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Electronic structure of high-T_c superconductors and related compounds

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Abstract. We study a 5-band Hubbard model for the CuO_2 planes in cuprate superconductors using Hartree-Fock mean-field theory including spiral spin density waves. For the half-filled case we recover a ZSA-like phase diagram *but with an additional new region characterized by strong covalency effects, which we call a covalent insulator region.* We also provide a nonperturbative calculation of J_{eff}, the effective in-plane antiferromagnetic interaction, as a function of parameters of the model. We suggest that the high-T_c cuprates are in or very close to the covalent insulator region and within this we show that a consistent explanation of apparently conflicting high energy spectroscopic and magnetic measurements of the high-T_c cuprates can be given.

Keywords. Electronic structure; oxide superconductors; phase diagram; transition metal compounds; covalency.

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A large number of experiments over the last few years have systematically provided limits on various energetics that models of the electronic structure of high-T_c cuprates and related compounds must conform to. There is an emerging consensus (Fujimori *et al* 1987; Shen *et al* 1987; Yarmoff *et al* 1987; Sondericker *et al* 1987; Bianconi *et al* 1987; van der Marel 1988; Eskes and Sawatzky 1988; Nucker *et al* 1988; Kuiper *et al* 1988; Sarma *et al* 1988; Anderson 1987, 1988; Fulde 1988) that large intra-atomic Coulomb interactions are operative within the Cu 3*d* manifold; that the charge carriers are primarily active in the CuO_2 planes; and that the mechanism of superconductivity in these cases is electronic in nature and is closely related to the strong antiferromagnetic correlations present in these systems, in general agreement with Anderson's original suggestions (Anderson 1988).

However, beyond the above mentioned agreement on certain general trends, several experiments have been interpreted in terms of contradictory electronic structure. A large number of magnetic measurements (probing the low-energy scale of the spin degrees of freedom) in the insulating cuprates have been interpreted in terms of a highly ionic picture (Chakravarty *et al* 1988; Horsch and van der Linden 1988; Vaknin *et al* 1987; Mitsuda *et al* 1987; Huse 1988; Aeppli *et al* 1989; Hayden *et al* 1990; Sinha 1987, 1990; Freltoft *et al* 1988; Birgeneau and Shirane 1989), with more than 90% of the moment being at the Cu site. On the other hand, high-energy spectroscopies (sensitive to the large energy scale associated with the charge degrees of freedom) (Fujimori *et al* 1987; Shen *et al* 1987; Yarmoff *et al* 1987; Sondericker *et al* 1987; Bianconi *et al* 1987; van der Marel *et al* 1988; Eskes and Sowatzky 1988; Nucker

et al 1988; Sarma 1988; Sarma and Taraphder 1989) and band-structure calculations (Mattheiss 1987; Haas 1989; Pickett 1989) suggest that these systems are highly covalent, with a strongly nonintegral d-occupancy. While there has been a recent attempt (Brandow 1990) to reinterpret the high-energy spectroscopy data with parameter values closer to the ionic limit, this very unsatisfactory situation has not yet been fully resolved. Furthermore recent angle-resolved photoemission studies (Takahasi *et al* 1988; Olson *et al* 1989; Campuzano *et al* 1990) show direct evidence of extensive band dispersion, indicating substantial covalency. Clearly a discussion of the electronic structure of these compounds must resolve the conflict between the two descriptions; and this will have an important bearing on theoretical attempts to explain the mechanism of superconductivity.

In this letter, we have attempted to understand these issues within a unified approach. We model the system along well-accepted lines (Anderson 1988; Fulde 1988; McMahan *et al* 1988; Hybertsen *et al* 1989; Stechel and Jennison 1990; Oles and Zaanen 1989), including a two-dimensional CuO_2 lattice, with O–$2p_{x,y}$ and Cu–$3d_{x^2-y^2}$ orbitals only. The bare charge transfer energy, $(\Delta = \varepsilon_d - \varepsilon_p)$, the various intra- and inter-atomic Coulomb interactions (U_{dd}, $U_{pp} = U_{p_yp_y} = U_{p_xp_y} = U_{p_xp_x}$ and U_{pd}) and all the near neighbour Cu–O as well as O–O hopping matrix elements ($t_{pd}, t_{p_xp_x}$ and $t_{p_xp_y}$) are taken into account leading to a 5-band extended Hubbard model (Anderson 1988; Fulde 1988; McMahan *et al* 1988; Hybertsen *et al* 1989; Stechel and Jennison 1990; Oles and Zaanen 1989). We treat this model using a Hartree-Fock mean-field approximation allowing for spiral spin density wave (SDW) states (Krishnamurthy *et al* 1990; Sarker *et al* 1991). The Hamiltonian is diagonalized self-consistently, with a convergence criterion that the differences between the order parameters (and the eigenvalues) in successive iterations were less than 10^{-6}. The calculations were performed with 256 k-points in the Brillouin zone, though checks were made extensively by increasing the number of k-points to 576 for various values of the parameters and few calculations were also done with 1600 k-points.

From this model at "half-filling" we obtain a Zaanen, Sawatzky and Allen (ZSA) like phase diagram (Zannen *et al* 1985) *with some important distinctions*. We argue that these differences between the original ZSA diagram and the present one have relevance to the electronic structure of the high-T_c cuprates. Furthermore, we analyse the magnetic properties in the insulating phase by mapping the results obtained with the spiral SDW onto a Heisenberg model with a J_{eff}. In all the calculations we use $t_{pd} = 1$ eV, $t_{p_xp_x} = 0{\cdot}19$ eV and $t_{p_xp_y} = 0{\cdot}5$ eV in accordance with the various estimates available (Fujimori *et al* 1987; Shen *et al* 1987; Yarmoff *et al* 1987; Sondericker *et al* 1987; Bianconi *et al* 1987; van der Marel *et al* 1988; Eskes and Sawatzky 1988; Nucker *et al* 1988; Sarma 1988; Sarma and Taraphder 1989; Mattheiss 1987; Haas 1989; Pickett 1989; McMahan *et al* 1988; Hybertsen *et al* 1989; Stechel and Jennison 1990; Oles and Zaanen 1989) for the high-T_c cuprates. While the detailed model is specific to the high-T_c cuprates, the results obtained are general enough to be relevant for any transition metal compound, just as in the case of the ZSA diagram (Zannen *et al* 1985).

In figure 1 we present the (metal-insulator) phase diagram obtained by us in the $U_{dd} - \Delta'$ plane, where $\Delta' = \Delta + 4t_{pp}$ is the bare Cu $3d_{x^2-y^2}$ level measured from the *bottom* of the O–$2p$ band. The solid line in the diagram, where the bandgap E_g vanishes, separates the metallic phase from the insulating phase when $U_{pp} = 0$ and $U_{pd} = 0$. The results with $U_{pp} \neq 0$ and $U_{pd} \neq 0$ are not qualitatively different and will

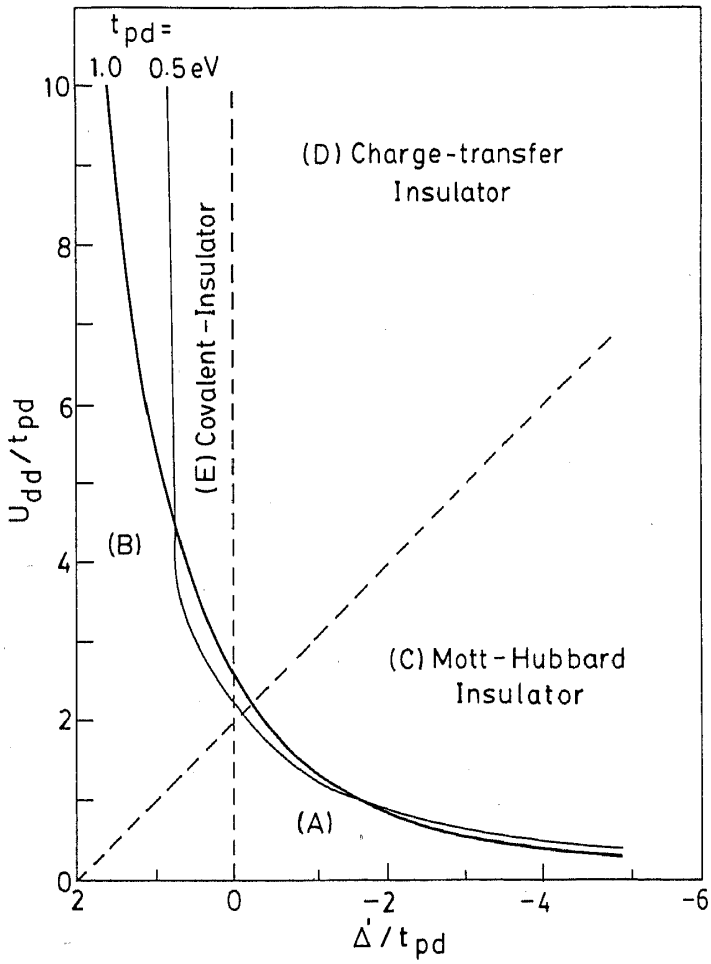

Figure 1. The "phase" diagram for the 5-band Hubbard model using the Hartree-Fock approximation including spiral SDW, to be compared with the corresponding ZSA diagram. Note the new "covalent insulator" region. The thick and the thin lines are $t_{pd} = 1{\cdot}0$ and 0·5 eV respectively. The dashed lines are the approximate boundaries between the different regions.

be presented elsewhere. We have marked five approximate regions A through E in figure 1 to denote different ground state behaviours. A is a normal *d*-band metal formed for small U_{dd}-values, and B is a mixed-valent metal formed due to the overlap of the partially filled *d* and oxygen *p*-band. The other three regions C, D and E represent insulating planes. Region C is the Mott-Hubbard insulator where E_g scales primarily with U_{dd} and Δ' has little effect on it. Region D is the charge transfer insulator where E_g is largely determined by Δ', with only small effect from U_{dd}. These four regions are the same as those discussed (Zannen *et al* 1985) in connection with the ZSA phase diagram and thus, are not discussed any further here. While we have obtained our results in a Hartree-Fock mean field approximation, but *for the full lattice problem*, ZSA used an impurity approach. The latter may not be so bad for rare-earth systems where the hybridization strengths are small and the valences are closer to integral values. But we believe that in the present strongly mixed-valent context, with large t_{pd}, our approach is superior (especially in view of the results discussed below).

The last region, marked E, has not been identified earlier. It has various interesting

properties that distinguish it from the charge-transfer region (D). First, it is found that the properties, for example the bandgap and the phase boundaries, in this region do not merely scale with U_{dd}/t_{pd} or Δ'/t_{pd}, but depend explicitly on the value of t_{pd} (cf. the phase-boundaries for $t_{pd} = 1{\cdot}0$ eV and $t_{pd} = 0{\cdot}5$ eV in figure 1). Note that in the absence of t_{pd}, the metal-insulator phase boundary would have been $\Delta' = 0{\cdot}0$ (dashed line in figure 1). Hence, the insulating phase obtained for $\Delta' > 0$ is actually driven by the finite t_{pd} values in presence of a large U_{dd}, whereas the charge-transfer insulator region D is insulating because of the bare charge-transfer gap, and even for vanishingly small t_{pd}-values. For this reason the d-count in region E will always be strongly nonintegral. Keeping in mind these distinct features, we term this region as a "covalent insulator". Here we would like to point out that the calculations presented by Zaanen *et al* (1985) also contain a region with finite gap for $\Delta' > 0$; however the authors had disregarded this feature in the phase diagram suspecting it to be an artefact of the impurity approximation. But the existence of this region also in the present multiband Hubbard model calculations establishes this region. We expect that compounds of late transition elements that exhibit metal-insulator transitions (as functions of parameters that do not change carrier concentration), e.g. $LaCoO_3$, NiS, $NiS_{2-x}Se_x$ etc. will belong to this covalent insulator regime in the insulating phase.

Band structure calculations have indicated (Mattheiss 1987; Haas 1989; Pickett 1989) that the insulating parent compounds of the high-T_c cuprates invariably exhibit a large t_{pd} and a small Δ, (in fact a positive Δ'). This is an indication that the high-T_c cuprates probably are in the vicinity of the covalent insulator regime, close to a metal insulator transition. Analysis of core-level photoemission data using the cluster approximation (Fujimori *et al* 1987; Shen *et al* 1987; Yarmoff *et al* 1987; Sondericker *et al* 1987; Bianconi *et al* 1987; van der Marel *et al* 1988; Eskes and Sawatzky 1988; Nucker *et al* 1988; Sarma 1988; Sarma and Taraphder 1989; Sarma and Ovchinnikov 1990) has always indicated a small Δ-value, which, in presence of oxygen p-band formation, will surely lead to a covalent description. In fact a recent analysis (Sarma and Ovchinnikov 1990) of all core-level data available from high-T_c and related compounds showed that a small Δ and substantially nonintegral d-occupancies are inevitable conclusions. An analysis of the core-level photoemission spectra beyond the cluster model has also shown (Sarma 1988; Sarma and Taraphder 1989) that La_2CuO_4 belongs to the covalent insulator regime with positive Δ', large t_{pd} and large U_{dd}. Analysis of the valence band spectra (Fujimori *et al* 1987; Shen *et al* 1987; Yarmoff *et al* 1987; Sondericker *et al* 1987; Bianconi *et al* 1987; van der Marel *et al* 1988; Eskes and Sawatzky 1988; Nucker *et al* 1988) also have consistently indicated that Δ' is rather small. Thus, it appears that the insulating ends of the high-T_c cuprates indeed belong to the class of covalent insulators or are close to the boundary. We find that for reasonable values of U_{dd} ($> 8{\cdot}0$ eV), we need a Δ value of about $-3{\cdot}0$ eV (i.e. $\Delta' = -1{\cdot}0$ eV) to obtain the experimental bandgap ($\sim 2{\cdot}0$ eV). This indicates that these cuprates belong to the boundary region between the covalent and charge-transfer insulators.

We are confident that the new insulating phase survives the inclusion of fluctuation corrections beyond the Hartree-Fock approximation. For, the presence of the charge gap suppresses charge fluctuations, and spin fluctuations are characterized by a small energy scale J_{eff} (see later) and cannot in any way destroy the large charge gap. In further support of our conclusion we have carried out exact calculations of finite clusters

with periodic boundary condition within a minimal model of a CuO chain including $d_{x^2-y^2}$ orbital on Cu and p_σ orbitals on the oxygens. The calculations for 4 and 6 Cu atoms clearly establish that there is a conductivity gap, E_g $(=2E(n)-E(n+1)-E(n-1))$ for parameter values belonging to the covalent insulator regime. Thus, for $t_{pd}=1{\cdot}0$, $t_{pp}=0{\cdot}5$, $U_{dd}=10$ and $\Delta'=0{\cdot}1$ eV, we obtain the conductivity gaps as 0·44 and 0·60 eV for 4 and 6 atoms in the chain. The slight increase in the conductivity gap is perhaps due to the greater stability of the $(4n+2)$ (n integer) system at half-filling due to aromaticity. While we cannot perform exact calculations for larger sized systems due to computational restrictions it is however possible to do a finite size scaling to arrive at a reasonable estimate of E_g in such a system. In this we follow the method of Fourcade and Spronken (1984) whose analysis for similar sizes gave very good extrapolated results in close conformity with the exact results in the case of a single-band one-dimensional Hubbard model. Following this method, we obtained conductivity gap of 0·73 eV for the infinite lattice, lending strong support to the existence of the covalent insulator regime. The Hartree-Fock treatment of the same model yields a conductivity or charge gap of 0·69 eV.

The above analysis of the electronic structure of the insulating cuprates imply a strongly nonintegral d-occupancy in complete disagreement with the ionic model proposed on the basis of various magnetic measurements. Hence in the following we examine the magnetic data vis-a-vis the results obtained in the present calculations.

The Hartree-Fock ground state obtained by us is a 2-sublattice antiferromagnet throughout the insulating range. With reasonable values of U_{dd} (5–10 eV), t_{pd} (1·0 eV), $t_{p_xp_x}$(0·19 eV) and (0·5 eV), and within the covalent insulator regime (including the boundary region, i.e. $\Delta' \geqslant -1$ eV) we find typical d-occupancies between 0·35 and 0·7. The Cu sites are found to have a moment between 0·3 and 0·66 μ_B, to be compared with the observed moments in the insulating compounds (about 0·4 μ_B for La_2CuO_4 (Vaknin *et al* 1987; Mitsuda *et al* 1987; Huse 1988) and 0·65 μ_B for $YBa_2Cu_3O_6$ (Tranquada *et al* 1988a, b; Li *et al* 1988)). We take the point of view that the reduction of the observed moment from the ionic value of μ_B (for Cu^{2+}) comes partly from the covalence or hybridization effects, as discussed above, and partly from the quantum fluctuations (Chakravarty *et al* 1988; Horsch and van der Linden 1988; Sinha 1990; Birgeneau and Shirane 1989). We also find that the moment on the oxygen sites is exactly zero, and hence the form-factor of the moment is entirely that of a Cu^{2+} ion, in spite of a large covalency. In fact, the zero moment at oxygen sites is a consequence of the symmetry present in the problem. For, in the presence of the antiferromagnetic order of the Cu moments, the up- and down-spin orbitals at every O-site are degenerate and equally populated. Within our model, the same conclusion in fact holds for each lobe of the oxygen orbitals. For, the oxygen p-orbitals (that point to the neighbouring Cu sites) are symmetric with respect to the oxygen site, whence each lobe of a given spin-orbital has the same net charge. This ensures that the moments associated with the up- and down-spin orbitals in each lobe (and not just at each site) cancel exactly. Thus, the experimental observation (Sinha 1987, 1990; Freltoft *et al* 1988; Birgeneau and Shirane 1989) that each lobe of the oxygen orbitals has a negligible moment can be understood within the strongly covalent description, and does not require an ionic description of the compounds. The neutron scattering data has been interpreted (Sinha 1987, 1990; Freltoft *et al* 1988; Birgeneau and Shirane 1989) as indicating a finite, though small ($<0{\cdot}1\ \mu_B$) moment on each lobe of the oxygen orbitals. We could explain this if we were to extend our model to include processes that permit a spin dependent

distortion of the lobes of the O–p orbitals (e.g., a mixing of O–3s orbitals with Cu $d_{x^2-y^2}$, which would then cause them to mix in a spin-dependent way with the O–p orbitals). Preliminary calculations including the O–3s indeed show such a trend.

The Hartree-Fock theory, with the inclusion of spiral states, provides a clean, nonperturbative way (i.e., one which does not assume weak or strong coupling) of mapping the multiband Hubbard model at "half-filling" to the Heisenberg model, and hence of calculating J_{eff}. We first obtain the energy E of the Hartree-Fock spiral SDW state (Chakravarty *et al* 1988; Horsch and van der Linden 1988), with a frozen spin configuration $\langle S_i^+ \rangle = b_0 e^{i\mathbf{Q}\cdot r_i}$, as a function of its wave-vector $\mathbf{Q}$ around $\mathbf{Q}_0$ (π, π) of the antiferromagnetic ground state, by minimizing the energy with respect to b_0. This energy spectrum $E(\mathbf{Q})$ is then compared with the energy of such a state for the spin-1/2 Heisenberg model (the two energies have the same $\mathbf{Q}$ dependence) to obtain J_{eff}. We have evaluated the J_{eff} for the insulating phase as a function of Δ for two constant values of U_{dd} (5 and 10 eV) as well as a function of U_{dd} for two fixed values of Δ ($-3{\cdot}0$ and $-6{\cdot}0$ eV). The results are shown in figures 2a and b. In the covalent insulator regime, the calculated J_{eff} is close to 0·2 eV. For reasonable values of U_{dd} ($\geqslant 8{\cdot}0$ eV) J_{eff} at $\Delta = -3{\cdot}0$ eV ($\Delta' = -1{\cdot}0$ eV) is about 0·14 eV in excellent agreement with the experimental value of J_{eff} (0·16 eV) for La_2CuO_4, indicating that the system

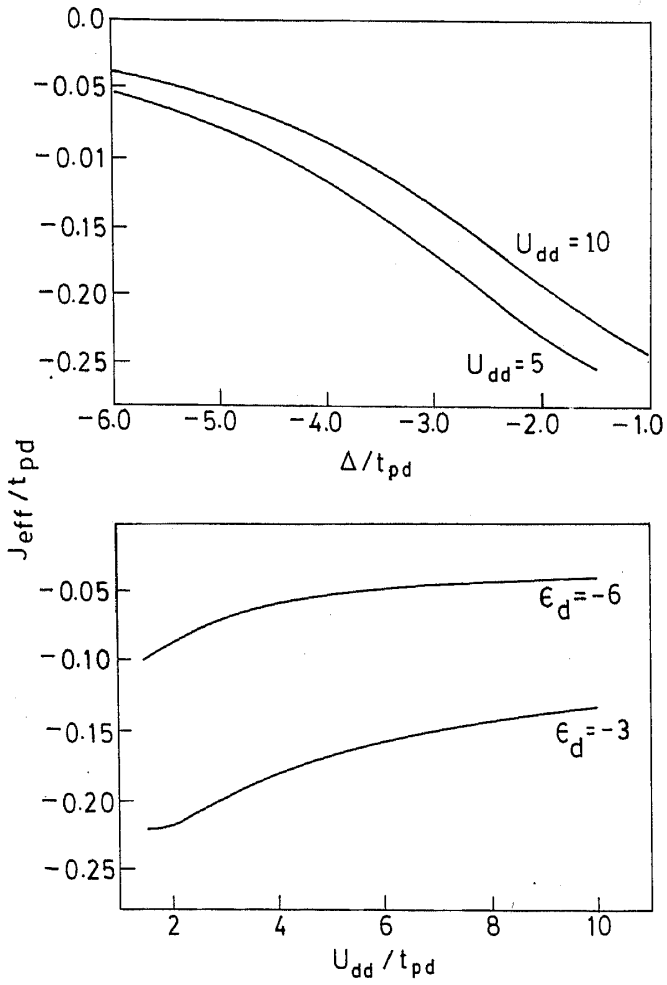

Figure 2. J_{eff}, the effective antiferromagnetic exchange interaction for the 5-band Hubbard Model, obtained using the Hartree-Fock spiral SDW states.

is at the boundary region between the charge transfer and the covalent insulator regimes.

Multiband Hubbard models of the type discussed here have also been treated by slave-boson mean-field theories (MFT) in the $U \to \infty$ limit (Newns and Pattnaik 1989). An essentially similar MFT for the finite U case has been presented by Balseiro *et al* (1989), who also obtained a ZSA-like phase diagram at half-filling. However, these treatments have the limitation (Pattnaik *et al* (to be published)) that the insulating phase at half filling is obtained as a zero slave-boson amplitude ($b = 0$) solution (implying infinite effective mass for the charge carriers); and for the choices of parameters in the model for which $b = 0$ at half filling, the effective masses away from half-filling are too large. Furthermore, the covalent insulator phase is missed in these treatments, and the $b = 0$ solution implies an ionic picture of the insulator which we have argued as in conflict with high energy spectroscopy data. As suggested in (P C Pattnaik *et al* (to be published)), we believe that, if the parameters are such that the slave-boson MFT is relevant well away from half-filling, then there is most likely a phase transition to a groundstate of the type discussed here sufficiently close to half-filling.

In conclusion, we have obtained a phase diagram separating the metallic and insulating phases at half-filling within a Hartree-Fock mean-field theory of the multiband Hubbard model. We have discussed the characteristics of the different phases and have characterized a new insulating region which we term "covalent insulator". We have also shown that a reasonably consistent understanding of the apparently conflicting experimental results from the high-energy spectroscopic studies and magnetic studies can be achieved within the present treatment. Specifically we have indicated that the ground state electronic structure of these compounds is highly mixed-valent. We have also carried out an extension of the treatment presented here to the case with finite doping (i.e. away from half-filling) which permits a clean, nonperturbative discussion of the mapping to the t-J model which holds for a wider range of parameters than the treatment of Zhang and Rice (1988), Shastry (1989). Furthermore we find that the states which are populated due to hole-doping have a large oxygen p character and that there is a marked asymmetry between electron and hole doping at the level of detailed modelling. The details will be discussed elsewhere (Seva Nimkar *et al* (to be published)).

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