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Charged magnetic domain lines and the magnetism of high-T_c oxides

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The breakdown of the antiferromagnetism in the high-T_c oxides is studied taking into account the $3d$ charge fluctuations. We point out a tendency towards the formation of charged magnetic domain lines if holes are introduced in such a state, which can be viewed as a generalization to two dimensions of the soliton of Su-Schrieffer-Heeger. In the ground state these domain lines line up, explaining the incommensurate spin phase observed in the high-T_c superconductors.

It is well established that the compounds La_2CuO_4 and $YBa_2Cu_3O_6$ exhibit rather strong antiferromagnetism. It has been argued that this antiferromagnetism can be well explained using a spin-$\frac{1}{2}$ quantum Heisenberg system,[1] indicative of a complete localization of the $3d$ electrons. However, there is accumulating evidence that at least in some respects a bandlike picture is more appropriate. In superconducting samples metallic Fermi edges and a BCS-like gap[2] have been observed in photoemission, and in angular resolved photoemission[3] dispersions are seen in the immediate vicinity of the Fermi level. From a theoretical viewpoint, this is not surprising. The localization of the Cu d holes is not only controlled by the ratio U/V (where U is the on-site d-d Coulomb interaction), but as well by D/V, where V is the p-d hybridization and D the energy cost of the charge fluctuation $d^9 \rightarrow d^{10}+p$-hole (charge-transfer energy).[4,5] Both the analysis of electron spectroscopy data,[6,7] as well as local-density approximation (LDA) calculations for the parameters to be used in the model Hamiltonians[7,8] point at $D/V \leq 1$. The controlling factor for localization is the d-hole count ($\langle n_d \rangle$). Assuming that $0.5 < \langle n_d \rangle < 0.7$ the mass renormalization is at most of order 3 in either the Gutzwiller[9] (slave boson[10]) or T-matrix[11] theories, and together with the highly dispersive σ bands in the vicinity of the Fermi level, bandlike behavior is to be expected.

There is no conflict between this bandlike picture and the situation at half filling. Using the Gutzwiller approximation,[12] or cluster calculations,[13] it can be shown that at half filling a gap of 2 eV is expected in this parameter regime and, obviously, the macroscopic physics can be described with a spin-only Hamiltonian. Another issue is the breakdown of the magnetic order under doping and, at the end, the metal-insulator transition. The experimental facts to be explained are the rapidly decreasing antiferromagnetic (AF) correlation length[14] and the appearance of quasistatic incommensurate spin order,[15] as observed in the $La_{2-x}Sr_xCuO_4$ system. As has been stressed by Schrieffer, Wen, and Zhang,[16] the $3d$-charge fluctuations will give rise to a coupling between the excess holes (introduced by doping) and the *magnitudes* of the spins, and not only to the orientation of the spins as in the localized regime.[17] In this Rapid Communication we will show that, according to electronic mean field, this leads to a condensation of the excess holes in charged solitons. We argue that this "striped phase" may well account for the experimental observations.

It is generally accepted that only the $d_{x^2-y^2}$ band is of relevance and the electronic structure can be modeled by the well-known two-band Hamiltonian[18]

$$H = H_0 + H_1, \quad H_0 = \sum_{j\sigma} \varepsilon_p p^\dagger_{j\sigma} p_{j\sigma} + \sum_{i\sigma} \varepsilon_d d^\dagger_{i\sigma} d_{i\sigma} + \sum_{\langle ij \rangle \sigma} V_{ij} (d^\dagger_{i\sigma} p_{j\sigma} + \text{H.c.}), \quad H_1 = \sum_i U n_{di\uparrow} n_{di\downarrow}, \tag{1}$$

with $D = \varepsilon_p - \varepsilon_d$. In the Hartree-Fock approximation (HFA), H_1 is approximated by

$$H_1^{MF} = U \sum_{i\sigma} \Delta_{i\sigma} n_{di-\sigma} + U \sum_i (\Delta_{i-} d^\dagger_{i\uparrow} d_{i\downarrow} + \Delta_{i+} d^\dagger_{i\downarrow} d_{i\uparrow}) - U \sum_i (\Delta_{i\uparrow} \Delta_{i\downarrow} + \Delta_{i-} \Delta_{i+}), \tag{2}$$

with $\Delta_{i\sigma} = \langle n_{di\sigma} \rangle$, $\Delta^*_{i-} = \Delta_{i+} = -\langle d^\dagger_{i\uparrow} d_{i\downarrow} \rangle$. Alternatively, we can write

$$\langle n_{di\uparrow} \rangle = (\langle n_{di} \rangle + \langle m_{di} \rangle \cos\theta_i)/2,$$

$$\langle n_{di\downarrow} \rangle = (\langle n_{di} \rangle - \langle m_{di} \rangle \cos\theta_i)/2,$$

and

$$\langle d^\dagger_{i\uparrow} d_{i\downarrow} \rangle = \sin\theta_i (\cos\phi_i + i \sin\phi_i) \langle m_{di} \rangle / 2.$$

$\langle n_{di} \rangle$ and $\langle m_{di} \rangle$ refer to, respectively, the total d-hole count and the magnitude of the spin at site i, while θ_i and ϕ_i refer to the canting angles of the spins with respect to the quantization axis. In this way we account for the proper [O(3)] symmetry of the order parameter.

At half filling the system is unstable towards the formation of a Néel state, which leads to a gap in the charge excitation spectrum. Using the spectroscopic parameters [$D \approx V$, $V \approx 1.8$ eV, $U \approx 8$ eV (Ref. 6)], one finds from Eqs. (1) and (2) $\langle m_{di} \rangle \approx 0.5$ and $\langle n_{di} \rangle \approx 0.6$ for the perovskite planes. The magnitude of the charge excitation gap is then ≈ 2 eV, in agreement with what is experimentally known [≈ 2.0 eV (Ref. 19)]. At least in this respect a reasonable picture is derived from HFA.

Our mean-field Hamiltonian is similar to the Su-

Schrieffer-Heeger (SSH) Hamiltonian[20] used to describe the solitons in polyacetelene, except for the continuous [O(3)] symmetry of the order parameter. In the localized limit, this difference is rather crucial as the experience with the $J-t$ model shows. However, if the $3d$-charge degrees of freedom are not neglected, the amplitude fluctuations of the spins are on a larger (electronic) energy scale than the transverse fluctuations ($\propto J$) if excess charge is present, as is well known from itinerant magnetism. On the mean-field level this leads to an effective symmetry lowering; the order parameter responds on an excess charge as if it has a discrete symmetry. For example, numerical evaluation of Eqs. (1) and (2) on a finite lattice show that at half filling domain walls spread out to infinity, while in the presence of excess charge the amplitudes $\Delta_i \pm$ are essentially zero in the converged solutions and in one dimension (1D) the familiar charged SSH solitons[20] are found.

In contrast to 1D, SSH-type Hamiltonians are to our knowledge not extensively investigated in 2D.[21] We report here on numerical results obtained for Eqs. (1) and (2) applied to the perovskite planes for parameters representative for the superconductors ($D/V=1$, $U/V=4$). For numerical convenience, we use large two-dimensional supercells (with up to 10×10 elementary CuO_2 unit cells) subject to periodic boundary conditions. The supercells are sufficiently large to make the results essentially independent of their size, and our findings can be taken to be representative for the infinite system. As in the case of SSH solitons, the results depend on the topological constraints coming from the boundary conditions. We can use this to study excitations of the system.

Having one excess hole per supercell, a spin bag[16,22] is found in the ground state. As in the localized regime, the majority of the charge is localized on the "oxygen" ions ($\approx 70\%$), while the spins reside mainly on the "Cu" ions. The energy gain due to self-localization can be found by comparing the total energy of the spin-bag solution with the total energy of the supercell containing one extended excess hole, and we find the bag to be stabilized by an amount $E_{\text{bag}} \simeq 0.06V$. Figure 1(a) shows the result for the ground state of a 9×10 supercell containing 10 excess holes. This corresponds to a 10 unit-cell long *charged Néel line* running along the (0,1) direction in the perovskite plane. In Fig. 1(b) we show such a result for a 10×10 unit cell also containing 10 excess holes. In this case we also find a 10 unit-cell long domain line, which now forms a closed ring. Because of the periodic boundary conditions, a straight line would be accompanied by a neutral Néel line in this supercell, and it is energetically less expensive to bend the line. We can determine the energy per elementary unit cell in the domain line (ε_{dl}) by using $\varepsilon_{dl}=[E_{\text{tot}}-(N_xN_y-N_{dl})\varepsilon_{af}]/N_{dl}$, where N_x, N_y correspond to the x and y dimensions of the supercell, and N_{dl} to the number of elementary domain-line unit cells. E_{tot} is the total energy of the supercell and ε_{af} is the energy per unit cell at half filling. ε_{dl} is now nearly independent of the size of the supercell for doping concentrations $<10\%$. In this way we find that the (1,0) line is slightly more stable ($0.005V$/ carrier) than the (1,1) line. Assuming that the energy of the closed domain line can be separated into the energies of the corners and the straight sections, we find that it costs $E_{\text{corn}} \simeq 0.025V$ to make a 90° corner in a straight line. Using appropriate boundary conditions, we can force the lines to cross and this costs $\simeq 2E_{\text{corn}}$. Having $2N$ holes in a $N\times N$ unit cell leads to two parallel lines repelling each other. Finally, by comparing the energy of an isolated spin bag ($N_{dl}=1$) with that of a straight domain line, we find that the latter is more stable by $\simeq 0.05V$/carrier. *The ground state of the infinite system at low doping concentration thus consists*

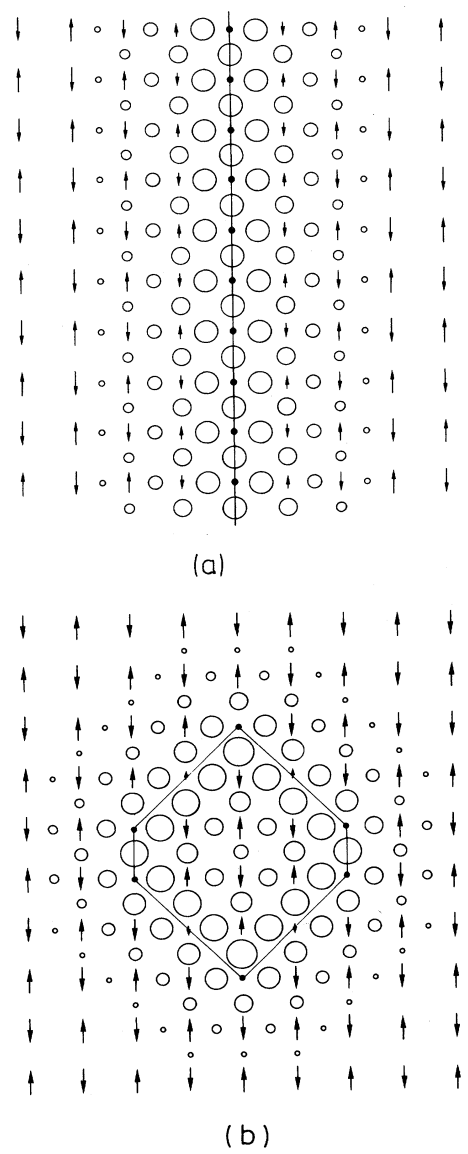

FIG. 1. Results of numerical simulations (see text) for holes on antiferromagnetic perovskite planes. The density of excess holes on the oxygen ions is proportional to the radius of the circles and the spins on the Cu lattice are represented by the arrows. Note that according to these calculations the spins do not cant. (a) Ten holes condense into a charged Néel line in a 9×10 supercell. (b) In a 10×10 supercell the line is forced to form a ring, because of the periodic boundary conditions.

of straight, charged Bloch lines aligned parallel to each other. This result is not totally surprising. It is similar to the so-called striped phase, known from the study of two-dimensional incommensurate systems.[23] In our case, the two competing length scales are, on the one hand the lattice parameter, and on the other hand $1/k_F$, proportional to the number of holes.

From the numerical results we can extract a qualitative picture of the electronic structure. Apart from the valence and conduction bands, separated by the antiferromagnetic gap, we find an *approximately midgap band* corresponding to the domain-line structure. Each domain-line unit cell contributes one state to the band and the line is optimally stable if this band is completely filled with the excess holes. For instance, if we add one more hole to the domain line in the uneven by uneven unit cell we find that an additional spin bag is formed and removing a hole leads to a neutral unit cell in the line, both of which are unfavorable. It is also costly to polarize the line magnetically ($\simeq 0.15V$ per spin flip). The width of the midgap band is governed by the relative orientation of the line with respect to the underlying lattice. For instance, for the (1,0) case, in contrast to the (1,1) case, the nearest-neighbor Cu atoms along the domain line share nearest-neighbor O atoms, and hopping of holes along the domain line is efficient. The calculations show that the width of the midgap band is larger in the (1,0) case ($\simeq 0.3V$) than in the (1,1) case ($\simeq 0.1V$). The gap between the filled valence band and the empty domain-line band shows the opposite trend (respectively $\simeq 0.1$ and $0.3V$). We thus find that this domain-line phase is electronically characterized by *zero spin, a new charge excitation gap which is appreciably smaller than the AF gap, and finally, a counting rule stating that the number of domain-line unit cells is equal to the number of carriers.*

Our calculations suggest that the spatial extent ("coherence length") of the isolated bags is already quite small in this parameter regime,[24] suggesting that an effective strong-coupling [i.e., *J-t* (Ref. 25)] model would make sense. In that case, one expects that the condensation energy of the domain lines would be outweighted by the kinetic energy of the isolated bags, which is neglected in HFA.[26] However, in trying to do better than HFA one should be careful. In this respect, it is interesting to consider what the outcome would be if the Gutzwiller approximation (GA) is used instead of HFA. The structure of GA is similar to that of HFA (Ref. 12) and it is also expected that a stable striped phase will be found. However, in GA the coherence length would be much larger because the paramagnetic state, realized in the core of the domain line, is more stable than in HFA for the same charge distribution, gap, etc. This only arises from a better treatment of the charge fluctuations.[12] At least with respect to the center-of-mass motion of the bags, this would bring the problem closer to the weak-coupling limit. Another aspect is the longer-ranged Coulomb interaction. Although the electrostatic energy destabilizes the striped phase at very low hole concentrations, it is also expected that at least the nearest-neighbor Coulomb interactions help domain-line structures.[27,28]

Turning to experiment (see also Refs. 29 and 30): According to our domain line picture the AF incommensurability $\Delta Q = Q_0 n_h$ where Q_0 is the AF wave number and n_h is the number of excess holes. This simple proportionality between hole count and incommensurability seems to be confirmed by neutron measurements.[29] Birgeneau *et al.* studied in detail $x = 0.12$ samples[15] and here the incommensurability is of the right order ($n_h \simeq 0.08$), although somewhat smaller than expected from our picture, assuming $x = n_h$. It would be interesting to see what the real n_h in these samples is, i.e., one expects that less holes are present than suggested by Sr concentration because of oxygen deficiency. Also the unit-cell doubling along the c axis observed by Birgeneau *et al.*[15] can be understood in this framework as originating in the Coulomb repulsion of lines in different planes.[29]

Finally, also with respect to the electronic structure this domain-line picture is suggestive. The simplest explanation of the unoccupied density of states in doped La_2CuO_4 samples, as measured by electron-loss spectroscopy,[30] is in terms of a hole-induced midgap band. Also, optical measurements seem to point in this direction.[19,31] As we pointed out, our striped phase is insulating. According to Birgeneau *et al.*,[15] the vast majority of samples showing the incommensurate spin structure is not superconducting and apparently insulating at low temperature, while superconducting samples are exceptional. In this respect, one would expect that the stability of these charged domain lines would be influenced by lattice (defect) structures. It is interesting that some Bi and Tl cuprates are characterized by lined up BiO (TlO) vacancies, i.e., a striped phase, in the BiO (TlO) planes[32] with a period of the same order as expected for the magnetic lines. It turns out that this vacancy-order prohibits superconductivity.[32] Obviously, these discommensurations tend to stabilize the magnetic striped phase, suggesting that the *striped phase and superconducting phase are actually competing.* It would be interesting to see if in a similar way Sr-impurity ordering can explain the puzzling variation of transport properties in the La_2CuO_4 samples.

Although a direct relationship between magnetism and superconductivity in the cuprates remains to be proven, in at least one respect this domain-line picture is suggestive. As is well known, the same interactions can give rise to either charge density wave or superconducting instabilities. Our domain-line phase is analogous to the former, giving support for the spin-bag picture of Schrieffer *et al.*[16] as the theory for the latter.

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