Li Induced Spin and Charge Excitations in a Spin Ladder

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A lithium dopant in a cuprate spin ladder acts as a vacant (non-magnetic) site accompanied by an extra hole bound by a Coulomb potential. We find that, although the undoped ladder spin gap is not essentially altered by Li doping, a dopant-magnon bound state appears within the gap. Contrary to the case of Zn-doped ladders, we predict that, in the Li-doped ladder, the spin liquid character is very robust against antiferromagnetism. We also predict the spatial dependence of the density of states in the vicinity of the dopant which could be measured by local spectroscopic probes.

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Impurity doping of the high-Tc cuprate superconductors is an effective tool for exploring the low temperature physics of these strongly correlated electronic systems. The similarities or differences observed upon doping non-magnetic zinc (Zn) and lithium (Li) ions must find their explanations both in the strongly correlated nature of their host and in the peculiarities of each dopant. For example, when doped into the antiferromagnetic (AF) phase of La2CuO4, Li is far more effective at suppressing the Néel temperature than Zn, although both enter the same planar Cu(2) site. However, Li, by releasing an extra hole (due to the difference of formal valence of Cu2+ and Li+) can theoretically lead to an essential perturbation while Zn was shown to only slightly enhance the AF correlations in its vicinity.

Strikingly, despite different behavior in AF systems, non-magnetic Zn and Li ions behave quite similarly in conducting YBa2Cu3O6+x (YBCO) by inducing local magnetic moments. This suggests that, in the case of Li, the extra hole is not bound, unlike La2Cu1-xLi1.xO4. The induced moment sits predominantly on the four nearest neighbor (NN) Cu of the dopant and exhibits static and dynamic susceptibilities reminiscent of a Kondo-like behavior with a large range of Kondo temperatures. In fact, screening of the induced moment by pairing was also investigated. Possible reduction of screening due to pairing was also investigated.

Spin ladders offer a perfect system to investigate impurity doping in a spin liquid or Resonating Valence Bond (RVB) state, i.e. a state with short AF correlations and a spin gap. Clearly, a reliable microscopic description of dopants in ladders would greatly contribute to the understanding of their two dimensional (2D) analogs as well. In fact, Zn doping into the spin-1/2 Heisenberg two-leg ladder SrCu2O3 leads to local moments (as in metallic YBCO) and to an instability of the gapped spin-liquid state towards an AF ordered state at low enough temperature. Formation of local moments as well as a rapid suppression of the spin gap was obtained theoretically in a Heisenberg ladder using the vacant-site impurity model. Further numerical simulations have led to an effective model of local moments interacting via an exchange interaction which rapidly decays with the impurity distance.

Doping Li into a two-leg spin ladder is still an open and fascinating problem both experimentally and theoretically. Novel physics can be expected due to the additional (w.r.t the case of Zn) intrinsic doping associated with the substitution of a divalent (Cu2+) by a monovalent (Li+) ion. In this Letter, we address this issue using a specific vacant-site model for Li+ embedded into a correlated host. We show that Li doping does not introduce low-energy spin excitations as in the case of Zn but instead leads to the formation of a magnon-impurity...

FIG. 1. Schematic representations of a Li-doped spin ladder. The cross (resp. the circle) stands for the impurity site (resp. injected hole) and the thick lines correspond to the attractive potential. (a) Singlet GS where spins are paired up into singlet spinlets (shown as shaded areas). Sites are labelled for convenience. (b) Lowest triplet excitation: dopant-magnon bound state.
bound state (BS) just below the spin gap of the undoped ladder. This suggests that, unlike Zn-doped ladders, Li-doped ladders would keep a robust spin liquid character at low temperature.

For dilute concentrations, a single dopant in the geometry of Figs. 1 suffices. We model the substitution of a Cu$^{2+}$ ion by a Li$^+$ atom by a vacant inert site which is a hard-core potential for holes. Because of the different valences of Cu$^{2+}$ and Li$^+$ a hole is injected into the spin ladder and it feels a static attractive Coulomb potential centered on the Li$^+$ site. A realistic Hamiltonian reads:

$$H = J \sum_{<i,j>} (S_i \cdot S_j - \frac{1}{4} n_i n_j)$$

$$-t \sum_{<i,j>,\sigma} (c_{i,\sigma}^\dagger c_{j,\sigma} + \text{h.c.}) - V \sum_{l_i} (1 - n_i),$$

where the notations are standard and the prime means that the sum over NN links $<ij>$ is restricted to the bonds not connected to the dopant site. We have modeled the attractive hole potential by a static NN potential $-V$ as depicted on Figs. 1. The summation over $l_i$ only runs over the NN sites of the impurity. Note that, for simplicity, we restrict ourselves to the case of a magnetically isotropic ladder i.e. with equal rung and leg couplings, $J$. Numerical results discussed in the following are obtained by Exact Diagonalisations (ED) of small periodic ladder rings (up to $2 \times 12$) supplemented by Density Matrix Renormalisation Group (DMRG) calculations on larger open systems (up to $2 \times 64$). In open ladders, the impurity site is placed in the center.

First, we investigate the localization of the injected hole vs the strength of the Coulomb potential. Following Ref. [1], we calculate the so-called hole-dopant binding energy defined as,

$$\Delta_{1,\text{imp}, 1 h}^{S = 0} = E_0(1h, 1i) + E_0(0h, 0i) - (E_0(1h, 0i) + E_0(0h, 1i)),$$

where $E_0(nh, mi)$ is the GS energy of a ladder with $n = 0$ or 1 ($m = 0$ or 1) holes (dopants). This quantity is negative when the hole and Li ion form a stable bound state. Since Li-doping results in the simultaneous removal of two spins we expect a magnetically inert state i.e. a singlet ($S = 0$). As seen on Fig. 2(a) a stable BS is found for almost all couplings, even in the case $V = 0$. It should be noticed that the magnitude of the binding energy is slightly larger than in the case of a 2D planar geometry. Fig. 2(a) offers a pictorial representation of such a state where the absence of any local moment can be clearly understood from the RVB nature of the host (all remaining spins are paired up in singlets). Although, our calculation shows that a single Li-dopant binds the injected hole in its vicinity, caution is required at a finite concentration. In this case the possibility of other “decay channels” has to be considered, e.g. 2 holes from 2 dopants recombining into an itinerant hole pair [15]. This possibility is ruled out by the fact that the dopant-hole binding energy is always smaller than half of the hole-pair binding energy $\Delta_{2,\text{holes}}$ calculated on an undoped ladder as seen on Fig. 2(a), even when $V = 0$. At low concentrations we then expect decoupled quenched dopants each binding one hole. Since the spatial extensions of the isolated BS’s are quite small (typically $\xi = 2$ to 4 rungs even when $V = 0$), the system is expected to remain insulating up to large doping.

We now turn to the magnetic properties of the Li-doped spin ladder in order to compare with Zn doping. For this purpose, we compute the dynamical spin structure factor $S(q, \omega)$ as measured in Inelastic Neutron Scattering (INS) experiments for an undoped ladder (shown on Fig. 3(a)), a Zn-doped ladder (modeling Zn as a neutral vacant site and shown on Fig. 3(b)) and a Li-doped ladder described by Eq. (1) (shown on Fig. 3(c)). Note that $S(q, \omega)$ quantifies the spectral weight given by the matrix element $|\langle n | S^z_q | 0 \rangle|^2$ between the singlet GS $|0\rangle$ and the magnetic excitation $|n\rangle$ at energy $\omega$. The RVB picture, in which spins are paired up into short range singlets (for example on the rungs as in Figs. 3), turns out to give a nice qualitative understanding of the magnetic properties. In the undoped ladder, a triplet excitation (magnon) is well described by exciting a rung singlet into a triplet. Fig. 3(a) clearly shows the single magnon dispersion with a minimum at $q = (\pi, \pi)$ which defines the spin gap $\Delta_0$ of the undoped ladder. Introducing a Zn atom on a rung releases a free spin-1/2 which leads to zero-energy spin fluctuations predominantly at $q = (\pi, \pi)$ as seen in Fig. 3(b) while the undoped ladder magnon mode still survives. When two Zn dopants are
introduced, our data, in agreement with Ref. [14], show that the two local $S=1/2$ moments experience a weak effective exchange interaction, $J_{\text{eff}}$, which decays rapidly with separation. In that case, a small spectral weight at $q=(\pi, \pi)$ and low energy $\omega \sim J_{\text{eff}}$ below the undoped spin gap is seen. The case of Li-doping shown on Fig. 3(c) is drastically different with no weight at small energy. Indeed, since a Li$^+$ dopant introduces simultaneously a bound hole, it does not release a free spin as for Zn but rather produces a new type of excitation located just below the unperturbed spin gap, namely a collective bound state of a (let us say rung) magnon with the hole and the Li ion as naively depicted on Fig. 2(b). Its binding energy defined as the energy separation w.r.t. the free magnon energy $\Delta_0$, remains in general quite small (in absolute value) as seen directly in Fig. 3(b) (and qualitatively in Fig. 3(c)). Therefore a drastic reduction of the spin gap does not occur in this case.

The physical origin of this BS is of particular interest. Simple qualitative arguments as well as extensive numerical data show that the origin of binding in this channel can be attributed to the hole kinetic energy gain associated with the spin polarization. DMRG calculations show that the spatial extents of the charge and spin perturbations associated with the BS are quite different. While the hole is localized on a scale of a few rungs from the dopant as seen in Fig. 3(a), the distribution of the rung magnetization can extend to large distances as seen in Fig. 3(b). This is in agreement with our other finding that the energy costs (i.e. the absolute value of the related binding energies $\Delta_0$) of the two virtual decay processes leading either to a free hole and a neutral dopant-magnon complex or to a free magnon and a dopant-hole complex are quite different in magnitude as seen in Figs. 2(b) and 2(c). In fact, the attraction $-V$ has the opposite effect on the two quantities; while increasing $V$ binds the hole more strongly, it also limits the ability of the hole to reduce its kinetic energy by moving in the spin polarized background of the magnon, and so the binding energy of the magnon to the dopant-hole complex is reduced [14]. From the analysis of the behavior of the magnetic size of the BS (deduced from Fig. 2(b)) or alternatively directly from its binding energy plotted on Fig. 2(c), we conclude that above a critical value of $V$ (typically one gets $V_C/t\simeq 2$ for $J/t=0.5$) the magnon escapes from the dopant.

For completeness, we calculate also the local density of state (DOS) in the vicinity of the dopant. Since this DOS might be accessible experimentally by Scanning Tunneling Spectroscopy it can test the validity of the model. Results are shown in Fig. 3 corresponding to spatially resolved DOS spectra in the vicinity of the dopant. The $\omega > 0$ ($\omega < 0$) spectra give the weights of the neutral (charged) target $S=1/2$ states which are accessible by removing the hole (adding an extra hole) in the singlet GS. The large peak seen at vanishing positive energy on site number 1 i.e. on the same rung as the dopant corresponds to a resonant magnetic spinon-dopant BS. Other spinon-dopant resonances of higher energies are also seen with larger spatial ranges i.e. spread over two or three sites of the leg opposite to the dopant. Similarly, $\omega < 0$ resonances are seen when adding an extra hole on the same leg as the dopant e.g on sites number 3 and 5. The lowest resonance energy (in absolute value) is obtained.

![Figure 3](image3.png)

**FIG. 3.** Spin structure factors $S(q, \omega)$ calculated on 2x10 ladders. Data for $q_z=\pi$ only are shown. From bottom to top, the different curves correspond to decreasing $q_{\perp}$ momenta, from $q=(\pi,\pi)$ (bottom) to $q=(0,\pi)$ (top). For clarity, reducing scaling factors (as indicated) are applied on some curves. (a) Undoped periodic ladder; (b) Spin ladder doped with a single Zn impurity (full line) or two Zn impurities separated by the maximum distance on the same leg (dashed line); (c) Ladder doped with a single Li impurity with $t=2J$ and $V/t=0.5$.

![Figure 4](image4.png)

**FIG. 4.** Hole rung density (a) and $S_z$ rung density (b) along the ladder direction in the lowest energy triplet state calculated by DMRG for $J/t=0.5$. The rung density is defined as the algebraic sum of the densities (if any) on the two sites of a given rung. Different values of $V/t$ (as indicated) are shown.
the dopant. Note also that the local density of states around a Li dopant should ensure that a bound hole in the latter case. Experiments to test these predictions are proposed.

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