ground state of a low-dimensional antiferromagnet can be seen as a state made of closely packed spin-1/2 singlet pairs, which are called valence bonds. When a non-magnetic impurity is doped, a valence bond is broken after one of the spin in a pair is lost and the counterpant subspin is liberated. Thus non-magnetic impurities induce nearly-free local magnetic moments around their doped sites and there is long-range correlation between them that keeps the bulk antiferromagnetic long-range order \cite{4}.

Recently, in the experiments using the spin-1 gapped magnet PbNi\textsubscript{2}V\textsubscript{2}O\textsubscript{8}, the effects of several species of magnetic impurities were systematically investigated and it was found that the impurity-induced transition temperatures show non-monotonic dependence on the magnitude of the impurity spin \cite{5}. In particular, compared with the transition temperatures induced by non-magnetic impurities, those done by Cu\textsuperscript{2+} are found to be much lower. These
results can be thought to be reasonable if we inspect the magnitude of the local magnetic moments that are expected to appear around impurities by the simplest picture analogous to the VBS one. There are two spin-1/2 effective magnetic moments around one non-magnetic impurity, and these are coupled by an effective ferromagnetic coupling mediated by the interchain couplings, making one spin-1 local magnetic moment. For a magnetic spin-1/2 impurity, there is one spin-1/2 local magnetic moment per one impurity. Thus the magnitude of the local magnetic moments is larger for the non-magnetic impurities than for the magnetic spin-1/2 ones. We can at least hope to understand the experimental result that the spin-1/2 impurities induce much lower transition temperature than the non-magnetic ones do. Motivated by this observation, we do quantum Monte Carlo simulations for the quasi-one-dimensional Heisenberg antiferromagnets with site impurities and see if our picture for the impurity-induced ordered state is valid or not.

1.2 Model and Method

We introduce our model following the observation described in the previous section. The pure system is weakly coupled spin-1 antiferromagnetic Heisenberg chains aligned in parallel on a simple cubic lattice. The Hamiltonian is written as follows.

$$
\mathcal{H} = J \sum_{x,y,z} S_{x,y,z} \cdot S_{x+1,y,z} + J' \left( \sum_{x,y,z} S_{x,y,z} \cdot S_{x,y+1,z} + \sum_{x,y,z} S_{x,y,z} \cdot S_{x,y,z+1} \right)
$$

(1.1)

Here the spin operator $S_{x,y,z}$ has the magnitude $S \equiv |S| = 1$ and $x, y, z$ denote the points on a simple cubic lattice. We consider only the nearest neighbor antiferromagnetic couplings. The intrachain (interchain) coupling is denoted by $J(J') > 0$. We set the $x$ axis parallel to the chains. The strength of the interchain coupling, $J'$, is set to be $0.01J$. This value is small enough to allow the Haldane gap [6] to be finite even in a three-dimensional space. The critical point between the gapped phase and the antiferromagnetically ordered one, $J'_c$, is estimated to be $J'_c = 0.013J$ [7, 8].

In the impurity-doped system, the $S = 1$ host spins are randomly replaced with the $S \neq 1$ impurity spins with the couplings between the spins left unchanged. The aim of using this simple model is to let the magnitude of local magnetic moments play a dominant role in determining the impurity-induced transition temperatures by letting all of the species of impurities share the equal strength of the effective couplings between the impurity neighborhoods when the concentration of the impurities is fixed. By this model we try to simulate the impurity-induced transition in which the non-magnetic impurities yield higher transition temperature than the magnetic spin-1/2 ones do.

The impurity-induced transition temperatures are found as follows. The physical observables are calculated by the quantum Monte Carlo with the
continuous-time loop algorithm [9, 10]. The correlation length is calculated using the second moment of the two-point correlation function [10, 11]. The ratio of the correlation length to the linear size of the system is plotted with respect to the temperature for several system sizes and the crossing point (if any is found) gives the position of critical point. By this simple analysis, enough accuracy in the data for the qualitative discussion in the present study is obtained. The concentration of impurities is fixed to be 10% and the impurity-induced transition temperatures (if any is found) are compared with each other between the species of impurities.

1.3 Results and Discussions

By the procedure described in the previous section, we find that all species of impurities induce phase transitions in our model. Specifically, it is found that non-magnetic impurities induce the Néel temperature at $T_N = 0.03 \pm 0.01$ and magnetic spin-1/2 ones do that at $T_N = 0.05 \pm 0.01$. As for the other magnetic-impurity-induced transition temperatures, we find that they are monotonically increasing as a function of the magnitude of the spin of impurities [12]. We must note that the transition temperature induced by the magnetic spin-1/2 impurities is higher than that done by the non-magnetic ones, which is in contrast with our expectation before simulations.

To check our presumption that the magnitude of the local magnetic moments, which should be relevant in the value of the impurity-induced transition temperature, must be larger for non-magnetic impurities than for magnetic spin-1/2 ones, we are going to take a look directly on the local magnetic moments around an impurity. We calculate the local field susceptibilities for each site $i$, which we denote by $\chi_{\text{local},i}$, on a single spin-1 chain with one impurity doped in the center site of the chain. The local field susceptibility is defined by the following formula.

$$\chi_{\text{local},i} = \frac{\partial \langle S_i^z \rangle}{\partial h_i} \bigg|_{h_i = 0} = \int_0^\beta d\tau \langle S_i^z(\tau)S_i^z(0) \rangle$$

Here $h_i$ is the local magnetic field applied on the site $i$, $\tau$ is the imaginary time introduced in the path-integral formalism that is employed in the loop algorithm, and $\beta$ is the inverse temperature. The real-space distribution of the local field susceptibility is shown in Fig. 1.1. Here the length of the chain is 64 and the temperature is 0.01$J$ at which the pure system is well near the ground state. We see that the total magnitude of the local magnetic moments is larger in the neighborhood of a non-magnetic impurity than in that of a magnetic spin-1/2 one. This result is consistent with our first expectation.

As the transition temperature is expected to be determined by both of the magnitude of the magnetic moments that contribute to the long-range order and the strength of the interaction between them, we are led to consider the interaction between the local magnetic moments more closely. First
we consider the neighborhoods of non-magnetic impurities. The two local magnetic moments with the effective magnitude $1/2$ around a non-magnetic impurity are ferromagnetically coupled mediated by the very weak interchain couplings. The strength of this coupling is estimated to be of the order $J'^2$. Near the transition temperature that is in energy scale comparable to the strength of the interchain coupling $J'$, the coupling of the order $J'^2$ between the two moments hardly contribute to the global antiferromagnetic order of the present interest. On the other hand, around a magnetic spin-$1/2$ impurity, two effective spin-$1/2$ moments at the edge of the pure chain and the impurity spin are directly coupled yielding an effective spin of the magnitude $1/2$. See Fig. 1.2. When the ground state of the three spins with the magnitude $1/2$ which are separated from the rest is inspected, they exhibit magnetic moments (whose magnitudes are less than $1/2$) antiferromagnetically aligned. This strongly suggests that the three spins contribute to the global antiferromagnetic order even at temperatures of the order $J'$, i.e., near the transition temperature. Thus it is reasonable to encounter the present simulational results that the impurity-induced temperatures are higher for the magnetic impurities than for the non-magnetic ones. Our first expectation may hold good in the ordering at temperatures below the order $J'^2$.

In real experiments, various kinds of interaction around impurities must be playing key roles, which may require the detailed examination specific to each kind of impurities [5].

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Fig. 1.2. Schematic figure for the local spin-spin couplings between local magnetic moments around (a) a non-magnetic impurity and (b) a spin-1/2 impurity in the gapped phase of the quasi-one-dimensional spin-1 Heisenberg antiferromagnet. The bold solid line represents a valence bond and the dotted line do the intrachain coupling. The arrows denote local spin-1/2 effective magnetic moments.

library “LOOPER version 2” developed by Dr. S. Todo and Dr. K. Kato, and “PARAPACK version 2” developed by Dr. S. Todo.

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