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Large moment formation and thermodynamic properties of disordered spin ladders with site dilution

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Low temperature properties of antiferromagnetic two-leg spin-1/2 ladders with bond randomness and site dilution (or doping with nonmagnetic impurities) are studied using the real-space renormalization-group technique. We find that for nonzero dopant concentrations the systems are driven into a phase dominated by large effective spins, i.e., the large spin phase. The susceptibility follows a universal Curie-like $1/T$ behavior at low temperature, regardless of the dopant concentration (as long as it is nonzero) and the strength of bond randomness. A very similar behavior has been found in ladders that are doped with magnetic impurities that carry spin-1.

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I. INTRODUCTION

Quantum effects in one-dimensional spin systems have attracted prolonged interest from both theoretical and experimental physicists. These include quasi-long-range order, topological order, and the fluctuation induced excitation gap (e.g., the Haldane gap) that are purely quantum mechanical effects enhanced by the low dimensionality of the systems. Among these quantum phenomena, the effects of disorder have been studied by many groups. It was found that disorder can qualitatively change the low temperature physics and produce rich disorder-dominated phases in these systems. One class of such systems which have received considerable attention is that of random antiferromagnetic spin chains. Most of the theoretical studies of random spin chains are based on the real space renormalization group (RSRG) method developed by Ma, Dasgupta, and Hu[1] in this context, and Bhatt and Lee[2] in the study of doped semiconductors. This RSRG method was extended further by Fisher,[3] and allows one to obtain results which are essentially exact for the random spin-1/2 chain. The application of this method to the other random spin chain models by a number of authors[4–11] has given us a better understanding of the behavior of these systems at low temperature.

Another example of a one-dimensional spin system that is of considerable recent interest is the two-leg antiferromagnetic spin-1/2 ladder.[12] It is known to have an excitation gap similar to the Haldane gap of integer AF spin chains, and a short-range spin-spin correlation. Compared to the spin chains, only relatively few theoretical studies have been devoted to the study of disorder effects in spin ladders. Several authors have investigated the effects of bond randomness.[13–15] It was found that the ladder is remarkably stable against *weak* bond randomness.[13] Stronger randomness introduces a large density of low-energy excitations into the system,[14,15] which can lead to *divergent* spin susceptibility in the limit $T \to 0$.[15] However, the spin-spin correlation remains short ranged,[15] contrary to what occurs in strongly disordered antiferromagnetic spin chains.[3,8]

In real systems, bond randomness is typically induced by impurities *away* from the ladder, which distort the lattice structure (and hence coupling constants) without affecting the spins that form the ladder. Experimentally, another way to introduce and control disorder in the system is to introduce dopants that go directly into the ladder, so that the ions that carry the half-spin (typically the Cu ion) are randomly replaced by nonmagnetic ions (like Zn), or ions with other spin sizes (like Ni which carries spin-1). Such disorder not only induces lattice distortion, but also changes the lattice structure of the spin ladder through site-dilution etc, and thus has more dramatic effects. A number of experimental[16,17] and theoretical[18–28] works have been devoted to study the doped two-leg spin-1/2 ladder, for example $Sr(Cu_{1-x}Zn_x)_2O_3$, where some Cu ions are replaced by nonmagnetic Zn ions. It was found experimentally that even a small amount of nonmagnetic doping is enough to change the low-temperature behavior of the systems drastically, and gives rise to divergent susceptibility at low temperature. Theoretically, it is understood that a *single* Zn impurity induces an effective, localized spin-1/2 moment in the vicinity of the dopant; such localized moments immediately destroy the spin gap.[19,22,25] When there is a small but finite density of dopants, these effective spin-1/2 moments interact with each other, and currently there is no consensus on what the asymptotic low-temperature behavior is. Sigrist and Furusaki[20] argued that the system can be mapped onto an effective model that is made of these *effective* half spins induced by the dopants forming a spin-1/2 chain, with random AF and ferromagnetic (F) couplings; this model is known to form large effective spins and exhibits Curie susceptibility at low T: $\chi \sim 1/T$.[5] On the other hand, Gogolin and co-workers,[27,28] used the bosonization method to map the problem to a Dirac fermion with random mass, and concluded that the low T susceptibility behaves as $\chi \sim 1/(T \log^2 T)$, which is the same as the random singlet phase;[3] no large moment formation was found in their work. Existing exact diagonalization[19] and quantum Monte Carlo calculation[19,21,23] do not have large enough system size to unambiguously resolve this discrepancy.

In this work we study disordered two-leg spin ladders with both bond randomness and site dilution (corresponding to Zn doping), using the RSRG method. As discussed earlier, in principle Zn doping introduces two types of disorder. Technically the presence of bond randomness is useful to us

in our study, as it introduces a separation of energy scales and justifies the usage of the RSRG method. Using this method we are able to study systems with sizes 100 times larger than those accessible in quantum Monte Carlo studies. In addition to the nonmagnetic (Zn) doping, we also study theoretically for the first time magnetic doping by replacing the Cu ions with Ni ions (or doping with $S=1$ impurities), a situation already realized experimentally.[29]

Our results are summarized as follows. As the RSRG procedure is carried out, effective spins (or moments) larger than 1/2 start to form; these large moments persist and grow without bound as the energy scale is lowered, regardless of the dopant concentrations (as long as it is nonzero) or the strength of bond randomness. Thus the presence of dopants drives the system into a new phase which is controlled by large spins, i.e., the large spin phase; the susceptibility at low temperature remains universal and follows $1/T$ behavior as T goes to zero. The $1/T$ Curie behavior comes from the spins coupled together forming larger effective spins. Such a behavior is very similar to that of random AF-F spin chain studied by Westerberg *et al.*,[5] as anticipated by Sigrist and Furusaki. While for any finite temperature range it is very difficult to distinguish between $\chi\sim 1/T$ and $\chi\sim 1/(T\log^2 T)$, we further find that the Curie coefficient of χ approaches that predicted by Sigrist and Furusaki based on analogy to the random AF-F spin chain. We thus conclude that the low energy behaviors of disorder spin ladders are the same as those of random AF-F spin chains.

The remainder of the paper is organized as follows. In Sec. II we introduce the model we use and review the application of RSRG to this model. In Sec. III we present our numerical results, compare them to previous works, and discuss the significance of our results. In Sec. IV we summarize our work and discuss the implications of our results.

II. MODEL AND REVIEW OF PREVIOUS RESULTS

The model we consider in this work describes a disordered antiferromagnetic two-leg spin-1/2 ladder. The Hamiltonian for this model is given by

$$H=\sum_{i=1}^{N-1}\sum_{j=1,2} J_{i,j}\mathbf{S}_{i,j}\cdot\mathbf{S}_{i+1,j}+\sum_{i=1}^{N} K_i\mathbf{S}_{i,1}\cdot\mathbf{S}_{i,2}, \qquad (1)$$

where $\mathbf{S}_{i,j}$ is a spin-1/2 operator when there are no dopants, and the positive coupling constants $J_{i,j}$ (couplings along the chains, or legs of the ladder) and K_i (couplings between the chains, or along the rungs of the ladder) are distributed randomly according to some probability distributions $P_{\parallel}(J_{i,j})$ and $P_{\perp}(K_i)$. N is used to represent the number of sites for a single chain. The dopant concentration is given by z, namely, we put $2Nz$ nonmagnetic impurities (Zn doping) or magnetic impurities (Ni doping) on the ladder; for nonmagnetic impurities we simply remove the spins at the impurity sites, while for magnetic impurities we replace the spin-1/2 operators by spin-1 operators at the impurity sites.

We use the real-space renormalization-group method to study this problem. Application of the RSRG procedure with proper extensions to the ladder systems has been discussed at length in Ref. 15, and we refer the readers to that paper for details. For the purpose of later comparison, here we briefly review some of the relevant results of that work, which studies the effects of bond randomness *without* non-magnetic or magnetic (spin-1) dopants going into the ladder.

When the RSRG procedure is carried out in ladders with no dopants, it was found that ferromagnetic (F) couplings are generated, and some effective spins with sizes bigger than 1/2 are formed because these ferromagnetic bonds may become the strongest bond in the system at some stage of the RG.[15] However, the percentage of these large effective spins remains low at all stages of the RG, and in the low-energy limit, their percentage *decreases* as the energy scale is going down, due to the fact that the overall strength of the ferromagnetic bonds becomes much weaker than that of the antiferromagnetic bonds, even though they have roughly the same numbers. Such a behavior may be understood in the following way. With nearest neighbor couplings only, the ladder has a bipartite lattice structure which means the system can be divided into two sublattices (A and B), and spins sitting on sublattice A are always coupled to spins sitting on sublattice B, and vice versa. In the absence of dopants, the number of spins in the two sublattices are strictly equal, and Marshall's theorem[30] dictates that the ground state is a total spin singlet in this case. Heuristically this is easy to see: the spins in the same sublattice tend to be parallel while those in opposite sublattices tend to be antiparallel, and there is a total cancellation when the number of spins are the same in the two sublattices. The disappearance of large effective spins in the low-energy limit is simply a reflection of this cancellation effect.

As we will see below, the situation becomes very different in the presence of dopants. In this case the dopants go onto lattice sites randomly, thus there are *fluctuations* in the numbers of dopants going onto the two different sublattices, even though *on average* they are the same. Such fluctuations destroy the perfect cancellation discussed above, and as we see below, lead to large moment formation in the long-distance, low-energy limit, which in turn changes the thermodynamic properties of the system qualitatively.

III. NUMERICAL RESULTS

We present numerical results for the spin ladder with the length of the ladder up to 100 000 (200 000 total spins). We decimate the strongest bond in the system defined as the bond with the largest energy gap, Δ_0, between the ground state and the first excited state. The decimation process is repeated until the number of spins left is less than 1% of the original number of spins in the system. The initial distributions are taken to be in power-law form:[31]

$$P_{\parallel}(J_{i,j})=(1-\alpha)J_{i,j}^{-\alpha},\quad 0<J_{i,j}<1,$$

$$P_{\perp}(K_i)=\frac{1-\alpha}{\Lambda^{1-\alpha}}K_i^{-\alpha},\quad 0<K_i<\Lambda. \qquad (2)$$

Here $0\leqslant\alpha<1$ is the measure of bond disorder (the bigger α, the stronger the randomness strength), and Λ is the anisot-

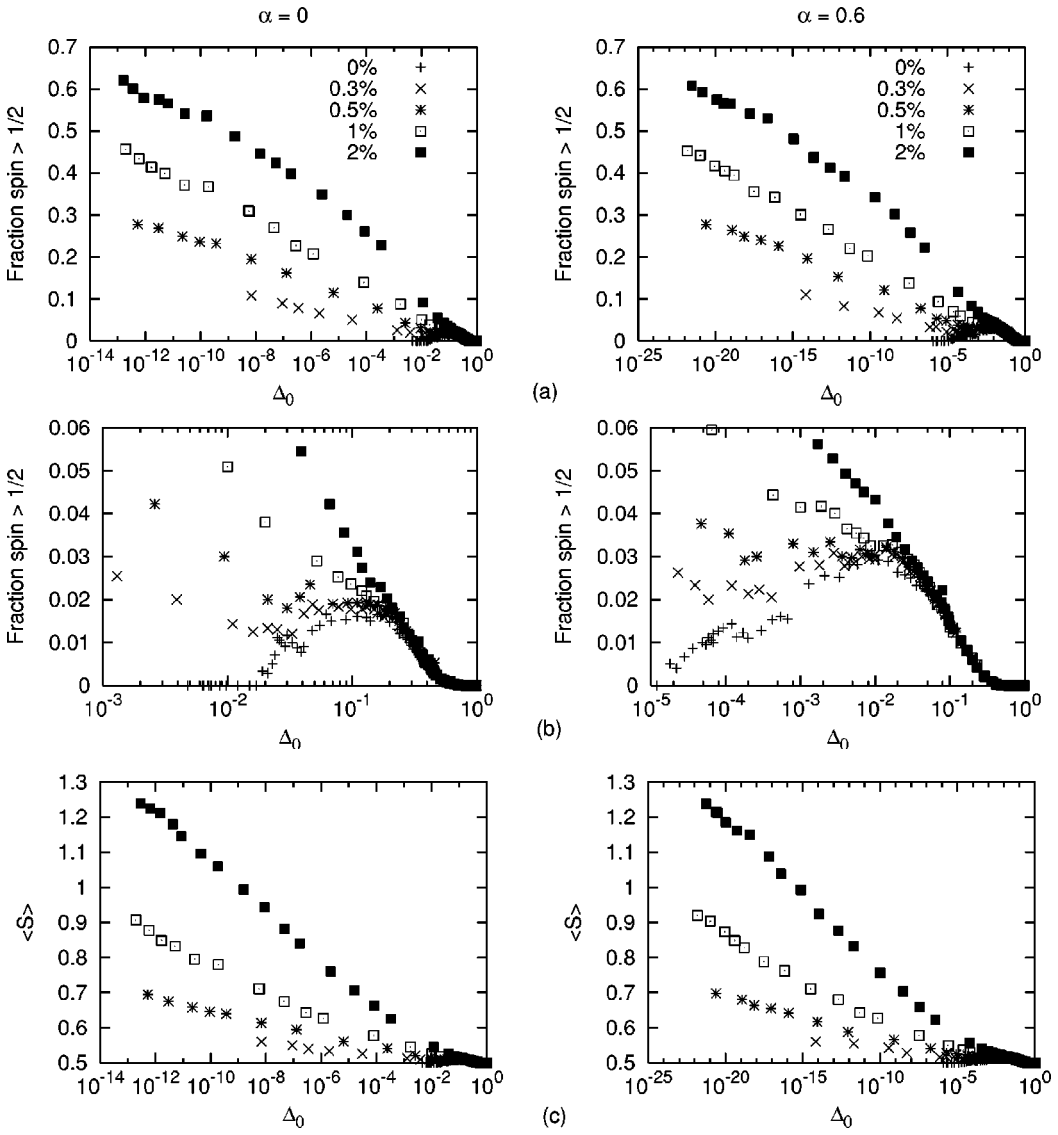

FIG. 1. The results from numerical calculations for nonmagnetic impurities. The left column is for $\alpha=0$ and the right column for $\alpha=0.6$, both with $\Lambda=1$. The number of spins on a single chain is $N=100000$. (a) and (b) The fraction of spins larger than 1/2, and (c) the spin size average as a function of cutoff Δ_0 with different dopant concentrations. All are sample averaged. A more detailed view of the high temperature part from (a) is shown in (b). The error bars, not shown in the figures, are comparable to the size of the data points.

ropy parameter; in the limit $\Lambda\rightarrow 0$ the two chains decouple. The nonmagnetic or $S=1$ magnetic dopants are distributed randomly throughout the system.

We start by discussing the effects of nonmagnetic dopants on the spin ladder. Due to the generation of F bonds in the RSRG procedure, effective spins with sizes bigger than 1/2 are formed as the RSRG procedure is carried out. The question whether or not these large effective spins proliferate at low energy is very important. In the undoped case (with bond randomness only), we have shown[15] that these large effective spins do *not* proliferate for the reasons discussed in Sec. II. The situation becomes completely different when a finite percentage of dopants are introduced into the ladder. This is shown in Figs. 1(a) and 1(b), where we plot the fraction of spins larger than 1/2, and in Fig. 1(c) where we plot the average spin size for different dopant concentrations as a function of cutoff Δ_0, for $\alpha=0$ and $\alpha=0.6$, both with $\Lambda=1$. For dopant concentrations bigger than 1% there is a very clear indication for large spin proliferation at low energy. The fraction of spins larger than 1/2 grows without bound as the energy scale is lowered. This picture is also supported by the results for the average spin size which show no sign of decreasing. This is in sharp contrast with zero doping, which is also included for comparison. The behavior for lower dopant concentrations ($<1\%$) is more interesting. In this regime we see a clear turnover in the graphs where the fraction of spins larger than 1/2 initially increases with decreasing energy scale, reaches a maximum, then it decreases before it begins to rise again [see Fig. 1(b)]. Our interpretation of this behavior is the following. For very low dopant concentrations, the effects of the dopants are very weak, and the system behaves like an undoped spin ladder at higher energy scales down to a certain energy scale Δ_c. Below Δ_c, the effect of these dopants kicks in and eventually dominates

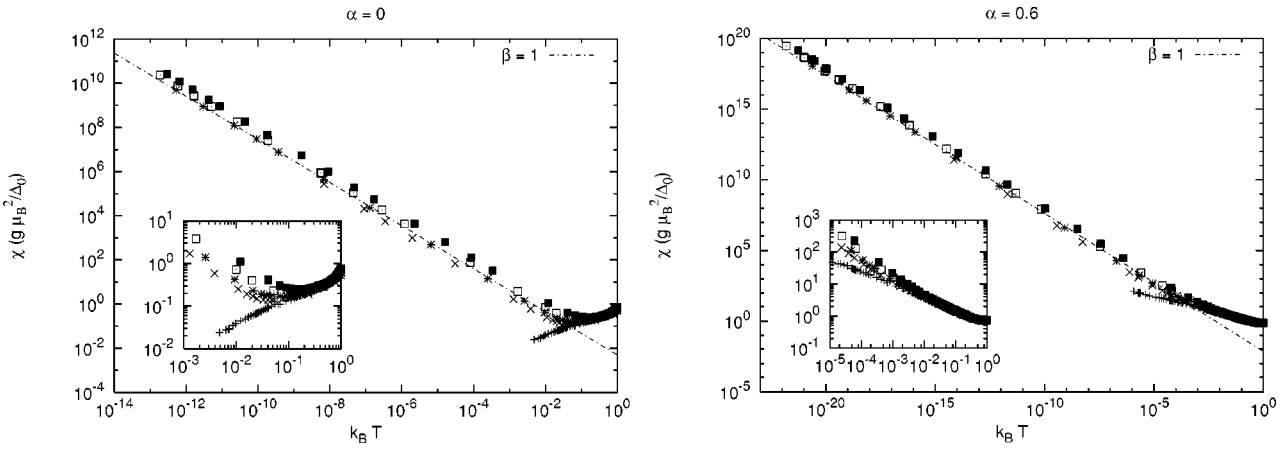

FIG. 2. The sample averaged susceptibilities per spin as a function of temperature with different nonmagnetic dopant concentrations for $\alpha=0$ and 0.6. The symbols have the same meaning as those in Fig. 1. The inset shows the part in susceptibility that crosses from one behavior to another. In both cases the $1/T$ line is drawn on the data as a guide to the eye. We do not include the error bars in the figures which are comparable to the size of the data points.

the physics, and the system flows into the large spin phase. Thus in a way the dopants are relevant perturbations in the RG sense.

The proliferation of large effective spins can be understood from the following analysis. The nonmagnetic impurities introduced into the system can go into either sublattice *A* or *B* with equal probability, so in *average* the number of dopants in the two sublattices are equal. However, due to statistical fluctuations, the number of dopants in the two sublattices are *not* equal in *specific* realizations of the random distributions. In particular, in any finite segments of the system, the fluctuations leave some of the half spins uncompensated for and destroy the perfect cancellation discussed in Sec. II, and the number of such spins grows as the square root of the size of the segment. The large effective spins that get generated under the RG have the same spin size as the ground state spin quantum number of the finite segments that they are made of; thus the proliferation of large spin at low energies is simply reflecting the growing fluctuation of the spin size of longer and longer segments.

The thermodynamic properties of the doped ladders are dramatically influenced by the presence of large effective spins in the system. Figure 2 shows the magnetic susceptibility for ladders with $\alpha=0$ and 0.6 with varying dopant concentrations, all with $\Lambda=1$. We associate the temperature with the cutoff Δ_0 where we stop the RG procedure and calculate the contribution from the active spins to the susceptibility. These active spins consist of undecimated half spins and effective spins larger than 1/2 generated during the decimation process. All the spins that have been decimated down to the cutoff Δ_0 do not contribute to the susceptibility. All the active spins are treated as free spins, so the contribution can be calculated using

$$\chi_{tot}=\frac{g\mu_B^2}{3k_BT}\sum_s N_s s(s+1), \tag{3}$$

where N_s is the number of spins left at energy scale $\Delta_0=k_BT$ for a given spin size s and the summation runs over all possible spin sizes.

In all cases we find that at low temperature the susceptibility can be fit very well to a Curie-like behavior $\chi\sim T^{-1}$, which is *insensitive* to specific details of the systems, like the strength of bond randomness and the dopant concentrations. Such a behavior agrees with the predictions of Sigrist and Furusaki[20] but it is very different from what we found earlier on the *undoped* ladders with bond randomness only, where the low-T susceptibility follows nonuniversal power laws with an exponent that depends on the bond disorder strength as well as the strength of the interchain interactions. We note that while our results agree with the Curie behavior predicted by Sigrist and Furusaki, due to the limited temperature range, they may also be fit to the Random-Singlet like behavior $\chi\sim 1/(T\log^2 T)$ that Gogolin and co-workers[27,28] suggested, which differs from the Curie behavior with a factor that only has a logarithmic dependence on T. In order to further clarify the situation, we study the dependence of the Curie coefficient on the parameters of the system and compare it with predictions made by Sigrist and Furusaki.

The $1/T$ Curie behavior is usually associated with free spins. In our case however, the $1/T$ dependence has a very different origin; it comes from the strongly correlated effective spins formed during the RG procedure, due to the existence of ferromagnetic couplings, which form clusters whose average size grow in a random walk fashion at low temperature. Sigrist and Furusaki,[20] in their effective model, have shown, using a random walk argument similar to that used in Ref. 5, that the Curie constant for finite dopant concentrations is given by

$$\chi T=z\mu_B^2/(12k_B). \tag{4}$$

On the other hand, if the effective spins induced by the dopants behave like free spins, and the Curie constant is given by

$$\chi T=z\mu_B^2/(4k_B). \tag{5}$$

We plot the Curie constants for $\alpha=0$ and 0.6, each with two different dopant concentrations, 2% and 4%, as a function of

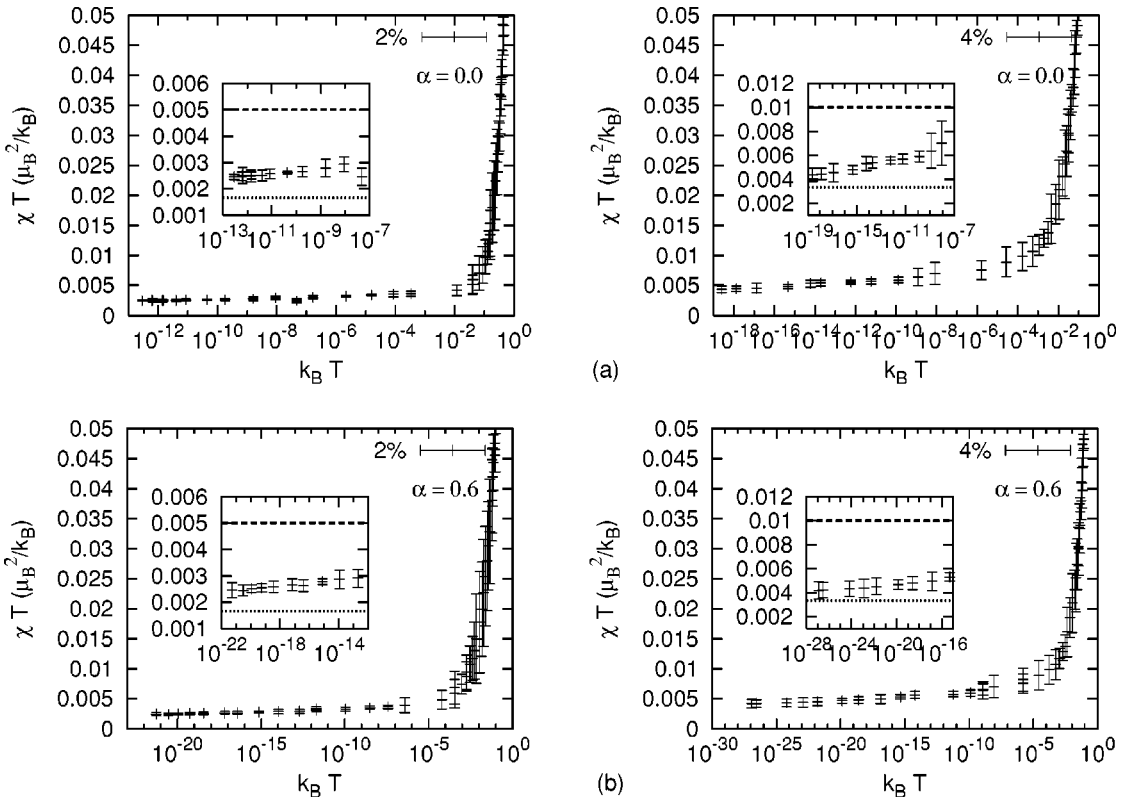

FIG. 3. The sample averaged Curie constants for (a) $\alpha=0$ and (b) $\alpha=0.6$. For each α we plot two different dopant concentrations. The dashed line in the inset is the Curie constant for free uncorrelated spin given by $z\mu_B^2/4k_B$ and the dotted line is the constant for strongly correlated spins given by $z\mu_B^2/12k_B$. In all cases, the Curie constants are always approaching the asymptotic limit given by Eq. (4) in the low temperature regime.

temperature in Fig. 3. The figure shows that, at low temperature, the Curie constants deviate significantly from the free spin Curie constant [Eq. (5)], and approach the asymptotic limit [Eq. (4)]. This strongly suggests that the effective spins are strongly correlated and the susceptibilities follow a $1/T$ behavior at low temperature due to the large moment formation. If the susceptibilities were to follow $1/(T\log^2 T)$, as Gogolin *et al.*[28] suggested, the Curie constants would go to zero at low temperature. While we do see that the Curie constants decrease with decreasing T, they are approaching constants given by Eq. (4) in the low-T limit, instead of going to zero. We thus conclude that our data strongly support the results of Sigrist and Furusaki.[20] We note that Miyazaki *et al.*[23] used the quantum Monte Carlo method to calculate the Curie coefficients of the doped ladder with different dopant concentrations. They were unable to obtain conclusive results for the coefficients due to the fact that the system size studied was not large enough to probe deep into the low temperature regime.

The temperature dependence of the susceptibility also gives us some information on how the system crosses over from one behavior (at high T) to another (at low T). In Fig. 2 we plot the susceptibility with different dopant concentrations for $\alpha=0$ and 0.6 as a function of temperature. The inset of each figure shows the part of susceptibilities where the crossover into a new behavior occurs. This crossover is particularly clear for $\alpha=0$. As we vary the dopant concentrations, from 0% to 2%, there is a clear turnover in the susceptibilities. In the undoped limit, the susceptibility goes to zero as $T\rightarrow 0$. For very small z, χ follows this behavior at higher T, as the effect of the dopants have not yet dominated the contribution to χ. However, at low enough temperature the effects of the dopants start to dominate; this is characterized as the susceptibility begins to increase and finally becomes divergent as the temperature is decreased below a certain crossover scale. The same behavior can also be seen for $\alpha=0.6$, although it is not as pronounced as for $\alpha=0$, because in the undoped limit χ is already divergent as a power law of T, and the power-law exponent is given by $\beta \approx 0.4$.[15] Introducing a small amount of dopants into the system alters the physics at sufficiently low temperature where the susceptibilities have different power-law exponents.

One important observation from the numerical results is that in the presence of a finite dopant concentration, the physics of the systems in the low-temperature limit is *not* sensitive to the choice of the distribution of the bond randomness. The low energy physics for two quite different bond randomness strengths $\alpha=0$ and 0.6, as shown in Figs. 1, 2, and 3, are essentially the same. In both cases the systems are controlled by large effective spins at low energy and the susceptibilities follow $1/T$ behavior at low temperature, and the Curie constants are approaching the same asymptotic limit, given by Eq. (4), which depends on the dopant concentration *only*. Thus the insensitivity of the results on the specific form of the bond distribution justifies our choice of

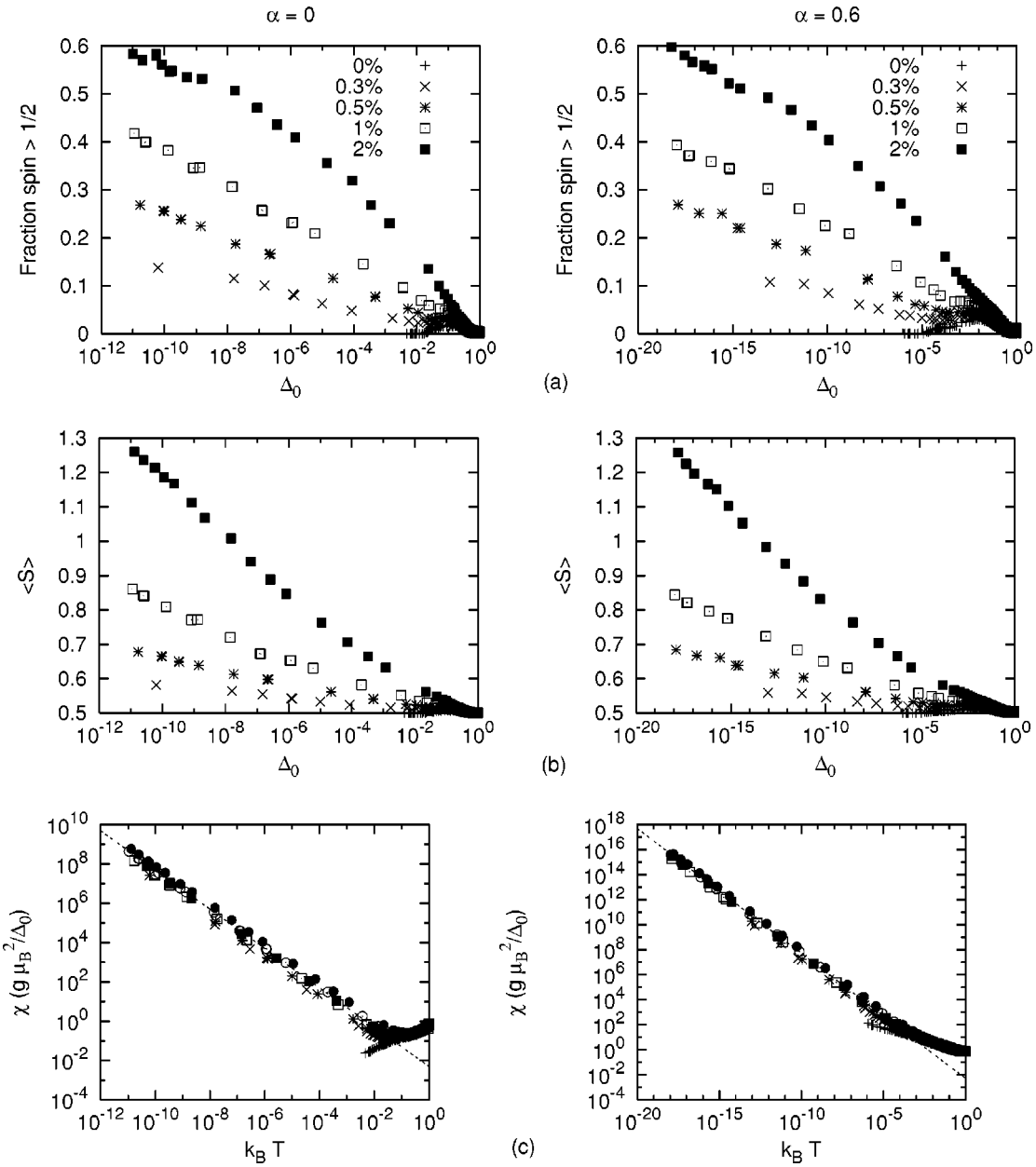

FIG. 4. The results from numerical calculations for magnetic impurities. The left column is for $\alpha=0$ and the right column for $\alpha=0.6$, both with $\Lambda=1$. The number of spins on a single chain is $N=100000$. (a) The fraction of spins larger than 1/2, (b) the average spin size as a function of cutoff Δ_0 with different dopant concentrations, and (c) the sample averaged susceptibilities per spin as a function of temperature with different magnetic dopant concentrations. The dashed line in (c) is the $1/T$ line drawn as a guide to the eye.

the bond distribution based on convenience. We note that it tends to flow to a power-law form even if it does not have such form initially; thus by choosing such a form, it puts one closer to the asymptotic form and reduces finite size effects.

We now turn our discussion to the effects of magnetic dopants with spin-1 on the spin ladders, which turn out to be very similar to those of nonmagnetic dopants. In Fig. 4 we plot the fraction of spin larger than 1/2, the average spin size as a function of cutoff Δ_0, and the susceptibility as a function of temperature with different magnetic dopant concentrations. As we can see from these figures, the qualitative behavior of the system doped with magnetic impurities at low energy is the same as that for a system doped with nonmagnetic impurities. Large spin formations are seen at low energies which grow continuously as the energy is decreased. The similarity in the effects of these two different types of dopants lies in the fact that they both induce spin-1/2 local moments, and uncompensated spins in finite segments, due to the fluctuation in the number of dopants going into the two different sublattices. This is the origin of the proliferation of large effective spins at low energies, and the $1/T$ Curie dependence of the susceptibility.

IV. SUMMARY AND DISCUSSIONS

We have studied antiferromagnetic two-leg spin-1/2 ladders with bond randomness and site dilution/magnetic impu-

rity by means of a real-space renormalization-group scheme. We found that there is proliferation of large effective spins at low energy for nonzero dopant concentrations. These large effective spins show the tendency of growing without bound as the energy scale is lowered. The susceptibility of the doped spin ladder follows a Curie-like $1/T$ behavior at low temperature. This behavior remains universal regardless of the dopant concentrations and the strength of bond randomness. We also find that the Curie coefficient is controlled by the dopant concentration only, and agrees with that predicted by Sigrist and Furusaki.[20] We conclude that nonzero dopant concentrations always drive the system into a phase dominated by the large effective spins. The large effective spins control the low temperature physics of the system, which makes the the doped ladder behave in many respects like a random spin chain with random ferromagnetic and antiferromagnetic interactions. This is very different from what we found in our earlier work for ladders with bond randomness only,[15] where no large spin proliferation was found, and the susceptibility at low temperature follows a nonuniversal power law: $\chi \sim T^{-\beta}$, with an exponent β that depends on the strength of bond randomness and the strength of interchain interactions.

Unfortunately at present we cannot make a direct comparison between our results and experiments, because in the systems studied so far the doped ladders all form long-range antiferromagnetic order at low temperature, due to the presence of three-dimensional interladder couplings not included in our study.

While reaching the same conclusions, we used a different approach in our study of the doped ladders as compared with the work of Sigrist and Furusaki.[20] They focused exclusively on the *effective* spins that are induced by the dopants, and neglected all the original spins, justified by the fact that without the dopants, and the effective spins they induce, the system is gapped. Thus the model they used is an *effective* model appropriate for describing the low-T properties of the system. In our study, on the other hand, we include all the original degrees of freedom (the original spins), and systematically lower the energy scale by decimating strong bonds one by one. Our approach thus treats high- and low-energy degrees of freedom on equal footing, and allows us to address both the high-T and low-T properties of the systems, and the crossover between them. Thus our study is complementary to that of Sigrist and Furusaki.

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[31] The specific distributions of the J's in real systems are not known. As we will see however, our results are not sensitive to the choice of these distributions; see below. So we may choose the distributions based on convenience of the study. Also from previous studies it is known that the distributions tend to flow to power-law forms at low energies, even if they do not take such forms initially.