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A single-chain analysis of doped quasi-one-dimensional spin-1 compounds: paramagnetic versus spin-1/2 doping

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Abstract. We present a numerical study of single-chain models of doped spin-1 compounds. We use low-energy effective one-dimensional models for both the cases of paramagnetic and spin-1/2 doping. In the case of paramagnetic doping, the effective model is equivalent to the bond disordered spin-1/2 chain model recently analysed by means of the real space renormalization group by Hyman and Yang. By means of exact diagonalizations in the XX limit, we confirm the stability of the Haldane phase for weak disorder. Above a critical amount of disorder, the effective model flows to the so-called random singlet fixed point. In the case of spin-1/2 doping, we argue that the Haldane phase should be destabilized even for weak disorder. This picture is not in contradiction with existing experimental data. We also discuss the possible occurrence of (unobserved) antiferromagnetically ordered phases.

1. Introduction

Much effort has been devoted over the last decades to understanding the behaviour of quasi-one-dimensional spin systems. These systems exhibit a rich variety of phases due to the enhanced quantum fluctuations in reduced dimensionality. One of the most famous and studied one-dimensional properties is the so-called Peierls and spin–Peierls instability. Organic compounds exhibiting such an instability were synthesized as early as the 1970s [1]. The more recent discovery of the inorganic CuGeO_3 spin–Peierls compound [2, 3] with a spin–Peierls temperature up to about 15 K renewed the interest in spin–Peierls systems, especially in the role of various possible doping mechanisms. In fact, it is possible to dope these inorganic compounds in a very controlled fashion with both magnetic and paramagnetic ions (see for instance [4] and [5] for Si doping, [6–8] for Zn doping and [9] for Ni doping). It turns out that an antiferromagnetic (AF) phase is induced even for very small impurity concentrations, with a maximal Néel temperature of the order of 4 K at doping to a few per cent. Neutron scattering experiments [5, 7] have unambiguously identified this phase as an antiferromagnet with a coexistence of gapless AF excitations at low energy and spin–Peierls excitations at higher energy [5, 8], and also with important disorder effects [7]. In a recent work, the present authors have shown that insight into the physics of these compounds can be gained by a single-chain model [10, 11], which does exhibit an enhancement upon doping of low-energy AF fluctuations, coexisting with higher-energy spin–Peierls features. More specifically, by means of the renormalization procedure proposed in [12], it was shown in [11] that this effective model belongs to the random singlet (RS) universality class of a random Heisenberg model [13]. Although within a single-chain model it is difficult to handle rigorously the interchain couplings that stabilize the Néel phase, nevertheless this

approach does emphasize the conjugated effects of low-dimensional quantum fluctuations and disorder, which seem to be an important ingredient in the physics of these compounds.

Another type of low-dimensional quantum phase of spin systems is the Haldane gapped phase [14] of a spin-1 AF chain, which has been observed in quasi-one-dimensional compounds [15]. Quite interestingly, a spin-1 chain and a dimerized spin-1/2 chain belong to the same quantum phase, with a non-vanishing string order parameter [16]. However, as we are going to argue, in spite of this analogy, the spin-Peierls and spin-1 chains behave very differently upon doping. Moreover, paramagnetic doping of spin-1 chains is very different from spin-1/2 doping. In fact, there is some experimental evidence that the Haldane phase is robust against paramagnetic doping: it was found in [17] that the Haldane gap persists up to 20% doping in the $\text{NiC}_2\text{O}_4\text{DMIZ}$ compound where Ni was substituted by Zn. On the other hand, it was shown in [18] that doping NENP compounds with less than 1% of Cu leads to a Curie behaviour in the susceptibility, with a Curie constant 4.6 times larger than the one of the free Cu^{2+} ions. These experiments thus show a very different behaviour of quasi-one-dimensional spin-1 compounds upon doping: weak paramagnetic doping preserves the Haldane gapped phase whereas weak spin-1/2 doping induces low-lying energy states. We argue that the behaviour of quasi-one-dimensional spin-1 systems upon both spin-1/2 and paramagnetic doping can be understood on the same basis as that of the doped spin-Peierls compounds, namely, in the framework of a one-dimensional effective model with a rigorous treatment of disorder effects. We also examine the issue of whether a strong-doping AF phase could appear for paramagnetic doping.

This article is organized as follows. In section 2 we introduce an effective one-dimensional model for paramagnetic doping. The zero-temperature phases of this model were analysed recently [19, 20]. We check the nature of the zero-temperature phases using exact diagonalizations of the effective spin-1/2 model at zero-temperature in the XX limit. We then argue that, in close analogy to our approach for spin-Peierls compounds, the zero-temperature Haldane phase of the effective model should be robust against switching interchain couplings whereas the RS phase should possibly turn into an AF phase. Section 3 is devoted to an analysis of an effective model for spin-1/2 doping. We show that in this case the Haldane phase is destabilized by an infinitesimal doping. In section 4, we analyse the question of whether an AF phase might appear for large paramagnetic doping. We carry out exact diagonalizations of small clusters, the interchain coupling being treated in mean field. These diagonalizations support our prediction, namely the possible emergence of a strong disordered AF phase. Finally, section 5 is devoted to some final remarks.

2. An effective one-dimensional model for paramagnetic doping

2.1. The model

Based on the valence bond (VB) description of a spin-1 chain [21], Hyman and Yang [19] first argued that bond-disordered spin-1 chains can be described by the following low-energy effective model: introducing bond disorder amounts as a first approximation to ‘breaking’ the chain into clusters of consecutive strongly coupled spins. As a consequence, effective low-energy spin-1/2 degrees of freedom appear at the edges of these segments [22]. Two neighbouring 1/2 spins, one at the right and the other at the left edges of two consecutive segments are then assumed to be weakly AF coupled.

This model should be also appropriate to describe the effects of doping by paramagnetic ions. In fact, if a magnetic ion is substituted by a non-magnetic impurity (as in the case of Zn doping in $\text{NiC}_2\text{O}_4\text{DMIZ}$ [17]), one expects that two spin-1/2 edge excitations appear

on the right and the left of the impurity, which are coupled by an AF exchange J_2 weaker than the bulk exchange J_H . In addition, two spin-1/2 moments on the opposite edges of a segment are also coupled by virtual polarization of the spin-1 background. Assuming that these two edge moments \mathbf{S}_1 and \mathbf{S}_2 are at distance l , the effective interaction mediated by the spin-1 background is [22]

$$\mathcal{H}_{1,2}(l) \sim J_H(-1)^l e^{-l/\xi_0} \mathbf{S}_1 \cdot \mathbf{S}_2 \quad (1)$$

where we have discarded a $1/\sqrt{l}$ prefactor, irrelevant to the physics we want to discuss.

2.2. Exact diagonalizations in the XX limit

Hyman and Yang [19], and also Monthus *et al* [20], carried out a renormalization group (RG) analysis of this effective model. These authors showed that, for weak disorder, the system remains in the gapped Haldane phase. As the disorder increases above a critical value, the system exhibits a second-order transition to the RS phase. The aim of this section is to check this prediction by means of exact diagonalizations in the XX limit. Due to the presence of ferromagnetic bonds, the XX limit is not expected to give a good description of the isotropic XXX model, in contrast to what is found when all bonds are AF. However, like the XXX model analysed in [19], the XX version may have in principle two different phases for weak and strong doping. Therefore, if one is only interested in confirming the existence of this phase transition, the XX model should be sufficient.

In the XX limit, this model can be mapped onto non-interacting spinless fermions via a Jordan–Wigner transformation [23]:

$$\mathcal{H} = \frac{1}{2} \sum_i J_i (c_{i+1}^+ c_i + c_i^+ c_{i+1}) \quad (2)$$

where the label i denotes the sites carrying spin-1/2 moments and J_i is calculated according to the aforementioned rules. We restrict ourselves to an even number of spin-1/2 magnetic moments, so that the fermions are periodic and the Fermi sea is half filled. Moreover, the impurities are distributed according to a Poisson law with an average impurity to impurity distance $\langle l \rangle$.

Increasing the strength of disorder can be done either by decreasing J_2 or by reducing the average impurity to impurity distance $\langle l \rangle$. We have found that in both cases increasing the strength of disorder leads to an increase of the correlation length and finally to power law correlations. We now present our calculations.

In order to calculate the $S_0^z S_R^z$ correlations, we first diagonalize the tight-binding Hamiltonian

$$\mathcal{H}^{(TB)} = \frac{1}{2} \sum_i J_i (|i+1\rangle\langle i| + |i\rangle\langle i+1|). \quad (3)$$

If $\Psi_\alpha = \sum_i \Psi_i^\alpha |i\rangle$ denotes the eigenstates of (3), the average correlations are $\langle S_i^z S_j^z \rangle = A_{i,j} - B_i/2 - B_j/2$, with

$$A_{i,j} = \sum_{\alpha, \beta \in FS} (\Psi_i^\alpha \Psi_j^\beta)^2 + \sum_{\alpha \in FS} \sum_{\beta \notin FS} \Psi_i^\alpha \Psi_i^\beta \Psi_j^\alpha \Psi_j^\beta \quad (4)$$

$$B_i = \sum_{\alpha \in FS} (\Psi_i^\alpha)^2. \quad (5)$$

The correlations $\overline{\langle S_0^z S_R^z \rangle}$ are shown in figure 1 in the strong-disorder regime. This correlation function follows quite nicely a $1/R^2$ decay, as in the RS phase [13].

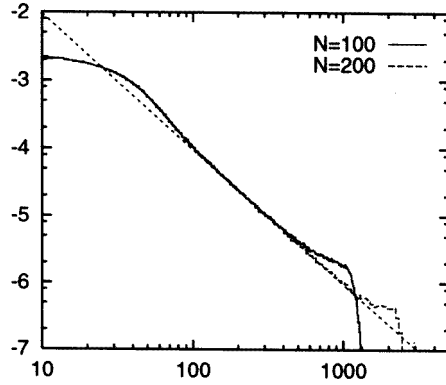


Figure 1. Logarithm of the correlation between the z component of the spins in the strong-disorder phase (paramagnetic doping), as a function of the spin–spin distance. The AF exchange between spins 1 is $J_H = 1$; the nearest-neighbour AF exchange is $J_2 = 0.1$; the correlation length in the Haldane phase is $\xi_0 = 20$ lattice spacings; the average distance between two impurities is $\langle l \rangle = 50$ lattice spacings. For $N = 100$ (200) impurities, we performed the average over 32 000 (2000) disorder realizations. The straight line is a fit to a $1/R^2$ behaviour.

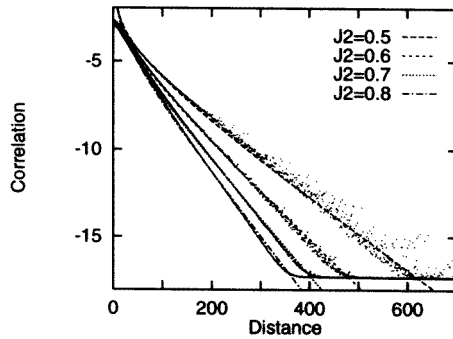


Figure 2. Logarithm of the correlation of the z component of the spins in the small-disorder phase (paramagnetic doping). This is a semi-log plot. The AF exchange between spins 1 is $J_H = 1$; the nearest-neighbour AF exchange is $J_2 = 0.5, 0.6, 0.7$ and 0.8 ; the correlation length in the Haldane phase is $\xi_0 = 20$ lattice spacings; the average distance between two impurities is $\langle l \rangle = 50$ lattice spacings. The correlations have been calculated for $N = 100$ impurities and averaged respectively over 50 000, 10 000, 6000 and 1300 realizations of the disorder. The solid lines are a fit to the form $\exp(-R/\xi)/R^2$, with respectively $\xi = 23 \pm 0.5, 17.5 \pm 0.5, 14.5 \pm 0.5, 13 \pm 0.5$. The saturation for correlations smaller than $\simeq 10^{-19}$ is due to round-off errors during the diagonalizations.

We now keep a constant dilution of impurities but increase the nearest-neighbour interaction J_2 across the impurities. This should drive the systems into the Haldane phase, with exponentially decaying correlation functions. This is clearly seen for the $S_0^z S_R^z$ correlation plotted in figure 2 in a semi-log plot for different values of J_2 . Notice in figure 2 an increase of the scattering of the data as the system approaches the zero-temperature critical point, in spite of an increase of the number of disorder realizations. This is a clear signature of critical fluctuations and thus consistent with the prediction that this zero-temperature transition is a second-order transition [19, 20].

We now consider the case $J_2 = 0.7$ in figure 2 and drive the system towards the infinite-

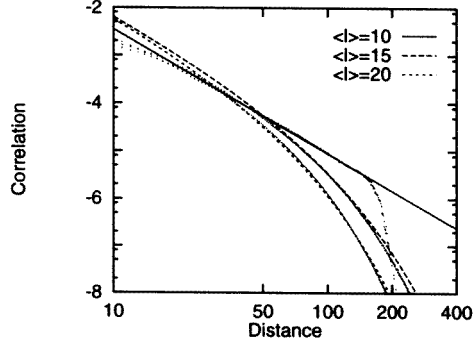


Figure 3. Logarithm of the correlation of the z component of the spins in the regime $\langle l \rangle < \xi_0$ (paramagnetic doping). This is a log–log plot. The AF exchange between spins 1 is $J_H = 1$; the nearest-neighbour AF exchange is $J_2 = 0.7$; the correlation length in the Haldane phase is $\xi_0 = 20$ lattice spacings; the average distance between two impurities is $\langle l \rangle = 10, 15$ and 20 lattice spacings. The correlations have been calculated for $N = 100$ impurities and averaged respectively over 3400, 10 000 and 25 000 realizations of the disorder. The solid lines are a fit to the power law $1/R^{2.4}$ in the case $\langle l \rangle = 10$; to the form $\exp(-R/\xi)/R^{2.4}$ with $\xi = 45$ lattice spacing for $\langle l \rangle = 15$; and $\xi = 30$ lattice spacings for $\langle l \rangle = 20$.

correlation-length phase by decreasing the average impurity to impurity distance $\langle l \rangle$. The resulting correlations are plotted in figure 3. We clearly see that decreasing the average distance between impurities drives the system towards a phase with critical correlations.

Although the effective model is valid only if $\langle l \rangle > \xi_0$, we have decided to investigate also the situation $\langle l \rangle < \xi_0$ just as a theoretical model. In this case we were not able to fit the correlation to the RS $1/R^2$ form. Instead, we found in the case of figure 3 a power law decay different from the RS one. Indeed, we find

$$\overline{\langle S_0^z S_R^z \rangle} \sim \frac{1}{R^{2.4}}. \quad (6)$$

This may be a finite-size effect: we know that the RG procedure is asymptotically correct since the disorder distribution becomes extremely large as the decimation is carried out. In the case $\langle l \rangle < \xi_0$, it is possible that, given the small sizes that we use, the distribution of bonds is not large enough, so that the correlation exponent has still not converged to the one of the RS. Also, we notice that the bond distribution is

$$P(|J|) = \frac{1}{2J_H} \frac{\xi_0}{\langle l \rangle} \left(\frac{|J|}{J_H} \right)^{-1 + \frac{\xi_0}{\langle l \rangle}} \theta(J_H - |J|) + \frac{1}{2} \delta(J - J_2). \quad (7)$$

In the case $\xi_0 < \langle l \rangle$, the smaller the exchange the larger the probability, which explains why in this regime the numerics works so well (see figure 1). In the opposite regime ($\xi_0 > \langle l \rangle$), the smaller the exchange the smaller the probability. In this case, we still expect the RS fixed point. However, it is not so obvious to exhibit it from a numerical point of view.

3. An effective one-dimensional model for spin-1/2 doping

3.1. The effective one-dimensional model

The effect of spin-1/2 doping in quasi-one-dimensional spin-1 NENP compounds was analysed in [18] where susceptibility measurements were carried out, and also in [22] by means of ESR measurements. These last experiments showed unambiguously that spin-1/2

doping amounts to releasing two spin-1/2 effective moments on the right and the left of the impurity, that are weakly AF coupled to the spin-1/2 impurity. This picture is not in contradiction with the susceptibility measurements [18], where the temperature variations of the susceptibility were found to have Curie behaviour. The effective model thus consists of disordered units of three consecutive spin-1/2 moments. The spin-1/2 moments experience a weak AF interaction J' and two consecutive units are coupled via the ferromagnetic or AF interaction (2).

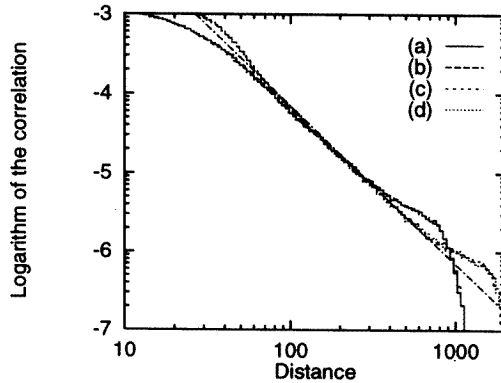


Figure 4. Logarithm of the correlations of the z component of the spins for the effective model of spin 1/2 doping. The data have been fitted to a $1/R^2$ dependence. We have chosen: $J_H = 1$, $\xi_0 = 20$, $\langle l \rangle = 50$ and (a) $J' = 0.8$, 102 spin-1/2 moments, (b) $J' = 0.1$, 102 spin-1/2 moments, (c) $J' = 0.8$, 204 spin-1/2 moments and (d) $J' = 0.1$, 204 spin-1/2 moments.

It is immediately clear that the RG description of this effective model is very different from the one in [19]. The reason is that, in the weak-disorder case, one first would replace all three spin-1/2 units by a spin 1/2. Therefore the effective model would be again a bond disordered $S = 1/2$ Heisenberg AF, but the bonds being either ferromagnetic or AF. This model has been investigated in [24] in quite some detail and shown to be unstable against a small disorder. In particular, the ground state is expected to have power law correlations. We check in section 3.2 that our effective model indeed has power law correlations in its ground state. However, the physics is expected to be very different from the RS: the low-temperature susceptibility has a Curie dependence whereas in the RS the low-temperature susceptibility diverges more slowly than a Curie behaviour ($\chi(T) \sim 1/(T \ln^2 T)$) [13]. Moreover, the low-temperature physics is dominated by large effective spins. Coming back to the physics of spin-1/2 doped quasi-one-dimensional spin-1 systems, we do not know whether, given this effective description, it would be possible to drive the system to a Néel phase by switching interchain couplings.

3.2. Exact diagonalizations in the XX limit

We repeat the numerical calculations of section 2.2 in the case of the effective model for spin-1/2 doping. As in the case of paramagnetic doping, we only consider the spin anisotropic XX model. The $\langle S_0^z S_R^z \rangle$ correlations are plotted in figure 4 for weak and strong disorder and exhibit a $1/R^2$ power law decay, as for the RS phase. This numerical calculation thus supports the previous argument that the effective one-dimensional model suited for spin-1/2 doping would flow to a massless fixed point as soon as disorder is introduced.

4. Exact diagonalizations of small clusters with paramagnetic doping

We now turn to exact diagonalizations of small clusters at finite temperatures, the interchain coupling being treated in the mean field. Notice that this problem is quite difficult and we cannot go to large sizes because (i) one must calculate *all* the eigenvalues and eigenvectors since we are interested in the finite-temperature behaviour, (ii) one must calculate the staggered field in the mean field, which implies solving for self-consistent equations and (iii) one must average over disorder. This kind of calculation has already been carried out in the context of disordered spin-Peierls systems [11], with results not in contradiction with experiments. We consider an eight-site chain with two paramagnetic impurities, and thus six sites carrying a spin 1 in the squeezed chain. There are three inequivalent ways of putting the impurities on the chain as shown in figure 5. We apply a finite staggered field and calculate the staggered magnetization response for different magnetic fields and temperatures. In practice, the temperature and the staggered field are varied between 0.01 and 2 with an increment of 0.01 and finally the self-consistent staggered field h_s^* is calculated as a function of temperature by imposing the condition $h_s^*(T) = J_{\perp} m_s(T, h_s^*(T))$, where J_{\perp} is the strength of the effective interchain coupling. The self-consistent staggered field is plotted in figure 6 for two values of the effective interchain coupling J_{\perp} . As in the case of the spin-Peierls system, we observe in figure 6 important fluctuations of the mean-field Néel transition temperature. At this point, we conclude that a transition to an AF ordered state should be possible in the strong-disorder regime.

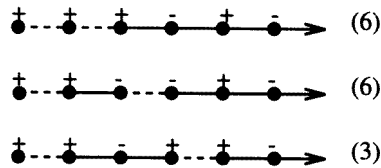


Figure 5. The three inequivalent disorder realizations, represented in the six-site squeezed chain. Arrows denote periodic boundary conditions. A dashed line denotes a weak bond. The sign of the staggered field is indicated by a + or – sign. The degeneracy is indicated in brackets.

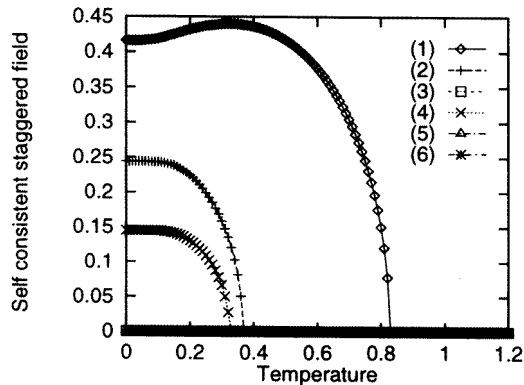


Figure 6. Self-consistent staggered field as a function of temperature calculated for $J_H = 1$, $J_2 = 0.5$ and $J_{\perp} = 0.8$ ((1)–(3)) and $J_{\perp} = 0.4$ ((4)–(6)). (1) and (4), (2) and (5), and (3) and (6) correspond to the three disorder realizations in figure 5.

In order to have an idea of the variations of a macroscopic observable, we plot in figure 7 the variations of the spin susceptibility as a function of temperature for two values of the interchain coupling. We observe that, unlike the case of the spin–Peierls system [11], the emergence of AF as the temperature is lowered does not induce a maximum of the susceptibility, but a jump in the slope of the susceptibility versus temperature.

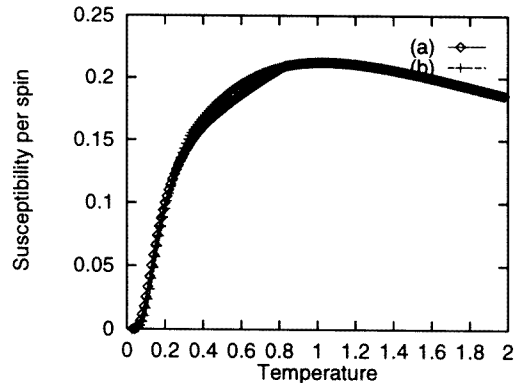


Figure 7. Variations of the uniform susceptibility per spin as a function of temperature for $J_H = 1$, $J_2 = 0.5$ and (a) $J_\perp = 0.8$, (b) $J_\perp = 0.4$.

Moreover, as a comparison between the two different doping mechanisms in a spin-1 chain, we note that eliminating the non-magnetic sites going from the original chain to the squeezed chain has more drastic consequences for the paramagnetic doping than for the spin-1/2 doping. As far as paramagnetic doping is concerned, applying a staggered field in the original chain amounts to applying a staggered field in the *same* direction on both the spin-1/2 moments released by the impurity (on the right and the left of the impurity) and thus to systematically frustrating the AF coupling J_2 across the impurity. Notice that in the case of spin-1/2 doping, this competition does not arise since the spin-1/2 impurity is coupled by AF bonds to the localized effective spin-1/2 moments. Hence, in the case of paramagnetic doping, this effect should act against the establishment of an AF phase.

5. Conclusions

We have thus carried out a detailed investigation of the effect of doping spin-1 chains by paramagnetic and spin-1/2 impurities. Our approach takes advantage of the quasi-one-dimensionality of these systems. We have shown how these systems can be mapped onto bond disordered spin-1/2 chains, the zero-temperature fixed points of which were recently analysed ([19] and [20] for the effective model appropriate to paramagnetic doping, and [24] for spin-1/2 doping). We have pointed out that paramagnetic and spin-1/2 doping are different physical situations since the effective one-dimensional models have a very different physics. Weak paramagnetic doping preserves the Haldane phase [17]. Above a critical disorder strength, the effective one-dimensional model flows to the RS phase. On the basis of our previous experience of spin–Peierls systems [10, 11], we argue that the RS phase, which was found to be the strong-disorder zero-temperature fixed point, would translate in these paramagnetically doped spin-1 chains into a possible AF phase above a critical disorder. Quasi-one-dimensional spin-1 compounds have already been found to be robust against paramagnetic doping [17], but, to the best of the authors’ knowledge,

no AF ordered phase has been found up to now. We have shown by means of exact diagonalizations of small clusters that a strong disorder AF phase is possible. In fact, the disorder favours the enhancement of magnetic fluctuations (as in the case of spin-Peierls compounds). Moreover, we have pointed out that in the paramagnetically doped spin-1 compounds, the interchain coupling always frustrates the AF coupling across the impurity. This mechanism does not exist in the spin-1/2 doped compounds, which in addition flow to a gapless phase for arbitrary weak disorder.

Although the spin-1/2 doping seems at first sight more promising, it is not clear at this stage whether it should display an AF phase or not when interchain coupling is allowed, since the physics of the effective one-dimensional model is dominated by clusters of spins coupled ferromagnetically, or, equivalently, by large spins.

On the other hand, a very difficult problem in the possible observation of an AF phase in the paramagnetic doping case is that the microscopic parameters should be tuned in such way that AF occurs for not too large impurity concentrations, when the compounds become unstable. This means that the AF coupling across the impurity should be as small as possible. Therefore, the issue of the appearance of AF upon doping spin-1 quasi-one-dimensional compounds requires still further experimental investigations.

Finally, even in the absence of AF ordering, neutron scattering experiments should reveal the presence of low-lying magnetic excitations along the chain directions, in both the spin-1 and spin-1/2 doping situations. The zero-temperature fluctuations should be critical for a small spin-1/2 doping. For spin-1 doping, the correlation length should remain finite below a critical amount of disorder, but the correlation length should increase with doping. To our knowledge, such measurements have not been carried out up to now in doped NENP or DMIZ compounds, even though neutron scattering experiments were performed in the doped quasi-one-dimensional transition oxide $Y_2CaBaNi_{1-x}Zn_xO_5$ [25]. However, this compound is not properly described by the type of localized spin models that we have analyzed here. Nonetheless, the results in [25] are interesting in view of our conclusions since the appearance of low-lying AF fluctuations upon doping is reported.

Acknowledgments

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