Rapid Suppression of the Spin Gap in Zn-doped CuGeO$_3$ and SrCu$_2$O$_3$

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The influence of non-magnetic impurities on the spectrum and dynamical spin structure factor of a model for CuGeO$_3$ is studied. A simple extension to Zn-doped SrCu$_2$O$_3$ is also discussed. Using Exact Diagonalization techniques and intuitive arguments we show that Zn-doping introduces states in the Spin-Peierls gap of CuGeO$_3$. This effect can be understood easily in the large dimerization limit where doping by Zn creates “loose” S=1/2 spins, which interact with each other through very weak effective antiferromagnetic couplings. When the dimerization is small, a similar effect is observed but now with the free S=1/2 spins being the resulting S=1/2 ground state of severed chains with an odd number of sites. Experimental consequences of these results are discussed. It is interesting to observe that the spin correlations along the chains are enhanced by Zn-doping according to the numerical data presented here. As recent numerical calculations have shown, similar arguments apply to ladders with non-magnetic impurities simply replacing the tendency to dimerization in CuGeO$_3$ by the tendency to form spin-singlets along the rungs in SrCu$_2$O$_3$.

PACS numbers: 64.70.Kb,75.10.Jm,75.50.Ee

I. INTRODUCTION

The recent discovery of a spin-Peierls (SP) transition in the inorganic compound CuGeO$_3$ has received considerable attention. The effect is presumably caused by the coupling of the spins along the Cu−O$_2$ chains with three-dimensional phonons. This coupling dimerizes the chains reducing (increasing) the lattice spacing for, e.g., the even (odd) links, and thus promoting the formation of spin singlets along the short bonds. A spin-gap is formed in the spectrum, which is roughly the energy necessary to break the even-bond spin singlets. Soon after the discovery of this material, studies where Cu$^{2+}$ (S=1/2) was substituted by Zn$^{2+}$, Ni$^{2+}$ or Mn$^{2+}$ were reported. Based on magnetic susceptibility $\chi$ data analysis, a rapid suppression of the SP spin-gap was observed, with a small 2% concentration of Zn being sufficient to induce a transition to a new phase. $\chi$, specific heat and inelastic neutron scattering (INS) measurements have unambiguously shown that this new phase is a three dimensional (3D) Néel state. The simultaneous existence in the experimental data of features indicating SP and Néel order has also been noticed. Similar results have been reported with Si substituting Ge, and Ni,Mn,Mg replacing Cu. These results are apparently inconsistent with the gapless SP state predicted by mean-field theory. Recently, INS measurements in good quality samples of the temperature dependence of the superlattice peaks caused by the lattice dimerization were interpreted as evidence that the SP actually survives up to 6% Zn-doping.

The experimental evidence accumulated recently in doped CuGeO$_3$ shows two interesting phenomena namely (i) the unexpectedly rapid suppression of the SP order by Zn-doping, and (ii) its replacement by 3D AF order. Since INS results for 2% Zn-doping show already an important reduction of the SP transition at temperatures well above those where AF order starts, we conjecture that these two phenomena could be studied independently. It is remarkable that similar experimental results have been observed in a quasi one-dimensional “ladder” system, Sr(Cu$_{1-x}$Zn$_x$)$_2$O$_3$, where a spin-gap exists at x=0 due to the formation of spin-singlets along the ladder rungs, without lattice deformations. This suggests that the rapid suppression of a spin-gap by Zn-doping and its subsequent replacement by 3D Néel order may be a general phenomenon independent of the origin of such a spin-gap.

In this paper, the effect of non-magnetic Zn-doping on the SP state of CuGeO$_3$ is studied. A simple generalization to the case of Zn-doped ladders is also discussed. Using numerical techniques we show that the SP spin-gap is rapidly suppressed with Zn-doping, as observed experimentally. The effect is caused by unpaired spins created by the breaking of spin-singlets upon doping (in agreement with conclusions reached in recent numerical studies of even-leg ladder systems). These unpaired spins coupled forming an effective random spin-1/2 chain. Our analysis does not include intra-chain couplings that could stabilize the 3D AF order, analysis that is postponed for a future publication (however, in Sec. VII below we discuss the enhancement observed numerically of spin correlations upon Zn doping. This effect may contribute to the stabilization of the 3D AF order in real materials). To study the dimerization of a spin-1/2 chain...
it is natural to use the standard Heisenberg model including a static modulation of the nearest-neighbor (NN) exchange to account for the lattice distortion. In addition, recent studies have shown that a next-nearest-neighbor (NNN) spin-spin interaction is also needed to properly fit experimental susceptibility data. This is reasonable based on the structure of CuGeO$_3$ where Cu-Cu interactions bridged by two oxygens, Cu-O-O-Cu, may not be negligible. Thus, the model used here is

\[
\mathcal{H} = J_1 \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle \langle ik \rangle \rangle} \mathbf{S}_i \cdot \mathbf{S}_k - \]

\[
J_1 \delta \sum_{\langle ij \rangle} (-1)^i \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)
\]

where $\langle ij \rangle$ ($\langle \langle ik \rangle \rangle$) denote NN (NNN) sites, $\delta$ regulates the amount of dimerization, and $J_1, J_2$ are the NN and NNN Heisenberg couplings, respectively. The rest of the notation is standard. A modulation of $J_2$ is a higher order effect neglected here. To model CuGeO$_3$, we here use $\delta = 0.03$ and $J_2/J_1 = 0.24$, but other set of parameters could be chosen. The qualitative results described below do not depend crucially on the actual values of $\delta$ and $J_2/J_1$. The introduction of a non-magnetic impurity at site $i$ is mimicked in Eq.(1) by removing the $J_1$, $J_2$ and $\delta$-terms associated with this site. Note that the $J_2$ NNN coupling between sites $i-1$ and $i+1$ is not influenced by an eventual impurity located at $i$. This coupling across impurities plays an important role in linking chain segments severed by Zn-doping.

II. LARGE DIMERIZATION LIMIT

To guide the intuition on how Zn-doping affects the properties of model Eq.(1), it is convenient to study the limit where the dimerization $\delta$ is large, inducing a well-defined pattern of dimers (spin-singlets) in the ground state of the system. To illustrate the physics of the problem consider in Fig.1 a simple case where two impurities are randomly distributed on an 8-site chain using periodic boundary conditions (PBC). Impurity-1 is located at site 1, and the second one at different sites along the chain. There are six independent cases shown in Fig.1. In Fig.2a, the longitudinal dynamical spin structure factor $S^{zz}(\pi, \omega)$, which is the spectral decomposition of the operator $(1/N) \sum_i (-1)^i S^z_i$, is shown for the different positions of the impurities ($N$ is the number of sites). We consider momentum $\pi$ because in this subspace is where the smallest gap is found in the undoped system. The calculation was performed with standard Exact Diagonalization (ED) techniques, and using $\delta = 0.5$ (providing a strong exchange $J = 1.5J_1$ for half the links of the chain, and a weak exchange $J = 0.5J_1$ for the other half). We observed that for such a large dimerization finite size effects are small. We also verified that using chains with open boundary conditions (OBC) the conclusions of this discussion are unchanged.

![Fig. 1. Eight site chain with PBC and two impurities located in the positions of the full dots. The thick (thin) links denote a large (small) spin-spin coupling of magnitude $J_1(1+\delta)$ ($J_1(1-\delta)$). The dashed line linking some pairs of sites illustrates the presence of the $J_2$ coupling across impurities. The other $J_2$ couplings are not shown explicitly. The case of impurities at sites 1 and 7 is identical to having impurities at 1 and 3.](image-url)
sites 1–6, the two free spins are also weakly coupled into a singlet (weight at small $\omega$ is thus expected in agreement with Fig. 2a), but now being at even and odd sites their contribution to $S_{zz}^{\pi}(\pi, \omega)$ is not suppressed by the sign modulation. The effective coupling is $J_{eff} \sim 0.07J_1$; (6) finally, for impurities at 1 and 8, the free spins are again weakly linked and the intensity of the pole signaling their singlet-triplet excitation is enhanced by the momentum $q = \pi$. Thus, weight is expected at small $\omega$, as in Fig. 2a. Note that in this case the effective exchange is the smallest of the cases shown in Fig. 1, $J_{eff} \sim 0.02J_1$, since it is mediated by two strong dimers.

The peak at high $\omega$ moves down in energy as $\delta$ is reduced, as expected since it is originated by the singlet-triplet excitations of the dimers. On the other hand, the weight at small $\omega$ increases slightly its energy as $\delta$ is reduced. This is also compatible with the expectation that this feature is created by the AF coupling between “free” spins located at large distances from each other. A smaller $\delta$ increases the coupling between those spins since the tendency to form tight intermediate spin singlets is reduced. The evolution with $\delta$ of Fig. 3a is smooth and suggests that the intuitive explanation for the filling of the gap found at large $\delta$ could be qualitatively correct even in the more realistic regime of $\delta \ll 1$.

In Fig. 2b, the equal-weight average of $S_{zz}^{\pi}(\pi, \omega)$ over the impurity positions, denoted as $S_{av}^{\pi}(\pi, \omega)$, is shown. The result in the absence of impurities is also presented. It is clear that the introduction of impurities broadens the main peak, and, more importantly, it introduces weight inside the original spin-gap. The peak the smallest in intensity is caused by the special case when two impurities are at close distance, while the weight at very small $\omega$ is originated by the weak coupling into singlets of unpaired spins at large distances. The weight inside the gap should diminish as the density of impurities is reduced. Note that the effective interaction between spins at large distances can be understood using a decimation procedure analog to that employed by Dasgupta and Ma [19], in their study of random spin-1/2 chains.

**III. REALISTIC PARAMETERS**

In Fig. 3a, $S_{av}^{zz}(\pi, \omega)$ is shown as $\delta$ varies from 0.5 to 0.03 now using a chain of 20 sites and still 2 impurities. The peaks in the spectral weight have been given a width $\epsilon = 0.03$. (b) $S_{av}^{zz}(\pi, \omega)$ vs $\omega$ for $\delta = 0.03$ with a resolution $\epsilon = 0.006$. The labels of the peaks refer to the position of a second impurity on a chain of 20-sites with the first impurity located at site 1. The missing sites are related to those shown by symmetry, or they have negligible weight.

On Fig. 3b, $S_{av}^{zz}(\pi, \omega)$ is shown for $\delta = 0.03$ but with a higher resolution than used in Fig. 3a. Each peak represents the position of an excited state of the system with spin 1. The labels refer to the position of the second impurity, the first one being always at site 1. With this resolution, we can identify the origin of the two dominant features observed in the spin structure factor Fig. 3a at small $\delta$, namely the presence of low-$\omega$ and high-$\omega$ spectral weight. Fig. 3b shows a clear difference between even and odd sites. To understand this effect, note that the case where the second impurity is on an odd-site corresponds to a partition of the chain into two segments each with an odd number of spins. This effectively produces two $S=1/2$ states in each segment which are coupled by the relatively small $J_2$. The fact that such an effective “free” spin-1/2 in each segment is now spread over more than one lattice spacing contributes to the reduction of the effective exchange leading to the very small energy excitation of, e.g., the case where the impurities are at
sites 1 and 9. Thus, the same reasoning leading to the appearance of weight at small $\omega$ in the large $\delta$ limit, can be repeated for small $\delta$ still using $J_2$ as the link between $S=1/2$ effective spins.

**IV. INTUITIVE PICTURE**

The basic idea found numerically in the previous section is that Zn-doping divides a $S=1/2$ chain into segments each carrying either a total spin 1/2 or 0, depending on the number of spins in the segment. Then, at low temperatures each segment can be replaced by just one effective spin with coupling that depend on the distance between impurities (i.e. on the length of the segments). This is reminiscent of a similar approach followed recently to study spin-1/2 chains with randomly distributed ferromagnetic and antiferromagnetic bonds, as realized in $\text{Sr}_3\text{CuPt}_{0.5}\text{Ir}_{0.5}\text{O}_6$. If such a simple picture were correct for Zn-doped CuGeO$_3$, then the low-energy states of the complete doped chain could be calculated using an effective Heisenberg Hamiltonian that couples the effective neighboring spins.

Let us verify this scenario with the example of a 15-sites chain with OBC, and two impurities located at sites 4 and 10 dividing the lattice into three segments, each with an odd number of spins (Fig.4a). This setup is supposed to produce three effective spin-1/2 that we denote as $S_1$, $S_2$ and $S_3$. Its low energy Hamiltonian should be

$$\mathcal{H} = J_{12}S_1 \cdot S_2 + J_{23}S_2 \cdot S_3,$$

with $J_{12}$ and $J_{23}$ denoting effective exchange couplings, which in principle correspond to the singlet-triplet energy separation $\Delta_{ST}$ for the case where two adjacent segments of Fig.4a are analyzed in isolation. Diagonalizing exactly these coupled segments of 3 and 5 sites (Fig.4b) and 5 and 5 sites (Fig.4c), respectively, $J_{12}$ and $J_{23}$ have been calculated, and the resulting couplings used in the full three-segment 15-site problem of Fig.4a, which can also be exactly diagonalized to gauge the accuracy of Eq.(2). Fig.5 shows that the agreement between the exact result and the effective model is excellent providing support to the simple picture outlined above.

![FIG. 4. (a,b,c) Clusters used to illustrate the main intuitive ideas explained in the text. Each of the three segments carry an effective spin 1/2, denoted by $S_1$, $S_2$ and $S_3$. The circles are the impurities. The thick (thin) bonds have large (small) exchanges. The dashed line denotes some of the $J_2$ couplings.](image)

![FIG. 5. Energy levels of the effective Hamiltonian Eq.(2) compared against those of the 15-site cluster shown in Fig.4a. The eigenvalues of Eq.(2) were shifted by a constant such that its ground state energy coincides with that of the 15-site chain.](image)
V. COMPARISON WITH AVAILABLE EXPERIMENTS

In Fig.6a, the dynamical spin structure factor $S_{zz}^{\pm}(q, \omega)$, corresponding to the spectral decomposition of the operator $(1/N) \sum_i e^{iq \cdot r} S_i^z$, is shown parametric with momentum $q$. As discussed before, this is an “equal-weight” average over the positions of the impurities. Note that in real materials we may have some other probability distribution of hole positions determined by energy and entropy factors. Keeping this detail in mind, we proceed with the analysis of Fig.6a assuming an equal-weight distribution. For all momenta there is a clear distinction between the low and high-$\omega$ features discussed before. Fig.6b contains the $q$-dependence of the centroid of the low and high-$\omega$ spectral weight. It is clear that the features at large $\omega$ are remnants of the $S=1$ excitations of the $J_1 - J_2$ model (here $\delta = 0.03$ plays only a secondary role). These results also clearly show that weight is generated for energies inside the original SP spin-gap of the undoped system upon non-magnetic doping of the chains. This weight has a mild $q$-dependence. Thus, based on the intuitive picture developed in this paper the spin-gap is, rigorously speaking, destroyed for an infinitesimal concentration of Zn-doping. However, the weight inside the gap is also proportional to the impurity concentration and it may not be experimentally detectable until a finite doping threshold is reached.

![Fig. 6.](image)

FIG. 6. (a) $S_{zz}^{\pm}(q, \omega)$ vs $\omega$ for a 20-site chain with 2 impurities, parametric with $q$. The triangle denotes the rough position of the low and high-$\omega$ spectral weight; (b) Position of the dominant high and low-$\omega$ features of (a) compared with results in the absence of impurities.

It is important to remark that even though the gap is closed with doping, in Fig.6a clear remnants of the original undoped system are observed. Then, according to our picture INS experiments should indicate the presence of dimerization and AF correlations for Zn-concentrations where other probes, specially magnetic susceptibility measurements, indicate the disappearance of the spin-gap. Short-distance dimerization correlations tested by INS can survive the introduction of doping up to large impurity concentrations. However, $\chi$ defined as

$$\chi \propto \lim_{q \to 0} \int_0^\infty d\omega \frac{S_{zz}^{\pm}(q, \omega)}{\omega}$$

is dominated by small energy excitations even if they carry small weight. Using $q = \pi/10$ in Eq.(3), we calculated numerically $\chi$ at zero temperature using a 20-site chain with 0 and 2 impurities. The ratio $\chi_{\text{doped}}/\chi_{\text{undoped}}$ is about 40. Therefore this explains why $\chi$ measurements can show no trace of a SP phase, while INS still suggest its presence. It is the short-against long-character of the correlations that establish the difference. $\chi$ reacts to the long distance structure, while INS captures fluctuations at all distances.

VI. EXPERIMENTAL PREDICTIONS

The numerical and analytical results discussed in this paper suggest that CuGeO$_3$ looses its gap immediately after an infinitesimal amount of Zn replaces Cu. This dramatic effect produces interesting experimental consequences in addition to those described in the previous section. The localized spin-1/2 states in the vicinity of the nonmagnetic ions interact weakly through a Heisenberg Hamiltonian with antiferromagnetic couplings having a strength that depends on the distance between the Zn impurities. Since this distance is random, the couplings are random and locate the low energy behavior of Zn-doped CuGeO$_3$ in the universality class of the random exchange Heisenberg model. This low energy effective model, defined by the Hamiltonian

$$H = \sum_{\langle ij \rangle} J_{ij} S_i \cdot S_j$$

in a standard notation, has been analyzed using decimation techniques. In this method the pair of spins with strongest coupling in the random chain is eliminated by considering the interaction with the neighboring pairs as a perturbation. With this procedure a new (small) coupling is generated between the two spins neighboring to the eliminated pair. The iteration continues until a single pair of spins remains. As this decimation procedure evolves, the original initial distribution $P_0(K)$ of random bonds ($0 < K < J$) changes into a power-law distribution $P(K) \sim K^{-\alpha} - 1$, where $\alpha$ depends only weakly with the "cutoff" $J$. In this asymptotic regime, corresponding experimentally to sufficiently low temperatures, the susceptibility behaves as $\chi \sim T^{-\alpha}$, and the magnetic contribution to the specific heat as $C \sim T^{1-\alpha}$ ("random singlet phase"). Since $\alpha$ is positive, the susceptibility diverges as the temperature approaches zero. For magnetic fields much stronger than the temperature but much weaker than $J$ (all in energy units), the magnetization behaves as $m \sim H^{1-\alpha}$ (note that the actual...
value of $\alpha$ is difficult to calculate within the accuracy of our techniques since we need to know the initial distribution of random exchange couplings). These predictions have been verified in organic materials and also in MgTiOBO$_3$ [22] where it was found that $\alpha \sim 0.8$. Thus, if indeed the low energy behavior of Zn-doped CuGeO$_3$ can be mapped into a random Heisenberg model, just one parameter ($\alpha$) is needed to fit the power-law behavior of the susceptibility, specific heat and magnetization as described above. This is a prediction of our theory for Zn-doped spin-Peierls systems in the regime of small Zn concentration, and it breaks down at higher concentrations when the experimentally observed transition to a long-range 3D Néel order takes place.

VII. CONCLUSIONS, EXTENSION TO LADDERS, AND ENHANCEMENT OF SPIN CORRELATIONS

Summarizing, here we have developed a simple picture of the effect of Zn-doping on compounds with a spin gapped ground state. The idea is that each non-magnetic impurity produces an effective S=1/2 “free” spin which may be an actual S=1/2 electron or hole located at a nearest-neighbor site (in the limit of large dimerization), or it may correspond to many-body S=1/2 states of severed odd-site chains (in the limit of weak dimerization). Similar concepts have been used in the context of other compounds that provide physical realizations of random ferro-antiferromagnetic exchange S=1/2 chains. [23] The interaction of the free spins is mediated by the rest of the spins, and thus it can be very small at low Zn concentration. Such a weak coupling induces low energy spectral weight inside the spin gap in calculations of the dynamical spin structure factor. This effect occurs at all Zn densities, with a weight proportional to such density. Clear remnants of the dynamical features corresponding to an undoped chain are observed. On the other hand, the magnetic susceptibility, which reacts to low energy excitations, indicates the rapid suppression of the spin gap at very low impurity concentrations. Thus, an analysis of INS and $\chi$ data may naively produce different conclusions unless we recall that INS reacts to fluctuations at all distances, while $\chi$ is affected mostly by the long-range order in the system.

The calculations presented in this paper have been carried out for the special case of Zn-doped CuGeO$_3$, but the results can be easily generalized to other system with spin-gaps. Of particular interest are the recent results reported in ladders, [14] where a rapid collapse of the spin-gap was observed. To understand this result consider the limit where the rung exchange $J_\perp$ is dominant (which is the analog of the large $\delta$ limit studied in Sec.II), and thus the ground state is made out of rung spin-singlets. [11] In this case each Cu replaced by Zn actually induces a loose S=1/2 spin, as recently observed numerically by Sandvik et al. [12] (see Fig.7).

Their interaction should lead to small spectral weight in the dynamical structure factor, similar to the results for the SP ground state. Then, INS studies of Zn-doped ladders at a small Zn concentration would indicate the survival of weight at energies of the order of the original spin-gap, in regions where susceptibility measurements show that the gap has collapsed (which occurs rapidly upon doping according to the intuitive picture discussed in previous sections and before in Ref. [12]). Recently, related work in the context of Zn-doped ladders have appeared with conclusions similar to ours.

An important issue that we have not addressed in detail in this work is the behavior of the spin-spin correlations after Zn doping of CuGeO$_3$. If these correlations are enhanced upon such doping, then a weak interchain coupling may lead to the stabilization of a 3D Néel order as observed experimentally.
the correlations is observed at short distances. Currently we are investigating the origin of this enhancement which seems to be generic of a variety of Zn-doped materials. Other systems with spin-gaps should behave similarly. S=1 chains have been recently studied \cite{24} and weight inside the Haldane gap was reported. In two dimensions, a similar behavior should be observed in Zn-doped cuprates in the temperature and density regime where a spin-gap is observed for the undoped case. \cite{22}

VIII. ACKNOWLEDGMENTS

We thank A. Sandvik, M. A. Continentino, S. Haas and A. Moreo for fruitful discussions. GBM acknowledges the financial support of the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq-Brazil). ED is supported by grant NSF-DMR-9520776. Additional support by the National High Magnetic Field Lab and the Supercomputer Computations Research Institute at Florida State University are acknowledged.

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