Spin ladder systems have attracted a lot of theoretical and experimental interest recently. The physics of these materials is believed to be closely related to that of planar cuprate superconductors. Theoretical studies have shown that the ground state of the undoped two-leg ladder is a short range singlet resonating valence bond (RVB) state with a finite spin-gap $\Delta \approx 0.5J$. This has been verified experimentally in materials such as SrCu$_2$O$_3$ which has a gap of about 400K. Doping of holes into such a ladder system will probably lead to superconductivity of preexisting RVB singlet pairs with d-wave-type symmetries. This seems to be confirmed by the discovery of superconductivity in the ladder compound Sr$_{0.4}$Ca$_{13.6}$Cu$_{24}$O$_{41.84}$.  

In this letter we concentrate on another aspect, that of the suppression of the spin-gap by non-magnetic impurities. This was first observed in the zinc-doped compound Sr(Cu$_{1-x}$Zn$_x$)$_2$O$_3$. Marked changes in the low temperature behavior occur upon partially substituting copper with magnetically inert zinc atoms. Susceptibility and specific heat measurements indicate that the impurities destroy the spin gap and lead to a long range Néel order at low temperatures. Doping with 1% impurities is enough to order the system magnetically at a Néel temperature of 3K. As the impurity density increases up to 4% doping, the transition temperature increases to 8.2K. Further doping however reduces the Néel temperature.

A theoretical model for this material is weakly coupled spin 1/2 Heisenberg ladders. Zinc doping leads to a random depletion of this ladder. Several aspects of an isolated depleted ladder have been studied. It was shown that a single impurity induces a local moment with $S = 1/2$ around it. The effective interaction between these two induced moments is ferromagnetic or antiferromagnetic depending on the sublattices of the two impurities, which does not contradict the Néel order. The magnitude of the interaction is determined by the distance between the two impurities, and decays exponentially. Based on this localize-moment picture, the enhanced staggered susceptibility has been discussed, but the actual magnitude of the staggered susceptibility, Néel temperature and its doping-dependence have not been established.

Our main purpose in this paper is to calculate explicitly the uniform and staggered susceptibilities and to discuss the origin of the antiferromagnetic ordering and the phase diagram. Using the quantum Monte Carlo loop algorithm, we obtain the temperature dependence of susceptibilities, with high precision, down to extremely low temperatures. By analyzing these reliable numerical results, we can check whether the localized-moment picture is valid or not for various values of impurity density. It is found that such a picture is valid for only a 1% doping case. Then, to determine the phase diagram, we consider the three-dimensional inter-ladder coupling. Actually, the interladder coupling inside the plane is frustrated ferromagnetic and its effect is not clear at present. Therefore in this paper we assume an unfrustrated antiferromagnetic coupling which originates from the interplane coupling. This interaction is assumed to be weak compared to the intra-ladder couplings and we treat it in a mean field approximation. We estimate the staggered susceptibility of the three-dimensional coupled ladder system as

$$\chi_{MF} = \frac{\chi_s}{1 - J' \chi_s}. \quad (0.1)$$

Here, $\chi_s$ is the staggered susceptibility of an isolated depleted ladder calculated by quantum Monte Carlo simulation and $J'$ is an effective inter-ladder coupling. It is shown that, with this simple mean field approximation, we can explain the features of the phase diagram. In particular, we explain why a very small doping concentration is enough for the Néel order and why overdoping reduces the ordering.

In this study we investigate ladders with randomly-depleted sites. So far, numerical calculations have been performed for periodically depleted spin ladders.
However we will show that the random distribution of impurities is essential for explaining the phase diagram. A typical configuration considered here is shown in Fig. 1. In the randomly-depleted case, the impurities can cut the ladder into disconnected non-interacting pieces as shown in the configuration in Fig. 1. Such configurations have not been studied before. In our quantum Monte Carlo simulations we have averaged more than twenty random configurations on a ladder of 1000 \( \times \) 2 sites. In the following discussion all the error bars include the statistical error of the Monte Carlo sampling and that arising from the different configurations of the disorder. Using this loop algorithm we can achieve Monte Carlo simulations down to extremely low temperatures such as \( T = 0.005J \), which cannot be achieved by other algorithms. Thus we can discuss the temperature dependence precisely.

First we discuss the uniform susceptibility. In Fig. 2, the temperature dependence of \( \chi T \) is shown for 1%, 5%, and 10% doping. Initially there is a Curie behavior of free spins at high temperatures (\( T \gg \Delta \)), but we are not interested in this region and it is not shown in Fig. 2. At intermediate temperatures \( T < \Delta \) and for a 1% doping case, we observe a plateau with \( \chi T = g^2 \mu_B^2 x / 12 k_B \), with \( x \) being the impurity density. The horizontal lines in the figure are the lines with \( g^2 \mu^2_B x / 4 k_B \). This is consistent with the speculation that a local moment with \( S = 1/2 \) is induced around an impurity. In this case the mean distance between the two impurities is so large that the induced moments are essentially non-interacting in this temperature range.

However at higher levels of doping such as 5%, we can see from Fig. 2 that such an independent localized-moment picture does not hold. In this case the interaction between the localized moments has already set in in this temperature range.

Renormalization group arguments for the random spin chain predict a limiting value of \( \chi T \rightarrow g^2 \mu^2_B x / 12 k_B \) for \( T \rightarrow 0 \). These values are shown by arrows in Fig. 2. Apparently the predicted zero temperature limit is not observed for any impurity density down to \( T = 0.005J \). Since the localized moment picture seems to hold for 1% doping, the present result suggests that the renormalization group argument becomes valid at temperatures much lower than 0.005J. On the other hand, for higher doping cases, the independent localized-moment picture is no longer valid, so that the temperature dependence is smeared out. In this sense, the renormalization group argument does not seem to be relevant for the experiments. The results of the uniform susceptibility do not change within our statistical errors if we restrict the simulations to connected configurations.

The staggered susceptibility (see Fig. 3) exhibits behavior different from that of the uniform susceptibility. This is already visible in the undoped case, where the uniform susceptibility vanishes exponentially at low temperatures. The staggered susceptibility has a finite value at zero temperature. Impurities lead to a divergent staggered susceptibility at zero temperature. The magnitude of the staggered susceptibility at a high impurity density, depends strongly on whether we allow disconnected configurations or not. If we disallow such configurations we see a monotonous increase of the staggered susceptibility for a wide range of dopings. If we allow disconnected configurations we see that for more than 10% doping the staggered susceptibility decreases again. This explains the drop of the Néel temperature at higher doping levels. Impurities are needed to induce local moments. Overdoping however reduces these moments again by dilution. We will discuss this effect more quantitatively later in this study.

In Fig. 4 we plot the temperature dependence of \( \chi_s T \). One can see immediately that the picture of free spin-1/2 moments induced by non-magnetic impurities does not describe the staggered susceptibility well even at 1% doping. This behavior, quite different from that of the uniform susceptibility, can be described as \( \chi_s T = (a + b/T^2) T \) at low temperatures. The constant term \( a \) comes from the spins far from the impurities, and is almost the same as for the undoped case. The Curie term comes from spins around the impurities. However, the magnitude of this term is about 30 times as large as the corresponding term in the uniform susceptibility. The magnitude of this term at 1% doping can be explained by integrating the distribution of the excess \( z \)-component of spin around the impurity site, which is given by the DMRG technique, with a weakly staggered field. Thus, the antiferromagnetic correlation between the impurities cannot be seen in our QMC method down to \( T/J = 0.005 \) at 1% doping. The crossover from the Curie behavior of induced spins to the coupled local moments is estimated to occur around 4% doping.

Nagaosa et al. suggested that the staggered susceptibility diverges as

\[
\chi_s \propto \frac{1}{T^{1+2\alpha}},
\]

with \( \alpha \approx 0.22 \), based on the renormalization group arguments of the induced-moment picture. However Fig. 4 shows clearly that such a divergent behavior does not exist at \( T = 0.005J \). This is for the same reason as the case of uniform susceptibility as discussed above: The renormalization group argument is valid only in temperatures much lower than 0.005J.

We argue here that, even if there is no strong divergence as in eq. (2), the significant enhancement of the staggered susceptibility leads to antiferromagnetic ordering when the ladders are weakly coupled. Using the results of staggered susceptibility and the mean field approximation of eq. (0.1) we calculate the Néel temperature \( T_N \) from

\[
\chi_s(T_N) = \frac{1}{J'}.
\]

The exact magnitude of the inter-ladder coupling, \( J' \), is not known. We use the following estimates. The intra-
ladder coupling is taken to be \( J = 1000K \) as this gives the correct order of magnitude for the gap in the undoped ladder. The inter-ladder coupling depends on the density as \( J' = J'^0(1-x)^2 \), where \((1-x)^2\) is the probability of having two spins on adjacent sites. \( J'^0 \) is taken as \( 0.02J \sim 20K \). This gives the same maximum Néel temperature as seen in the experiments and is consistent with the order of magnitude of the inter-plane coupling in other cuprate materials. In Fig. 5, we show the Néel temperatures as a function of impurity density estimated by this mean field approximation. As the impurity density increases up to 10% doping, the transition temperature increases to 6K. Further doping however reduces the Néel temperature.

This phase diagram agrees qualitatively with the experiments on Sr(Cu\(_{1-x}\)Zn\(_x\))\(_2\)O\(_3\) and can explain both the unexpectedly high Néel temperatures as well as the maximum at substantially low levels of doping. The optimal doping ratio is however overestimated as compared to the experiments. We believe this to be an artifact of the mean field approximation which takes into account neither the effect of disorder on the inter-ladder couplings nor the exact structure and frustration effects of the inter-ladder coupling in the plane.

Note here that we can apply the mean-field approximation for more than 5% doping where there is a correlation between the induced spins. For small impurity densities (less than 4%), the antiferromagnetic correlation between the impurities cannot be seen in our QMC method at temperatures as low as \( T/J = 0.005 \). Therefore the validity of applying the mean field approximation for such small densities is not clear and the actual Néel temperature is expected to be much lower than the obtained value. In this sense, the question as to why high Néel temperatures are observed for small densities in Sr(Cu\(_{1-x}\)Zn\(_x\))\(_2\)O\(_3\) is unanswered.

In summary, we have found that randomly depleted spin ladders with weak three-dimensional interactions have antiferromagnetic ordering at low temperatures. The proposed speculation of independent free spin seems to be valid only at very low levels of doping of about 1% but fails at higher levels of doping. For higher impurity density, random depletion, which includes disconnected configurations in the ladder, is important to suppress the magnetic ordering.

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**FIGURE CAPTIONS**

**FIG. 1.** Example of a randomly depleted ladder. Filled circles denote copper sites with \( S = 1/2 \) spin and open circles denote \( S = 0 \) impurity sites. The configuration depicted here includes two impurities on neighboring spins which break the ladder into disconnected parts.

**FIG. 2.** Curie constant per site for several impurity densities (1%, 5%, and 10%). The horizontal lines and arrows denote the predictions of the second plateau and the zero temperature limit respectively for these doping levels.

**FIG. 3.** Staggered susceptibility per site as a function of temperature. Results for the undoped system and for 1% and 5% doping are shown by triangles, open circles and closed circles, respectively. The inset shows results for 1%, 5%, 12.5%, 15% and 20% doping at low temperatures.
FIG. 4. Temperature dependence of $\chi_s T$ for 1%, 5% and 10% impurity doping. Note that the magnitude of the susceptibility for each impurity density is much larger than that of the uniform susceptibility.

FIG. 5. Néel temperatures as a function of impurity density as estimated in the mean field approximation for the inter-ladder coupling. We have assumed $J = 1000K$. 
Fig. 1 Miyazaki et al.
Fig. 2 Miyazaki et al.
Fig. 3 Miyazaki et. al.

\[ \chi_s \] 
[1/J]

\( T/J \) (x1000)
Fig. 4 Miyazaki et al.

\[ \chi_s T \]

\[ T/J \]
FIG. 5 Miyazaki et. al.