Role of the magneto-elastic coupling in spin-Peierls systems doped with impurities

P. Hansen[a], D. Augier[b], J. Riera[a,b] and D. Poilblanc[b]

[a] *Instituto de Física Rosario, Consejo Nacional de Investigaciones Científicas y Técnicas y Departamento de Física Universidad Nacional de Rosario, Avenida Pellegrini 250, 2000-Rosario, Argentina*

[b] *Laboratoire de Physique Quantique & Unité mixte de Recherche CNRS 5626 Université Paul Sabatier, 31062 Toulouse, France*

(June 15, 1998)

The role of the magneto-elastic coupling in spin-Peierls chains doped with spin-0 or spin-1 impurities is investigated by exact diagonalization and quantum Monte Carlo simulations. The lattice is treated either classically in the adiabatic approximation or in a fully quantum mechanical calculation. In the case of an isolated chain, strong bonds form next to the impurity, leading in general to the appearance of magneto-elastic solitons. In the case of nonmagnetic impurities, the solitonic excitations do not bind to the impurity. However, the interchain elastic coupling generates an effective attractive potential at the impurity site which can lead to the formation of a bound state.

PACS: 75.10 Jm, 75.40.Mg, 75.50.Ee, 64.70.Kb

Quasi-one dimensional (1D) spin-Peierls systems attract intense experimental and theoretical activity for their fascinating magnetic properties. Such systems usually consist in weakly coupled spin-1/2 Heisenberg chains. Due to spin-phonon coupling, these materials undergo at low temperature a transition towards a phase exhibiting a lattice dimerization and a spin gap [1]. Inorganic compounds like $CuGeO_3$ are easily doped by magnetic or non-magnetic impurities by substituting a fraction of the spin-1/2 Cu^{2+} ions by spin-0 Zn^{2+} or spin-1 Ni^{2+} ions. As shown by magnetic susceptibility measurements [2] and inelastic neutron scattering experiments [3], doping with impurities leads to a rapid collapse of the spin gap. Competition between the spin-Peierls phase and a new antiferromagnetic (AF) phase induced by doping has been established by magnetic susceptibility measurements [2,4], specific heat measurements [5], neutron scattering [6] and NMR experiments [7]. These experiments suggest that magnetic moments and enhanced staggered spin correlations are induced by impurity doping.

From a theoretical point of view, the relevant phonons in dimerized quasi-1D compounds like $CuGeO_3$ or NaV_2O_5 are often considered as three-dimensional, an assumption which, *a priori*, would justify a classical treatment of the lattice. In the dimerized AF Heisenberg chain, a model widely used in the literature to describe these materials, one introduces a fixed dimerization $J(1 \pm \delta)$ of the magnetic exchange integral leading to the opening of a spin gap $\propto \delta^{3/2}$. In $CuGeO_3$, it is believed that magnetic frustration (i.e. AF coupling between next nearest neighbor sites) plays a role. [8,9] The lowest energy excitations of the dimerized Heisenberg chain consist of spinon-spinon bound states [10,11] lying below the two-magnon continuum.

Extensive work on the effect of impurities in dimerized spin chains have been carried out [12–14]. The introduction of vacancies creates finite chains with open boundary conditions (OBC). So far, most treatments of the effects of the boundaries ignore the lattice dynamics [12,13] which, physically, is justified when the elastic coupling to the neighboring chains is large enough. In this case, a finite chain can end by either a "weak" or a "strong" bond depending on the sign of the dimerization on this bond. These two types of boundaries show very different magnetic properties: in contrast to the strong bond edge, a weak bond edge can localize a $S = 1/2$ magnetic excitation. This effect is responsible for the presence of strong AF correlations in the vicinity of weak edges. [15]

In the approach discussed above, the effects of impurities have been considered under the two assumptions that the lattice dimerization is (i) static and (ii) uniform in space. However, due to the magneto-elastic coupling, the presence of a spin-1/2 excitation is expected to, locally, distort the underlying lattice creating an elastic soliton [16]. Such effects were recently investigated in the context of the incommensurate phase of spin-Peierls systems under magnetic field [17,18]. In this work, we investigate the role of the spin-phonon coupling in Heisenberg chains in the vicinity of spin-0 or spin-1 impurities by exact diagonalization (ED) and quantum Monte Carlo (QMC) simulations. The lattice is treated either classically in the adiabatic approximation [17], i.e. allowing for non-uniform static lattice distortions, or in a fully quantum mechanical way [19] introducing, in addition, the lattice dynamics. In an isolated chain, we have found that strong bonds form next to an impurity. For nonmagnetic impurities, solitonic excitations do not bind to the impurity. However, in this case, the interchain elastic coupling generates an effective impurity–soliton attractive potential which leads to the formation of a bound state. The spatial extension of this bound state is governed by the strength of the interchain coupling.

The model we first consider is purely 1D and includes a static, but possibly non-uniform, lattice distortion,

$$\mathcal{H}_{\parallel} = J \sum_i (1+\delta_i) \mathbf{S}_i \cdot \mathbf{S}_{i+1} + \frac{1}{2} K \sum_i \delta_i^2 \,, \tag{1}$$

where the second part corresponds to the elastic energy lost within the chain. The role of the interchain elastic coupling $\mathcal{H}_\perp$ will be discussed later. The classical modulations δ_i living on the bonds have to be determined from a minimization of the total energy. The pure system (which corresponds to periodic boundary conditions, PBC) is dimerized with $\delta_i = \delta(-1)^i$.

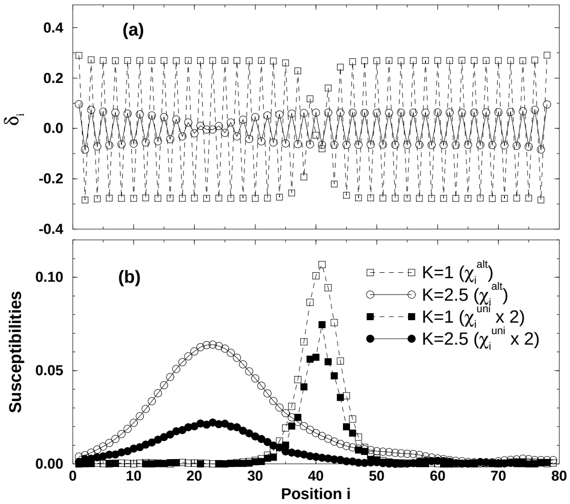

FIG. 1. World Line QMC calculation (at $T = 0.05$) of the modulation δ_i (a) and the local susceptibility χ_i (b) (decomposed into its uniform χ_i^u and alternating χ_i^a components) as a function of the position on a $L = 79$ chain with OBC. Data for $K = 1$ and $K = 2.5$ are shown.

We investigate the effect of spin-0 impurities by considering chains with OBC (open chains). Open chains with an odd number of sites must contain at least a spin-1/2 excitation. Previous QMC simulations supplemented by a self-consistent determination of the equilibrium distortion pattern δ_i (see Ref. [17] for details) have been extended to this new physical situation. Results for a $L = 79$ sites open chain shown in Fig. 1 for various parameters reveal the existence of a single solitonic excitation located away from the chain edges. The zero temperature local susceptibility $\chi_i = \sum_j \langle S_i^Z S_j^Z \rangle$ at site i corresponds physically to the average value of S_i^Z *with respect* to the global orientation of the total S^Z spin component. [20] Fig. 1(b) show that χ_i oscillates rapidly between positive and negative values (large staggered component) and has the largest amplitude of both its uniform and staggered components in the region where the dimer order parameter is suppressed, i.e. around the soliton. This result is quite different to that seen in fixed dimerization calculations [13] where it was observed that spin-1/2 excitations are bound to the chain edge. For increasing K (i.e. for decreasing spin-lattice coupling), the width of the soliton increases and the solitonic pattern continuously evolves into a sinusoidal distortion as expected in the weak coupling limit. It should be stressed that different QMC runs lead to random degenerate equilibrium configurations corresponding to the very same solitonic pattern centered on different sites in a wide area around the center of the chain. However, no changes occur at the two edges of the chains which, systematically, end with a strong bond (i.e. $\delta_i > 0$). This clearly indicates that there exists a short range repulsion between the impurity (edges) and the soliton. One should notice that, independently of the presence of the soliton (i.e. for odd or even chains), $|\delta_i|$ increases in the close vicinity of the edges, in contrast with the analytical results of Ref. [14].

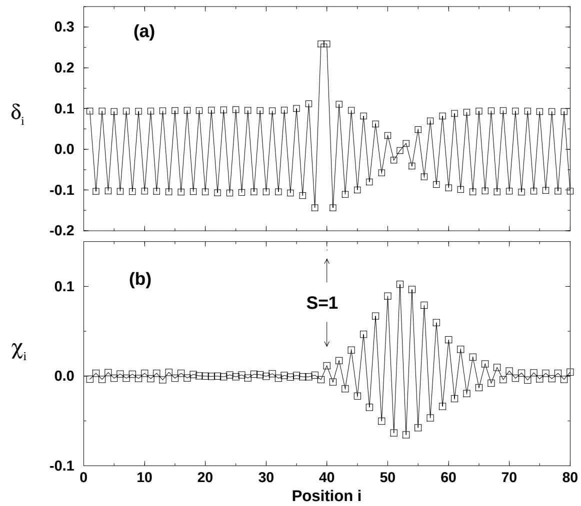

FIG. 2. World Line Monte Carlo calculation (at $T = 0.05$) of the modulation δ_i (a) and the local susceptibility χ_i (b) as a function of the position on a $L = 80$ chain with PBC and a spin-1 impurity at site $i = 40$. Data for $K = 2$ are shown.

The case of a spin-1 impurity has also been considered by assuming, for the sake of simplicity, the same exchange integral and elastic constant on the two bonds on each side of the impurity. Previous calculations [21] assuming a uniform dimerization of the chain have shown that spin-1 impurities lead to more localized states than spin-0 impurities (static vacancies). The QMC results shown in Fig. 2 reveal that the two bonds next to the spin-1 impurity become especially strong indicating that the impurity and the two neighboring S=1/2 spins form an effective spin-0 defect leading qualitatively to the same physics as in the case of a $S = 0$ vacancy. Indeed, the solitonic pattern shown in Fig. 2(b) resembles the ones obtained previously. However, it should be noticed that the new profile is not completely symmetric and that the soliton is always located close to the impurity. This might signal a small attraction in the vicinity of a spin-1 impurity. This might be due to the fact that the three-site system formed by the S = 1 impurity and its two spin S = 1/2 neighbors spends most of the time ($\approx 80\%$) but *not all the time* in the $S = 0$ state leading to a residual impurity-soliton attraction.

In order to investigate the role of the lattice dynamics, next we generalize the previous approach by assuming a coupling to dynamical phonons,

$$\mathcal{H}_\parallel = J \sum_i (1 + g(b_i + b_i^\dagger))\mathbf{S}_i \cdot \mathbf{S}_{i+1} + \Omega\, b_i^\dagger b_i \, . \qquad (2)$$

For sake of simplicity, optical dispersionless modes are considered here. While the adiabatic treatment discussed above is justified in the $\Omega \to 0$ limit, the lattice dynamics can not be neglected when Ω and J becomes comparable. In this case, the lattice modulation can be defined by $\delta_i = g\langle b_i + b_i^\dagger \rangle$. The treatment of the phononic degrees of freedom will relie here on a variational approach [22] which gives accurate results [19]. Previous calculations [19] have shown that the lowest $S = 1/2$ excitations of this model correspond also to massive solitons and antisolitons. Furthermore, soliton and antisoliton do not bind in the strictly 1D case.

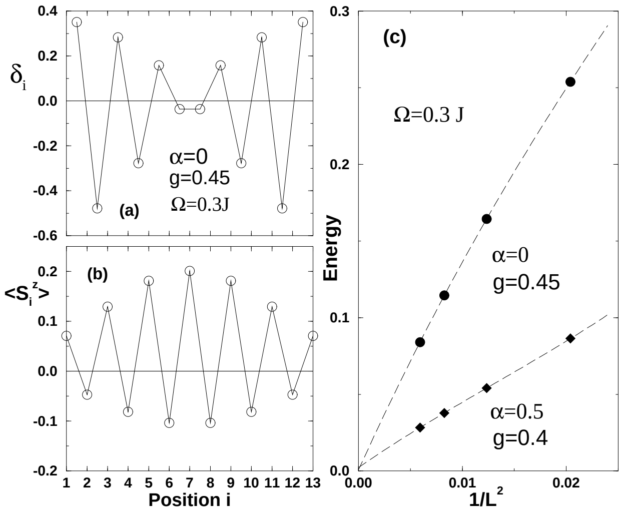

FIG. 3. Modulation δ_i (a) and spin density $\langle S_i^Z \rangle$ (b) in the GS of a $L = 13$ sites open Heisenberg chain coupled to dynamical phonons. Parameters are shown on the plot. (c) Finite size scaling of the soliton-impurity binding energy in the case of a vacancy (as indicated on the plot).

The results obtained by ED for an open chain with an odd number of sites are shown in Fig. 3. The lattice modulation pattern and the spatial variation of the spin polarization are very similar to the results obtained in the adiabatic treatment of the lattice. In particular, strong bonds ($\delta_i > 0$) also form at the chain ends and a soliton appears in the middle of the chain.

In order to get information on the effective interaction between a soliton and the chain ends, it is instructive to define the soliton-impurity binding energy on a $L = 2p+1$ chain as $E_B(L) = E_{IS}(L) - E_0^*(L) - e_S - e_I$, where $E_{IS}(L)$ is the ground state energy of an L-site chain with an impurity (e.g. with OBC for a $S = 0$ impurity), $E_0^*(L)$ is the energy of the pure system obtained for even number of sites and interpolated to L, and e_S (e_I) is the extrapolated soliton (impurity) energy (see Ref. [19]). The finite size scaling of E_B is shown in Fig. 3 and reveals no binding in the thermodynamic limit. The conclusion is also similar when a finite magnetic frustration $\alpha = J_2/J \neq 0$ is considered (in that case, a term $\mathcal{H}_F = J_2 \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+2}$ is added to Hamiltonian (2)). Preliminary analogous results for the case of S = 1 impurities indicate a nonzero binding between the impurity and the soliton. [23] The absence of impurity-soliton bound states for S = 0 impurities and the possible existence of binding for S = 1 impurities are consistent with the previous results obtained in the adiabatic treatment of the lattice.

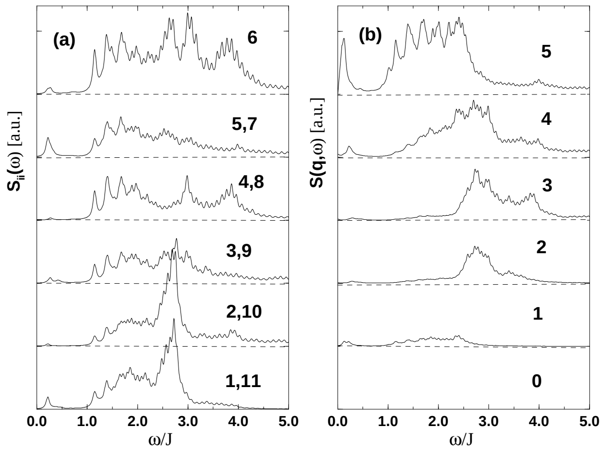

FIG. 4. (a) Local dynamical spin structure factor $S_{ii}(\omega)$ calculated on the various sites (indicated on the plot) of a $L = 11$ spin chain coupled to dynamical phonons. (b) Dynamical spin structure factor $S(q,\omega)$ for the same system as (a) (q in units of $2\pi/L$). Parameters are as in Fig. 3.

The local dynamical spin-spin correlation function is shown in Fig. 4(a). The reminiscence of the spin gap of the chain with PBC is clearly seen at an energy $\omega \sim J$. However, spectral weight appears at much lower energy. It can be attributed to the soliton excitation which behaves as a $S = 1/2$ object weakly connected to the rest of the system, consistently with the behavior discussed above with respect to Fig. 1. If one labels the sites from 1 to L starting from the left end of the chain, we observe a large low energy weight at the "odd" positions, $i = 2k+1$ due to the dimerization pattern, with the largest peak at the closest site to the center. In fact, this can be qualitatively understood by assuming that the (free) soliton can move by hopping "over" a strong bond. This feature also manifests itself in the low energy peak in $S(\mathbf{q},\omega)$ near $\mathbf{q} = \pi$ as it can be seen in Fig. 4(b). This is similar to what has been observed in fixed dimerization calculations. [12] The remnants of high energy branch of the pure system can be still seen.

Lastly, we investigate the role of a realistic interchain coupling $\mathcal{H}_\perp$. To illustrate the role of $\mathcal{H}_\perp$, let us consider the physical situation of a finite chain cut by two spin-0 impurities at its ends and immersed in the bulk. In the dimerized phase, the neighboring chains produce a $q = \pi$ potential of the form, $\mathcal{H}_\perp = K^\perp \sum_i \delta_i \delta_i^{ext}$. Here the modulation of the neighboring chains is treated in the mean-field approximation, i.e. $\delta_i^{ext} = (-1)^i \delta_0$, but the full spatial dependence in the chain with the impurities is retained. The amplitude of the external potential

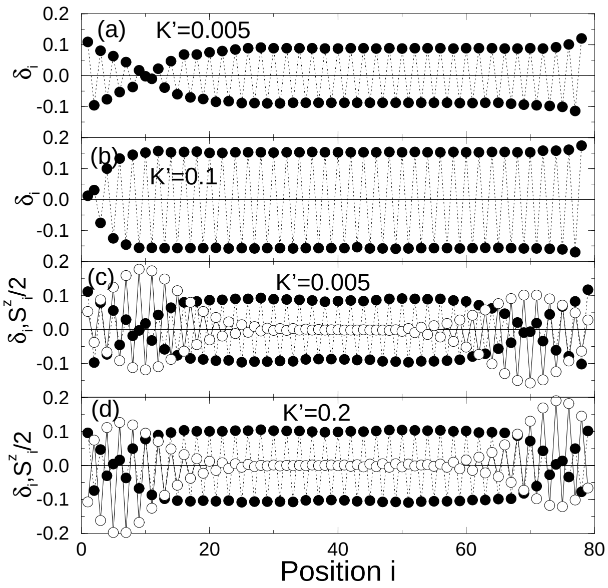

FIG. 5. (a), (b): QMC calculation (at $T = 0.05$) of the modulation δ_i vs i on a $L = 79$ chain with OBC coupled to a small external dimerization. (c), (d): the same for a $L = 80$ chain. In this case S^z_i vs i is also shown.

is then related to the elastic constant $K_\perp$ between the chains by $K' = K_\perp \delta_0$. In the case of finite chains with an odd number of sites, the external potential tends to form a weak bond on, e.g., the left end and a strong bond on the right end. Therefore, the soliton, which, according to the previous study, is free for vanishing K', will experience a confining force proportional to its separation from the left end. This attractive potential originates physically from the misfit between the dimerization pattern on the left side of the soliton and the dimerization pattern of the bulk. Our numerical calculations shown in Fig. 5 confirm this intuitive picture. As seen in Fig. 5(a), a very small coupling K' can suddenly push the soliton towards the left end and the equilibrium position is obtained when the small confining potential is equilibrated by the short range repulsive potential created by the impurity. For larger and larger K', the confining potential becomes steeper and steeper and, eventually, the soliton moves completely to the left end (Fig. 5(b)). The case of an open chain with an even number of sites is also particularly interesting (Fig. 5(c),(d)). If the external potential is out-of-phase with the open chain dimerization (i.e. δ_i for $K' = 0$ and δ_i^{ext} have opposite signs), the external potential will lead to the formation of a soliton-antisoliton pair in the center of the chain. For increasing K', the two $S = \pm 1/2$ excitations migrate towards the chain ends forming two localized excitations. The excess of S^z at the ends of the chain are fluctuating at low temperature between $+1/2$ and $-1/2$. These calculations strongly support the fact that soliton-impurity bound states are stabilized by the interchain elastic coupling. [24]

To summarize, the effects of impurities in spin-Peierls systems have been studied by numerical methods. In contrast to previous approaches, local non-uniform deformations of the lattice have been considered and major differences with respect to the case of a uniform dimerization have been outlined. In particular, by computing the impurity susceptibility and $S^z(i)$, we have observed that the spin-1/2 excitations are bound to the solitons. Besides, we have shown that, for a strictly 1D model the solitonic excitations do not bind to the nonmagnetic impurities. We thank IDRIS (Orsay) for allocation of CPU time on the CRAY supercomputers.

[1] M. Hase, I. Terasaki and K. Uchinokura, Phys. Rev. Lett. **70** 3651 (1993).
[2] M. Hase *et al.*, Phys. Rev. Lett. **71**, 4059 (1993).
[3] J. G. Lussier, S. M. Coad, D. F. McMorrow and D. McK. Paul, J. Phys. Cond. Matt. **7**, L325 (1995).
[4] T. Matsuda *et al.*, Phys. Rev. Lett. **80**, 18 May (1998); K. Kanabe *et al.*, preprint cond-mat/9805072.
[5] S. B. Oseroff *et al.*, Phys. Rev. Lett. **74**, 1450 (1995).
[6] L.-P. Regnault *et al.*, Phys. Rev. B **53**, 5579 (1996).
[7] J.-P. Renard *et al.*, Europhys. Lett. **32**, 579 (1995).
[8] J. Riera and A. Dobry, Phys. Rev. B **51**, 16098 (1995).
[9] G. Castilla, S. Chakravarty and V.J. Emery, Phys. Rev. Lett. **75**, 1823 (1995).
[10] G. S. Uhrig and H. J. Schulz, Phys. Rev. B **54**, R9624 (1996); A. Fledderjohann and C. Gros, Europhys. Lett. **37**, 189 (1997); D. Augier, D. Poilblanc, S. Haas, A. Delia, E. Dagotto, Phys. Rev. B **56**, R5732 (1997).
[11] E. Sorensen, I. A. Affleck, D. Augier and D. Poilblanc, preprint (1998).
[12] G. B. Martins, E. Dagotto and J. Riera, Phys. Rev. B **54**, 16032 (1996).
[13] M. Laukamp et al., Phys. Rev. B **57**, 10755 (1998).
[14] H. Fukuyama, T. Tanimoto, and M. Saito, J. Phys. Soc. Jpn. **65**, 1182 (1996).
[15] M. Fabrizio and R. Mélin, Phys. Rev. B **56**, 5996 (1997).
[16] S. A. Brazovskii, S. A. Gordynin and N. N. Kirova, JETP Lett. **31**, 456 (1980); M. Fujita and K. Machida, J. Phys. Soc. Jpn. **53**, 4395 (1984).
[17] A. E. Feiguin, J. A. Riera, A. Dobry, and H. A. Ceccatto, Phys. Rev. B **56**, 14 607 (1997); F. Schönfeld, G. Bouzerar, G.S. Uhrig and E. Müller-Hartmann, preprint cond-mat/9803084.
[18] For related experiments see e.g., V. Kiryukhin *et al.*, Phys. Rev. Lett. **76**, 4608 (1996).
[19] D. Augier, D. Poilblanc, E. Sorensen and I. Affleck, preprint cond-mat/9802053.
[20] S. Eggert and I. Affleck, Phys. Rev. Lett. **75**, 934 (1995).
[21] P. M. Hansen, J. A. Riera, A. D'Elia and E. Dagotto, preprint cond-mat/9711229.
[22] R. Fehrenbacher, Phys. Rev. Lett. **77**, 2288 (1996).
[23] D. Augier *et al.*, in preparation.
[24] D. Khomskii, W. Geertsma, and M. Mostovoy, Czech. J. Phys., **46**, Suppl. S6, 3239 (1996).