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Origin of spin incommensurability in hole-doped $S=1$ $Y_{2-x}Ca_xBaNiO_5$ chains

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Spin incommensurability (IC) has been recently experimentally discovered in the hole-doped Ni-oxide chain compound $Y_{2-x}Ca_xBaNiO_5$ [G. Xu *et al.*, Science **289**, 419 (2000)]. Here a two orbital model for this material is studied using computational techniques. Spin IC is observed in a wide range of densities and couplings. The phenomenon originates in antiferromagnetic correlations ''across holes'' dynamically generated to improve hole movement, as it occurs in the one-dimensional Hubbard model and in recent studies of the two-dimensional extended t-J model. The close proximity of ferromagnetic and phase-separated states in parameter space is also discussed.

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One of the most remarkable results in hole-doped two-dimensional (2D) high-temperature superconductors is the existence in neutron-scattering experiments of spin incommensurability (IC).[1] This result is compatible with the formation of stripes, with a π shift in the antiferromagnetic (AF) order across the stripes. Considerable theoretical work has been devoted to the explanation of these structures, but no consensus has been reached. Recently, a phenomenon analogous to the spin IC in cuprates has been reported in the one-dimensional (1D) compound $Y_{2-x}Ca_xBaNiO_5$.[2] The parent material ($x=0$) is a spin-one ($S=1$) chain described by a Heisenberg model, with a spin liquid ground state. Upon doping, the resistivity is drastically reduced,[3] and the magnetic IC arises.[2] These novel results raise the interesting possibility of a common origin of the spin IC phenomena observed in doped 2D cuprates and 1D nickelates.

The purpose of this paper is to present theoretical calculations searching for spin IC in models for doped $S=1$ chains. Theoretical studies of these systems have been presented before,[4,5] but the magnetic order upon doping has not been investigated in detail. The main result of our effort is that spin IC can indeed be observed when holes are doped into a spin-gapped $S=1$ chain. The effect originates in the local existence of dynamically induced robust AF correlations between the spins located at both sides of a mobile hole (''across-the-hole''). This spin structure is generated to facilitate the hole movement, namely to improve the kinetic energy portion of the Hamiltonian. This is qualitatively similar to results reported in 1D Hubbard models with spin-charge separation,[6] and also in studies by Martins *et al.* on ladders and 2D models with very mobile holes.[7] Based on the results reported here, it is natural to conjecture that the spin IC in 1D Ni oxides may originate in similar tendencies of these systems toward spin-charge separation. In the 1D $S=1/2$ system this separation is complete, while in 2D $S=1/2$ and 1D $S=1$ spin-gapped systems it appears to occur only at short distances. However, this is sufficient to induce a robust spin IC.

The proposed Hamiltonian for doped $S=1$ chains is

$$H=-\sum_{\langle ij\rangle ab\sigma} t_{ab}(c^{\dagger}_{ia\sigma}c_{jb\sigma}+\text{H.c.})+U\sum_{ia} n_{ia\uparrow}n_{ia\downarrow} + U'\sum_{i\sigma\sigma'} n_{i1\sigma}n_{i2\sigma'}-J\sum_{i\sigma\sigma'} c^{\dagger}_{i1\sigma}c_{i1\sigma'}c^{\dagger}_{i2\sigma'}c_{i2\sigma}, \quad (1)$$

where the notation is standard, namely $n_{ia\sigma}$ is the number operator at site i, orbital a, and spin σ, $a=1(2)$ corresponds to orbital $3d(3z^2-r^2)$ $[3d(x^2-y^2)]$ of the Ni^{+2} ion, and the chain direction is taken to be the z axis. The hopping amplitudes between nearest-neighbor sites are $t_{11}=4/3$, $t_{22}=t_{12}=t_{21}=0$.[8] U and U' are onsite intraorbital and interorbital Coulomb interactions, respectively, while $J(>0)$ is the Hund coupling which favors parallel spins on the same site. Note that orbital rotational invariance requires $U=U'+J$.[9,10] For simplicity, in this first attempt to understand the spin IC effect the oxygen ions are not explicitly included in the Hamiltonian. The use of Zhang-Rice ''doublets'' (instead of singlets) in previous literature of Ni oxides[5,11] justifies in part this approximation.

The many-body technique used here to analyze ground-state properties of Hamiltonian (1) is the density-matrix renormalization group (DMRG) method.[12] The finite-system variation of DMRG was applied, working with open boundary conditions (OBC). Truncation errors were kept around 10^{-5} or smaller, using about 100 states. In order to characterize ground-state properties, a variety of observables have been measured, such as the spin structure factor defined as

$$S(k)=\frac{1}{L}\sum_{jmab}\langle \mathbf{S}_{ja}\cdot\mathbf{S}_{mb}\rangle e^{i(j-m)k}, \quad (2)$$

where $\mathbf{S}_{ja}=\Sigma_{\alpha\beta}c^{\dagger}_{ja\alpha}\sigma_{\alpha\beta}c_{ja\beta}$, $\langle\ \rangle$ denotes ground-state expectation value, and L is the length of the chain. The charge structure-factor was also measured, but no evidence of charge ordering has been observed. Also the local density, $\langle n_i\rangle=\Sigma_{a\sigma}\langle n_{ia\sigma}\rangle$, and total-spin z component, $\langle \mathbf{S}^z_j\rangle=\Sigma_a\langle \mathbf{S}^z_{ja}\rangle$, were studied, but the main results were obtained focusing on spin correlations.

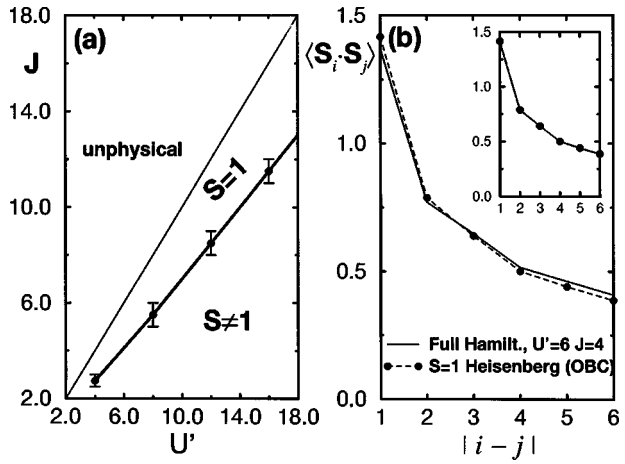

FIG. 1. Results at $n=2$ for a 16-site chain. (a) Phase diagram. (b) Spin correlations at $(U',J)=(6,4)$. For comparison, the correlations of the AF $S=1$ Heisenberg chain are also shown. The inset shows the same but for $(U',J)=(16,13)$. Details can be found in the text.

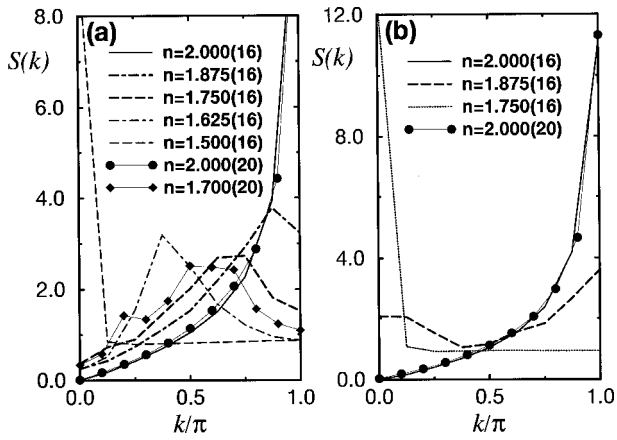

FIG. 2. The spin structure factor for systems of 16 and 20 sites (indicated in parentheses) and different electron densities: (a) $U'=6$ and $J=4$; (b) $U'=16$ and $J=13$.

Let us consider density $n=2$ first, expected to represent the undoped material. Figure 1(a) shows the (U',J) phase diagram for a system of 16 sites and 32 electrons. Each of the three marked regions have distinct features. In the regime above line $J=U'$ the Hund coupling J provides an electronic attractive interaction which competes with the U' repulsion. In order to have the expected overall repulsive total interorbital interaction, $J<U'$ is required.[9] In addition, to create a spin-1 chain, as observed in experiments, a robust Hund coupling is needed. In fact, inside the region labeled $\mathbf{S}=\mathbf{1}$ it was observed that the total spin at site j, defined as $\mathbf{S}_j=\mathbf{S}_{j1}+\mathbf{S}_{j2}$, is to an excellent approximation equal to one[13] for any site j. This is the region of interest in the present problem. In the regime labeled $\mathbf{S}\neq\mathbf{1}$, the mean spin is less than 1 since J is not sufficiently strong.[14] The frontier $\mathbf{S}=\mathbf{1}$–$\mathbf{S}\neq\mathbf{1}$ has error bars caused by finite-size effects, estimated from chains with $L=8$, 16, and 20 sites.

In the region of local $\mathbf{S}=\mathbf{1}$ states it is necessary to verify whether a Heisenberg spin-1 chain provides a proper effective model for the system. For this purpose, ground-state spin correlations were calculated for several couplings inside the $\mathbf{S}=\mathbf{1}$ region. As shown in Fig. 1(b), these correlations behave quite similarly for two typical sets of couplings (U',J), and in addition they are nearly identical to those arising from the much simpler spin-1 Heisenberg chain model. In both cases, the results at a given distance were averaged over the entire chain to improve the convergence to the thermodynamic limit when different OBC system sizes are used.

Inside the $\mathbf{S}=\mathbf{1}$ region, it can be shown that not only the ground-state properties, but also the low-energy physics of Eq. (1) reduces to the spin-one Heisenberg chain, with an effective antiferromagnetic coupling J_{Heis} as an overall scale, which depends on U' and J. This interesting result confirms that our model at $n=2$ is realistic. In fact, for several values of (U',J) and several lattice sizes, the effective Heisenberg coupling J_{Heis} was estimated by comparing energy gaps of Eq. (1) with those of the spin-1 Heisenberg chain. For given values of (U',J), data coming from systems with sizes between 4 and 20 sites yield approximately the same value for J_{Heis}. Even though a closed form for the dependence of J_{Heis} with couplings is unknown, it is clear from our results that J_{Heis} is inversely proportional to U' and J. Typical values of J_{Heis} are 0.14 ± 0.01 for $(U',J)=(4,3)$, 0.042 ± 0.002 for $(U',J)=(16,13)$, and 0.019 ± 0.002 for $(U',J)=(32,28)$, showing that the expected spin-gaps are very small in the units that are natural for Hamiltonian Eq. (1). Reducing J at fixed U', the $\mathbf{S}\neq\mathbf{1}$ region is accessed where the on-site $S=1$ and $S=0$ states start mixing, producing deviations in the spin correlations from those of the Heisenberg model.

The introduction of mobile holes in the $\mathbf{S}=\mathbf{1}$ region, through the reduction of the electronic density, produces the most interesting results of our investigations. In the regime of moderate couplings, both U' and J less than 10, the presence of *spin incommensurate* tendencies are clearly observed [Fig. 2(a)], in a wide range of densities, through the appearance of a prominent peak at a momentum different from π. No fine tuning of parameters is needed, the effect exists in a wide range of couplings.[15] Note that although experiments[3] suggest that the material $Y_{2-x}Ca_xBaNiO_5$ remains insulating upon doping, the observed drastic change in the resistivity[3] justifies the use of a metallic description of the compound, as observed in our studies where charge is not localized.[16] It may occur that holes in the real material are mobile over several lattice spacings, but defects destroy the complete metallicity of the samples (see also Ito *et al.*[17]).

Insight into the origin of the spin IC can be gained by investigating spin correlations in the vicinity of the holes. This can be obtained by projecting the hole on a giving site out of the ground state, and then measuring spin correlations around and across that hole. This projection procedure has been successfully previously applied in studies of models for the cuprates.[7,18] Figure 3 shows a pictorial representation of a 16 sites system doped with one, two, and four holes.[19] The holes were projected onto their most probable locations on the chain (remember that OBC are used). Since hopping is restricted within the $3d(3z^2-r^2)$ orbitals, the projection is

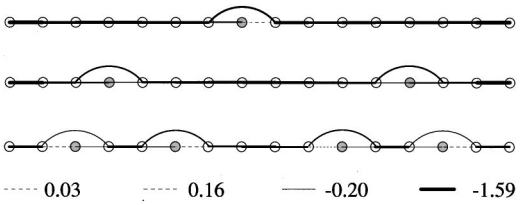

FIG. 3. Spin correlations for one ($n=1.9375$), two ($n=1.875$), and four ($n=1.75$) holes, doped on a 16 sites system at $n=2$. Gray circles indicate the sites where the holes were projected at orbital 1. The thickness of the lines is proportional to the spin bond strengths, according to the scale shown. Full (dashed) lines indicate AF (FM) bonds. The couplings are $(U',J)=(4,3)$. In all cases shown, across-the-hole bonds are AF and $S(k)$ has spin IC, as in Fig. 2(a).

always carried out onto orbital 1. After the projection, the spin bond strengths $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle$ were measured for i and j being nearest-neighbor sites, or sites adjacent to the same hole. The bond strength is proportional to the thickness of the line connecting the sites in Fig. 3. In that figure, the AF $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle < 0$ (FM $\langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle > 0$) bonds are represented by a full (dashed) line.

The results in Fig. 3 were obtained for $(U',J)=(4,3)$, but they are representative of tendencies found in most cases studied here, for small and moderate couplings. Across the projected holes, the clear presence of AF bonds is observed. It is this π shift in the staggered spin pattern that causes the incommensurability. Intuitively these π shifts improve the mobility of holes. Without them, the hole movement would lead to the creation of ''strings'' of ferromagnetic bonds, increasing the energy. This across-the-hole structure is reminiscent of those observed in models for cuprates in 1D and 2D,[6,7] with the conceptual difference that the present results exist in the spin-liquid regime (nonzero spin gap). It appears that the local tendency to produce spin IC, in the form of across-the-hole AF correlations, is present regardless of the spin-liquid vs critical or long-range ordered character of the spin background. Note that in fact for a $S=1$ chain it is possible to use the concept of ''AF order'' in the spin background as long as the distances considered are smaller than the AF correlation length, which for the $S=1$ Heisenberg chain is about six lattice spacings. This is sufficiently large to accommodate the small across-the-hole structure observed in our studies. This also suggests that even if holes are localized in $Y_{2-x}Ca_xBaNiO_5$, with a distorted region of only a few lattice spacings,[2,17] it is still possible for holes to generate spin IC.

The results shown in Fig. 2(a) are representative of data gathered at several couplings, and they not only contain information on spin IC, but on other nontrivial spin patterns, as well. In particular, it was observed that for a fixed U', the spin IC survives longer the decrease in electronic density by reducing J, but eventually a *ferromagnetic* (FM) regime always appears if the hole doping is large enough, as shown in Fig. 2(a) where a prominent peak at zero momentum is observed at density 1.5. Then, the phase diagram of models for doped $S=1$ chains must contain a FM phase at low enough electronic density. The existence of a competing FM phase is in agreement with previous calculations using effective

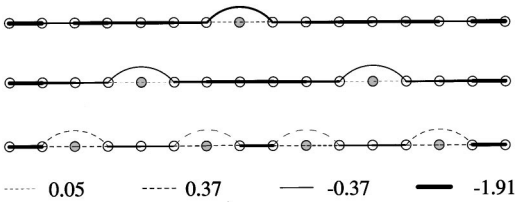

FIG. 4. Same as Fig. 3 but for $(U',J)=(8,6)$. Although in all cases shown $S(k)$ is still incommensurate, the system doped with four holes resembles a polaronic lattice, with the holes located at the centers of small FM islands.

t-J-like models valid at low energies,[11] also with expectations coming from manganite investigations where the relevant densities are close to 1,[8,20] and it could be tested experimentally by increasing the hole doping in the $S=1$ chain compounds.

In addition, doping at stronger couplings than those shown in Fig. 2(a) produces *phase separation* (PS) between hole-rich and hole-poor regions, if J is close to the lower boundary of the $\mathbf{S}=\mathbf{1}$ region of Fig. 1(a). This is the case for the results shown in Fig. 2(b). Such PS regime is characterized by a negative inverse compressibility, the presence of two simultaneous peaks in the spin structure factor at a fixed total electronic density, and a tendency to charge clustering in the local density. Tendencies toward FM and electronic PS found here are similar to those encountered in Kondo-like models for the manganites,[20] and effective models for nickelates.[11] Manganite models also have two orbitals, but in addition they contain localized t_{2g} spins. In the $S=1$ chains considered here, the orbital (x^2-y^2) is fully occupied, and plays a role analog to that of those localized spins of Mn oxides. Then, the tendencies to ferromagnetism observed here may be caused by a mechanism similar to double exchange, which could be operative in highly-doped Ni oxides.

The analysis of the previous paragraphs leads to a second possible rationalization of the spin IC. As explained before, hole doping eventually drives the system into a FM phase. But the transition between the IC phase discussed previously and the FM phase is not always abrupt. Figure 4 contains results analogous to those of Fig. 3 but now for $(U',J)=(8,6)$. In all three doped systems discussed, $S(k)$ was still found to be incommensurate.[21] In Fig. 4, the spin correlation pattern when one or two holes are projected to their most likely position is similar to Fig. 3, but for the case of four holes the spin configuration is clearly different. Instead of AF bonds across the holes, a small three-site island of ferromagnetism is formed around each hole, resembling a magnetic polaron. The presence of FM polarons was already observed in recent studies of doped two-orbital chains with strong Hund's rule couplings.[22] These polarons are coupled in such a way that they generate an overall spin IC (in particular note the way in which the two central polarons are coupled antiferromagnetically). Then, there is a second mechanism to generate spin IC in this context. However, since experimentally no evidence of FM has been reported,[2] the first explanation with AF correlations across holes appears more realistic.

Note that the presence of phase-separation tendencies re-

veal a source of attraction among holes. Actually, in previous literature[11] it was already shown that holes attract in a large region of parameter space, using t-J-like models for Ni-chains. This is reasonable, since each hole creates a spin distortion, and sharing distortions reduces the energy. The pairing of holes in a spin-gapped system has before been documented in models for ladders,[23] and it appears in some models for doped $S=1$ chains.[11] Studies of two-orbital models have also revealed robust singlet-pair correlations.[22] Then, it is tempting to speculate that the material $Y_{2-x}Ca_xBaNiO_5$ may become superconducting upon the application of pressure, as it happens with the two-leg ladder compound [14-24-41]. Experimental studies of doped $S=1$ chains under high pressure may lead to interesting results.

Summarizing, studies of hole-doped $S=1$ chains show that spin incommensurate correlations can be generated in a realistic model,[24] as observed in experiments. The mechanism is similar to the phenomenon recently discussed in $S=1/2$ ladders and planes, namely the generation of AF correlations across holes to optimize the hole movement.[7] It is expected that this mechanism may contribute, at least in part,[25,26] to the understanding of the recently unveiled spin IC tendencies in $Y_{2-x}Ca_xBaNiO_5$.

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[13] The criterion $\langle(\mathbf{S}_j)^2\rangle = S_j(S_j+1) < 1.90$ was used to establish the $S=1$ vs $S\neq 1$ boundary in Fig. 1(a) ($\hbar=1$).

[14] Due to the use of a realistic model with Coulomb interactions and $U=U'+J$, here it is not possible to simply select $J=\infty$ to enforce the $S=1$ state at every nondoped site.

[15] Note that the height of incommesurate peak in Fig. 2(a) increases when the density is reduced from $n=1.750$ to 1.625. This effect is caused by the presence of ferromagnetic islands in the ground state, as illustrated in Fig. 4.

[16] Future calculations of the optical conductivity using Eq. (1) or the model of Refs. 5 and 11 will clarify how robust is the Drude contribution at low energy.

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[19] DMRG calculations for one hole on an odd number of sites chain lead to results very similar to those at the top of Fig. 3, where an even number of sites was used. In both cases robust AF correlations across the hole are clearly observed.

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[21] While a system doped with six holes at $(U',J)=(6,4)$ is also incommensurate [Fig. 2(a)], for $(U',J)=(8,6)$ it is already FM.

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[24] Studies in simpler models of $S=1$ spins and $S=1/2$ holes as proposed in Refs. 5 and 11 may also lead to spin IC features. Results in Ref. 5 already suggest such behavior.

[25] However, more work is needed since the present approach predicts a peak in $S(k)$ located at π-δ, with δ proportional to x, effect that has not been observed yet in experiments (Ref. 2). A complete understanding of doped $S=1$ chains may require other ingredients, such as disorder and interchain coupling. In addition, it would be important to confirm experimentally whether δ indeed tends to a finite number as $x\to 0$, as described by Xu *et al.* (Ref. 2). If this is correct, qualitative important differences exist between doped cuprates and nickelates, not contained in the model studied here.

[26] Other mechanisms may be operative. See I. Bose and E. Chattopadhyay, cond-mat/0012006, Int. J. Mod. Phys. B (to be published).