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Antiferromagnetism of CuO_2 layers within a slave-boson approach

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The antiferromagnetic ground state of CuO_2 layers is studied at the mean-field level within a slave-boson approach to a multiband Hubbard model which includes the $d_{x^2-y^2}$ orbital for Cu and the p_x and p_y orbitals for O. The stability of the antiferromagnetism is tested by varying the separation Δ between p- and d-hole energy levels and by allowing direct O-O hopping. Results for the effective exchange integral, the antiferromagnetic gap, the percentage of d character of holes at the Fermi level, and the Cu magnetic moment are discussed by varying Δ.

The experimental finding of quasi-two-dimensional antiferromagnetic (AF) long-range order in both $La_{2-x}Sr_xCuO_4$ (Ref. 1) and $YBa_2Cu_3O_{6+x}$ (Ref. 2) has commonly been interpreted in terms of a superexchange picture,[3] whereby the spins are assumed to be localized about their lattice sites. The CuO_2 layers appear, however, to lie somewhere at the border line between a localized and an itinerant description of magnetism. Although there exists, in fact, general consensus on the strongly correlated character of the CuO_2 layers owing to the large value of the Coulomb repulsion at Cu sites, the experimental uncertainties on the charge-transfer gap Δ makes both descriptions possible *a priori*.

In this Rapid Communication the AF ground state of the CuO_2 layers is studied within a slave-boson approach to a multiband Hubbard model. This approach is appropriate to strongly correlated systems and allows for a smooth interpolation between the itinerant and the localized limits by varying the parameter Δ. We consider a mean-field approximation which is expected to capture the basic physical properties we are interested in. The effects of the three-dimensional coupling between layers as well as the systematic inclusion of fluctuations about the mean field may, in fact, be addressed separately upon selecting specific properties of interest (such as the spin-wave velocity, etc.).

The stability of the AF ground state is studied by comparison with its paramagnetic counterpart. Although the method allows us to study a CuO_2 layer with an arbitrary number of holes, we report on the case of one hole per Cu site. It will also be shown that for large values of Δ our approach recovers the canonical transformation method for a charge-transfer insulator.[4]

The method we consider extends to a multiband Hubbard model the treatment by Kotliar and Ruckenstein for a one-band Hubbard model.[5] The extension is physically nontrivial since the charge excitations that are controlled by the on-site Coulomb repulsion in a one-band Hubbard model (Mott insulator), are controlled instead by the gap Δ in our case (charge-transfer insulator). The $d_{x^2-y^2}$ orbital for Cu and the p_x and p_y orbitals for O are included with nearest- (t_1) and next-nearest- (t_2) neighbor hopping. The relevant Hamiltonian thus reads (in hole notation)

$$H=\sum_{ij}\sum_{\nu\sigma}t_1(d_{i\sigma}^{\dagger}p_{j\nu\sigma}+\text{H.c.})+\sum_{jj'}\sum_{\nu\nu'\sigma}t_2(p_{j\nu\sigma}^{\dagger}p_{j'\nu'\sigma}+\text{H.c.})+\varepsilon_d\sum_{i\sigma}d_{i\sigma}^{\dagger}d_{i\sigma}+\varepsilon_p\sum_{j\nu\sigma}p_{j\nu\sigma}^{\dagger}p_{j\nu\sigma}$$
$$+U_d\sum_i d_{i\uparrow}^{\dagger}d_{i\uparrow}d_{i\downarrow}^{\dagger}d_{i\downarrow}+U_p\sum_{j\nu}p_{j\nu\uparrow}^{\dagger}p_{j\nu\uparrow}p_{j\nu\downarrow}^{\dagger}p_{j\nu\downarrow}\,, \tag{1}$$

where the lattice sums are limited to neighboring sites, the index ν distinguishes p_x and p_y orbitals (with suitable phase convention for the orbitals and for the corresponding hopping integrals), σ is the spin projection, $\varepsilon_p-\varepsilon_d=\Delta$, and U_d and U_p are the Coulomb repulsions at the Cu and O sites, respectively. Typical tight-binding fits to the band structure of La_2CuO_4 give $t_1\cong 1.4$ eV and $|t_2/t_1|\cong\frac{1}{4}/\frac{1}{2}$.[6] The value of Δ/t_1 (which includes Coulomb and Madelung effects) is rather controversial, ranging from 0 to 4. Reasonable estimates for U_d/t_1 and U_p/t_1 are about 7 and 1.5, respectively, allowing one to take U_d as the largest energy scale. In the following, we shall consider the limit $U_d\approx\infty$.

In this limit, slave bosons which specify empty and singly occupied states are assigned to Cu sites to deal with the infinite Hubbard repulsion. We indicate by e_i and $s_{i\sigma}$ the associated bosonic operators, respectively. The local repulsion on O sites is instead regarded to be small and treated conveniently in a Hartree-Fock decoupling. Within the enlarged Fock space, the Hamiltonian (1) is then replaced by

$$H=\sum_{ij}\sum_{\nu\sigma}t_1(z_{i\sigma}^{\dagger}d_{i\sigma}^{\dagger}p_{j\nu\sigma}+\text{H.c.})+\sum_{jj'}\sum_{\nu\nu'\sigma}t_2(p_{j\nu\sigma}^{\dagger}p_{j'\nu'\sigma}+\text{H.c.})+\varepsilon_d\sum_{i\sigma}d_{i\sigma}^{\dagger}d_{i\sigma}+\varepsilon_p\sum_{j\nu\sigma}p_{j\nu\sigma}^{\dagger}p_{j\nu\sigma}+U_p\sum_{j\nu\sigma}(n_0p_{j\nu\sigma}^{\dagger}p_{j\nu\sigma}-\tfrac{1}{2}n_0^2)\,, \tag{2}$$

with the condition $\langle p_{jv\sigma}^{\dagger} p_{jv\sigma} \rangle = n_0$ at the O sites. Here $z_{i\sigma}$ is a bosonic operator introduced by the mapping of the kinetic term of the original Hamiltonian (1) into the enlarged Fock space.[5] Its form is not unique because this mapping is redundant. Attention must be paid to the choice of $z_{i\sigma}$ while dealing with approximations.

In the original description (1) each Cu site is at most singly occupied by holes. The Hamiltonian (2) must thus be supplemented by constraints which avoid multiple bosonic occupation at a given Cu site:

$$\text{(I)}\ e_i^{\dagger} e_i + \sum_{\sigma} s_{i\sigma}^{\dagger} s_{i\sigma} = 1 , \qquad \text{(II)}\ d_{i\sigma}^{\dagger} d_{i\sigma} = s_{i\sigma}^{\dagger} s_{i\sigma} . \tag{3}$$

As for a single band, a convenient way to handle the constraints (3) is to resort to a functional integral approach for the coupled Bose-Fermi problem, whereby the constraints are enforced by introducing Lagrange multiplier fields, λ_i^{I} and $\lambda_{i\sigma}^{\mathrm{II}}$ in the order, at each Cu site i. This approach provides a mean-field description of the antiferromagnetism of the CuO_2 layers by suitably selecting the saddle point for the bosonic part of the functional while satisfying the constraints (3) on the average. One then has to solve seven nonlinear coupled equations for the seven parameters e, $s_{\sigma} = s_1 + \sigma s_2$, the chemical potential, λ^{I}, and $\lambda_{\sigma}^{\mathrm{II}} = \lambda_1^{\mathrm{II}} + \sigma \lambda_2^{\mathrm{II}}$. [By convention, the parameters are specified for the atoms of the A sublattice for which $\exp(i\mathbf{Q}\cdot\mathbf{R}_i) = 1$, where $Q = (\pi/2a)(1,1)$ and $\mathbf{R}_i$ ranges over all Cu sites, a being the Cu-O distance. For the atoms of the B sublattice it is sufficient to replace $\sigma \rightarrow \bar{\sigma} = -\sigma$.]

The arbitrariness of the operator $z_{i\sigma}$ must be resolved within the mean-field approximation so as to reproduce known results and to avoid spurious outcomes. Kotliar and Ruckenstein[5] have shown that the choice

$$z_{i\sigma} = e_i^{\dagger} \frac{1}{(1 - D_i^{\dagger} D_i - s_{i\sigma}^{\dagger} s_{i\sigma})^{1/2} (1 - e_i^{\dagger} e_i - s_{i\bar{\sigma}}^{\dagger} s_{i\bar{\sigma}})^{1/2}} s_{i\sigma} + s_{i\bar{\sigma}}^{\dagger} \frac{1}{(1 - D_i^{\dagger} D_i - s_{i\sigma}^{\dagger} s_{i\sigma})^{1/2} (1 - e_i^{\dagger} e_i - s_{i\bar{\sigma}}^{\dagger} s_{i\bar{\sigma}})^{1/2}} D_i \tag{4}$$

reproduces the independent-particle limit within the mean-field approximation to a single-band Hubbard model. [The operator D_i, which specifies the double occupancy of a given Cu site, needs in general to be retained in Eq. (4) for finite U_d.] One may argue that it is not evident *a priori* that the choice (4) should also apply to a strongly correlated multiband system with AF order, for which $s_{i\uparrow}$ differs from $s_{i\downarrow}$. It turns out from our calculation that the choice (4) reproduces the AF energy of a Néel state with one hole per Cu site in the limit of large separation Δ (i.e., for $\Delta/t_1 \gtrsim 10$), when comparison with perturbation theory in t_1/Δ is meaningful. Specifically, our numerical results for the ground-state energy can be fitted by the expression[4]

$$E_{\mathrm{AF}} \approx N(-4t_{\uparrow}^2/\Delta + 16 t_{\uparrow}^4/\Delta^3 + \cdots) \tag{5}$$

(N being the number of Cu sites) that holds in the present form when $t_2 = 0$. In this limit, the slave-boson approach also accounts for the clustering of the magnetically ordered ground state into molecularlike orbitals, by suitably renormalizing the hopping for the two spin projections. Specifically, for atoms of the A sublattice $z_{i\uparrow}$ tends quickly to unity for increasing Δ while $z_{i\downarrow}$ vanishes (vice versa for the atoms of the B sublattice). This implies that, because of correlations, when Δ is sufficiently large a hole with spin up can hop freely between a type-A Cu and the four surrounding oxygens, while its hopping to a neighboring type-B Cu is hindered. The slave-boson approach, upon affecting directly the hopping integrals for the two spin projections and the two sublattices, accounts then for the superexchange mechanism[3] in the limit of large Δ. We thus feel confident in adopting the choice of (4) for all values of Δ.[7]

It is also interesting to verify whether the corresponding perturbative expression for the total paramagnetic energy at half filling, namely,

$$E_P \approx N(-4t_{\uparrow}^2/\Delta + 18 t_{\uparrow}^4/\Delta^3 + \cdots) , \tag{6}$$

is reproduced by the present slave-boson approach. Kotliar and Ruckenstein[5] have shown that, in the single-band case, their approach reduces to the Gutzwiller representation of the Mott insulator when the Hubbard repulsion exceeds a finite critical value. In this situation, the effective hopping integral and the free energy vanish identically. This result is a shortcoming of the Gutzwiller approximation which inhibits the electronic fluctuations in the localized phase. A similar result is obtained in the multiband case we consider, where the Gutzwiller representation of the charge-transfer insulator emerges from $\Delta \geq \Delta_c$ with $\Delta_c/t_1 \cong 4\sqrt{3}$. A direct numerical comparison of the ground-state energies per Cu site in the two phases, to extract an effective exchange integral J_{eff}, is then bound to give too large values for J_{eff} when $\Delta \gtrsim \Delta_c$. For instance, when $\Delta/t_1 = 10$ we would obtain $J_{\mathrm{eff}} \cong 0.75 t_1$ while comparison of Eqs. (5) and (6) gives $J_{\mathrm{eff}} \equiv 2(E_P - E_{\mathrm{AF}})/N = 4t_{\uparrow}^4/\Delta^3 = 4 \times 10^{-3} t_1$.

A more reliable estimate of the curve $J_{\mathrm{eff}}(\Delta)$ can be obtained by interpolating the total paramagnetic energy between its asymptotic expression (6) valid for $\Delta \gtrsim \Delta_c$ and the corresponding numerical results for smaller Δ. The resulting $J_{\mathrm{eff}}(\Delta)$ is reported in Fig. 1. Notice that $J_{\mathrm{eff}}(\Delta)$ shows a maximum at intermediate values $\Delta/t_1 \approx 3$, thereby fulfilling one's physical intuition that either too little or too much kinetic energy disfavors the occurrence of antiferromagnetism. The reported experimental value $J_{\mathrm{exp}} \cong 0.14$ eV for La_2CuO_4 (Ref. 8) is seen from Fig. 1 to be consistent with both $\Delta/t_1 \cong 0.5$ and $\Delta/t_1 \simeq 4.5$ when $|t_2/t_1| = \frac{1}{4}$; these two values lie within the interval of the reported value for Δ and correspond possibly to an itinerant and to a localized picture of magnetism, respectively.[9] Notice also from Fig. 1 that the inclusion of the O-O hopping t_2 changes the picture somewhat quantitatively.[10]

The Cu magnetic moment μ is also experimentally accessible, a typical value for La_2CuO_4 being about half the Bohr magneton.[11] The function $\mu(\Delta)$ is seen from our calculation to range smoothly from $\cong 1$ when $\Delta/t_1 \gtrsim 10$ to 0.3/0.4 when $\Delta = 0$, the reduction originating from the spread of the Cu Wannier orbital on the neighboring O

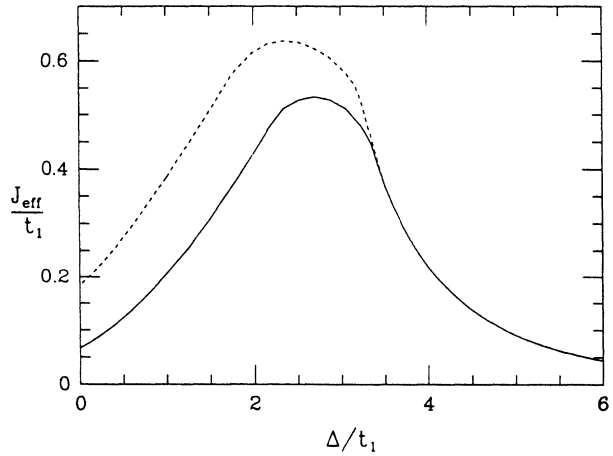

FIG. 1. Effective exchange integral vs Δ with (solid line) and without (dashed line) inclusion of the O-O hopping t_2.

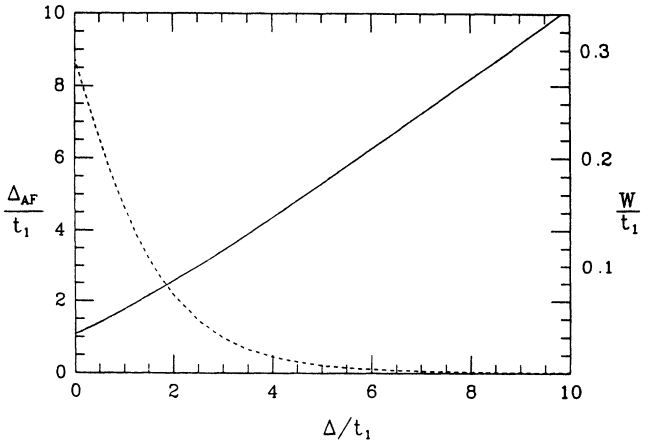

FIG. 2. Antiferromagnetic gap Δ_{AF} (solid line) and width W of the lowest band (dashed line) vs Δ when $t_2=0$.

sites by decreasing Δ. For large Δ it is known that the limiting value of μ is reduced (by about a factor of 0.6) due to the occurrence of two-dimensional quantum fluctuations in the Heisenberg antiferromagnet for spin $\frac{1}{2}$.[12] Conventional spin-wave theory, however, cannot be used to estimate the reduction of μ for moderate values of Δ when the spins partly itinerant. To this end, it would instead be necessary to include the fluctuations about the saddle point within the functional integral formulation we are adopting.

The effective one-particle picture associated with our mean-field approach is obtained by diagonalizing the Hamiltonian matrix of Eq. (2) [with the addition of the terms originating from the constraints (3)]. At half filling the lowest (degenerate) spin-up and spin-down bands are filled and separated by an energy gap Δ_{AF} from the higher empty bands. In Fig. 2 we report Δ_{AF} vs Δ when $t_2=0$. The AF gap is found to be $\cong\Delta$ for large values of Δ/t_1 (thereby identifying Δ_{AF} with the charge excitation gap) and to remain finite for vanishing Δ. The latter result can be understood by the occurrence of a strong nesting condition in two dimensions at half filling, which stabilizes the antiferromagnetism against charge fluctuations. In Fig. 2 we also report the width W of the lowest (filled) band versus Δ. This band becomes quickly rather narrow and well separated from the higher bands for increasing Δ. From the band structure one can also extract the percentage of d character of holes at the Fermi level. For instance, when $\Delta/t_1\cong 3$ we get about $\frac{3}{4}$ for this value, in agreement with cluster calculations.[6] These results change quantitatively with the inclusion of t_2. The absolute gap Δ_{AF} is, however, seen to disappear at a critical value $|t_2/t_1|_c$ which depends on Δ. For instance, one gets $|t_2/t_1|_c\cong 0.4+0.3\Delta/t_1$ when $\Delta/t_1\lesssim 2$.

We can finally compare our results with a full Hartree-Fock decoupling on both Cu and O sites. The latter approach has recently been followed by Mila to assess the itinerant versus localized nature of the antiferromagnetism of the CuO_2 layers.[13] The outcome of Mila's calculation for the effective exchange integral versus Δ is similar to our result.[14] There are, however, quantitative differences between the two approaches, as for the quantity $\Delta_{AF}-\Delta$ which is about a factor of 2 larger within the Hartree-Fock approach.

In conclusion, we have presented results for the AF ground-state properties of CuO_2 layers within a mean-field approach. The method we have adopted is specifically suited to deal with the strong correlation at Cu sites, and as such it should provide an optimized single-particle description of several experimentally accessible quantities. Full comparison with experiments as well as the assessment of the role of antiferromagnetism on the superconducting mechanism await, however, the inclusion of fluctuations about the saddle point.

Note added. After our work had been submitted for publication, two papers presenting work similar to ours have appeared. The paper by A.M. Oleś and J. Zaanen [Phys. Rev. B **39**, 9175 (1989)] relies on a Gutzwiller approach and arrives at similar conclusions as the present paper although for a one-dimensional case. In the paper by Balseiro *et al.* [Phys. Rev. Lett. **62**, 2624 1989)] the Kotliar and Ruckenstein approach is also used to obtain the instability of the paramagnetic phase of the CuO_2 layers against antiferromagnetism. The assumption of a constant density of states by Balseiro *et al.*, however, makes their results rather questionable since in that way they completely miss the nesting condition of the Fermi surface.

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[1]G. Shirane *et al.*, Phys. Rev. Lett. **59**, 1613 (1987).

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[4]J. Zaanen and A. M. Oleś, Phys. Rev. B **37**, 9423 (1988).

[5]G. Kotliar and A. E. Ruckenstein, Phys. Rev. Lett. **57**, 1362 (1986).

[6]F. Mila, Phys. Rev. B **38**, 11 358 (1988).

[7]We have addressed the question whether a different choice from (4) would account for the correct expression (5) for large Δ. In particular, one may notice that the independent-particle result, which has led Kotliar and Ruckenstein to the choice (4), is not affected upon replacing in the square roots of Eq. (4) $s_{i\sigma}^2$ by $\alpha s_{i\sigma}^2+(1-\alpha)s_{i\bar{\sigma}}^2$ and $s_{i\bar{\sigma}}^2$ by $\beta s_{i\bar{\sigma}}^2+(1-\beta)s_{i\sigma}^2$, with $0\leq\alpha\leq 1$ and $0\leq\beta\leq 1$. It can be shown, however, that any other choice different from Eq. (4) leads to the appearance of an unphysical critical value of Δ above which antiferromagnetism cannot be sustained any longer.

[8]K. B. Lyons, P. A. Fleury, J. P. Remeika, and T. J. Negran, Phys. Rev. B **37**, 2353 (1988).

[9]A *direct* experimental check on which side of the maximum of the curve $J_{\text{eff}}(\Delta)$ the CuO_2 layers of La_2CuO_4 actually lie could be done by measuring the Néel temperature upon applying uniaxial pressure to the layers so as to decrease the ratio Δ/t_1. Notice, however, that the quantity $J_{\text{eff}}(\Delta)$ which we have derived using the difference in energy of the AF and the paramagnetic states may not be directly related to the spin-wave velocity for small Δ. The effective exchange integral related to the latter follows from the singularity of the transverse spin susceptibility and thus requires the inclusion of Gaussian fluctuations about the AF saddle point.

[10]Inclusion of t_2 is, in principle, important since it removes the perfect nesting condition of the square Fermi surface in the paramagnetic case at half filling.

[11]D. Vaknin *et al.*, Phys. Rev. Lett. **58**, 2802 (1987).

[12]Including this reduction factor, the limiting experimental values $\mu=(0.48\pm 0.15)\mu_B$ for La_2CuO_4 are recovered by our calculation when t_2 is nonvanishing for $\Delta/t_1\cong 1$ and $\Delta/t_1\simeq 5$, respectively (the average value $\mu=0.48\mu_B$ corresponding to $\Delta/t_1\cong 2.5$).

[13]F. Mila, Europhys. Lett. **8**, 555 (1989).

[14]The reason why a Hartree-Fock decoupling works for strong coupling (as far as the ground-state energy is concerned) is probably the good choice of the trial wave function for the broken symmetry phase in the form of a Néel state.