Origin of charge density wave in the coupled spin ladders in \(\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}\)

Krzysztof Wohlfeld,1,2 Andrzej M. Oleś,1,2 and George A. Sawatzky3

1Marian Smoluchowski Institute of Physics, Jagellonian University, Reymonta 4, PL-30059 Kraków, Poland
2Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany
3Department of Physics and Astronomy, University of British Columbia, Vancouver B. C. V6T-1Z1, Canada

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We formulate a d-p multiband charge transfer model for \(\text{Cu}_2\text{O}_2\) coupled spin ladders, relevant for \(\text{Cu}_2\text{O}_3\) plane of \(\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}\), and solve it using Hartree-Fock approximation. The results explain that (i) the charge density wave (CDW) with its periodicity dependent on doping is stabilized by purely electronic many-body interactions in a single spin ladder and (ii) the inclusion of the interladder interactions favors (disfavors) the stability of the CDW with odd (even) periodicity, respectively. This stays in agreement with recent experimental results and suggests the structure of the minimal microscopic model which should be considered in future more sophisticated studies.

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\(\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_{24}\text{O}_{41}\) (SCCO) is a layered material with two distinctly different types of two-dimensional (2D) copper oxide planes separated by \(\text{Sr}/\text{Ca}\) atoms (i) the planes with almost decoupled \(\text{Cu}_2\text{O}_2\) chains and (ii) the \(\text{Cu}_2\text{O}_3\) planes formed by \(\text{Cu}_2\text{O}_5\) coupled ladders (see Fig. 1). The latter ones exhibit the non-BCS superconducting (SC) phase for \(x = 13.6\) in SCCO under pressure larger than 3 GPa or a spin-gapped insulating charge density wave (CDW) state in broad range of \(x\) and under normal pressure. By means of the resonant soft x-ray scattering it was found that this CDW is driven by many-body interactions (presumably just Coulomb on-site interactions since the long-range interactions are screened in copper oxides), and it cannot be explained by a conventional Peierls mechanism. Hence, the observed competition between the CDW (also referred to as the “hole crystal” due to its electronic origin) and SC states in spin ladders resembles the one between stripes and the SC phase in \(\text{Cu}_2\text{O}_2\) planes of a high \(T_c\) superconductor (HTS), which makes the problem of the origin of the CDW phase in SCCO both generic and of general interest.

Furthermore, recently it has been found that the only stable CDW states are with period \(\lambda = 5\) for \(x = 0\), and with period \(\lambda = 3\) for \(x = 11\) (and with a much smaller intensity for \(x = 10\) and 12), while such a CDW order has not been observed for \(1 \leq x \leq 5\). These striking results, which contradict the previous suggestions that the CDW order occurs in the entire range of \(0 \leq x < 10\), need to be explained by considering hole density per \(\text{Cu}\) site increasing with \(x\). Recently, a much higher hole density in the ladders was reported than believed before, i.e., \(n_h = 1.20\) (number of holes/Cu ion) for \(x = 0\), \(n_h = 1.24\) for \(x = 4\), and \(n_h = 1.31\) for \(x = 11\). The aim of this paper is to explain theoretically these puzzling features of the CDW order using the above hole density.

On the one hand, it is widely believed that a two-leg spin ladder described by the \(t-J\) model captures the essential physical properties of SCCO. The idea that merely on-site Coulomb interactions could lead to charge ordering was already suggested by White et al. using density matrix renormalization group (DMRG) — they found that a CDW of period \(\lambda = 4\) is the (possibly spin gapped) ground state at \(n_h = 1.25\). It is, however, quite remarkable that such a CDW has not been observed. On the other hand, the validity of the \(t-J\) model for \(\text{Cu}_2\text{O}_5\) coupled spin ladders is not obvious since: (i) unlike the \(\text{Cu}_2\text{O}_2\) plane of a HTS, a single \(\text{Cu}_2\text{O}_5\) ladder lacks the \(D_{4h}\) symmetry making the Zhang-Rice (ZR) derivation of the \(t-J\) model questionable and (ii) \(\text{Cu}_2\text{O}_5\) spin ladders are coupled through the on-site Coulomb interactions between holes in different \(O(2p)\) orbitals, so new interactions could arise.

This suggests that the multiband charge transfer model11 adapted to the \(\text{Cu}_2\text{O}_5\) ladder geometry, similar to those introduced earlier for \(\text{Cu}_2\text{O}_2\) planes12 or \(\text{Cu}_2\text{O}_3\) chains13 of HTSs, could be more appropriate to cap-

FIG. 1: (Color online) Schematic picture of two coupled \(\text{Cu}_2\text{O}_2\) ladders (white and grey) with a CDW order of period \(\lambda = 3\) and \(\lambda = 4\). The \(\text{Cu}_2\text{O}_2\) unit cell with two \(3d_{\pm x-y^2}\), three \(2p_x\), and two \(2p_y\) orbitals is indicated by dashed line. The arrows stand for hole spins in \(\text{Cu}\) and \(O\) orbitals, with their (large) small size corresponding to \(+1.0\) (+0.25) hole charge. The ovals show ranges with enhanced hole density in the CDW phase. The dotted ovals in the grey ladder of (b) show the two possible degenerate states, see text.
tured the essential physical phenomena. As parameters the charge transfer model includes: the energy for oxygen 2p orbital $\Delta$, the $d$-$p$ hopping $t$ between the nearest neighbor Cu and O sites, and the on-site Coulomb repulsion $U$ ($U_p$) on the Cu (O) sites, respectively. By solving this model in the Hartree-Fock (HF) approximation, we investigate whether: (i) the Coulomb on-site repulsion stabilizes the observed CDW of the respective period $\lambda$ for a given number of holes $n_h$, (ii) the different stability of the CDW order with odd/even period follows, and (iii) the ZR singlets are formed in the spin ladder geometry.

We consider the charge transfer model in hole notation

$$\mathcal{H} = \Delta \left( \sum_{j \in R,L,\sigma} n_{j\sigma} + \varepsilon \sum_l n_{l\sigma} \right) + \left\{ \sum_{m,j \in R,L,\sigma} t_{mj} d_{m\sigma}^\dagger x_{j\sigma} \right. $$

$$+ \sum_{m,j \in R,L,\sigma} t_{mj} d_{m\sigma} y_{j\sigma} + \sum_{m \in R,L,\sigma} t_{ml} d_{l\sigma} b_{i\sigma} + \text{H.c.} \right\} $$

$$+ U \left( m \in R,L \right) \sum_{m \in R,L,\sigma} n_{m\sigma} n_{m\sigmaL} + U_p \left( j \in R,L,\sigma \right) \sum_{j \in R,L,\sigma} n_{j\sigma} n_{j\sigma} $$

$$+ U_p \sum_{l \in b} n_{l\sigma} n_{l\sigmaL} + \left( 1 - 2 \eta \right) \sum_{j \in R,L,\sigma} n_{j\sigma} n_{j\sigmaL} + \left( 1 - 3 \eta \right) \sum_{j \in R,L,\sigma} n_{j\sigma} n_{j\sigma} $$

$$\left. \left( \left( \left( 1 - 3 \eta \right) \right) n_{j\sigma} n_{j\sigma} + \left( 1 - 3 \eta \right) n_{j\sigma} n_{j\sigma} \right) \right\},$$

(1)

where $|t_{mj}| = |t_{ml}| = t$, $\sigma = x, y$, and $\bar{\sigma} = -\sigma$ for $\sigma = \uparrow, \downarrow$. The parameter $\eta = \Delta/U_p = 0.2$ stands for a realistic value of Hund’s exchange ($U_p$ is the intra-orbital repulsion) and $\varepsilon = 0.92$ yields the correct orbital energy at bridge positions. The model of Eq. (1) includes seven orbitals per CuO$_2$ ladder unit cell (see Fig. 1): two Cu(3d$_{x^2-y^2}$) orbitals on the right or left ($R$ or $L$) leg, two O(2p$_y$) orbitals on the $R/L$ leg, two O(2p$_x$) orbitals on the rung of the ladder. We emphasize that the last two terms in Eq. (1) account for interladder interaction – the holes within two different orbitals on a given oxygen ion in a leg belong to two neighboring ladders (shown as white/grey orbitals in Fig. 1), and are described by charge operators $n_{jx(y)\sigma}$ with/without tilde sign in Eq. (1). This makes the model Eq. (1) implicitly 2D, though the band structure is one-dimensional (1D) when the interoxygen hopping $t_{pp'}$ is neglected (in fact $t_{pp'} \ll t$). A priori, $\tilde{n}_{j\sigma}$ should be treated as particle number operators belonging to the Hilbert subspace of the neighboring ladder, resulting in a 2D many-body problem. Here we simplify it and treat $\rho_{j\sigma}$ as “external” classical fields adjusted to the symmetry of the CDW state, which are self-consistently determined within the HF approximation.

We have solved the Hamiltonian (1) for various values of the model parameters $\{U, \Delta, U_p\}$, and for three different hole densities $n_h = 1.20, 1.25, 1.33$ (which correspond to the actual filling in SCCO in the range of $0 \leq x \leq 11$) using HF approximation, i.e., we decouple $n_{j\mu} n_{j\mu L} \rightarrow \langle \tilde{n}_{j\mu} \rangle \langle n_{j\mu L} \rangle + \langle \tilde{n}_{j\mu} \rangle \langle \tilde{n}_{j\mu L} \rangle - \langle \tilde{n}_{j\mu} \rangle \langle \tilde{n}_{j\mu L} \rangle$, where $\mu = d, x, y, b$. The ground state was found by diagonalizing the resulting one-particle Hamiltonian in real space for a single ladder with 60 unit cells, separately for spin up and spin down. The classical fields $\{\rho_{j\sigma}\}$ and $\{\langle \tilde{n}_{j\mu} \rangle\}$ were determined self-consistently with the initial values for these fields suggested by recent experiment, see Fig. 1. While a uniform spin density wave (SDW) is stable for $n_h = 1.0$, one finds a CDW superimposed on the SDW order for realistic hole densities $n_h \geq 1.20$. The stability of this composite order follows from the 1D polaronic defects in the SDW state. We limit the present analysis to the stability of this particular CDW phase, while we do not study here the possible competition with other phases.$^6$

For each state we evaluate the CDW order parameter

$$p = \sum_{i \in \text{rung}} \langle n_{id} + n_{ib} + n_{ix} \rangle - \frac{1}{\lambda - 1} \sum_{i \in \text{rung}} \langle n_{id} + n_{ib} + n_{ix} \rangle $$

$$+ \sum_{i \in \text{rung}} \langle n_{iy} \rangle - \frac{2}{\lambda - 2} \sum_{i \in \text{rung}} \langle n_{iy} \rangle,$$

(2)

Here and in what follows by “rung” we mean the “rung with enhanced hole density” which consists of seven O (four $y$, two $x$ and one $b$) orbitals and two Cu orbitals (see the ovals in Fig. 1). Hence, in both above definitions the mean values of the particle number operators are calculated for these rungs ($i \in \text{rung}$) or for all remaining sites ($i \notin \text{rung}$). Note that in the ideal CDW phase (shown in Fig. 1) $p = 2$ and $\sigma^2 = 0$, irrespectively of the actual period $\lambda$. We also introduce $\text{rung}$ hole densities on O and Cu sites

$$n_p \equiv \sum_{i \in \text{rung}} \langle n_{ib} + n_{ix} + n_{iy} \rangle, \quad n_d \equiv \sum_{i \in \text{rung}} \langle n_{id} \rangle.$$  

(4)

Similarly, magnetic order parameters are

$$m_p \equiv \left| \sum_{i \in \text{rung} \cap L} n_{ix} + n_{iy} \right| + \left| \sum_{i \in \text{rung} \cap R} n_{ix} + n_{iy} \right|,$$

(5)

$$m_d \equiv \left| \sum_{i \in \text{rung}} n_{id} \right|,$$

(6)

where the magnetization for orbital $\mu$ at site $i$ is $m_{i\mu} = \langle n_{j\mu} - \langle n_{j\mu} \rangle \rangle$. We recall that when holes on the rungs form two localized ZR singlets next to each other, then $n_d = m_d \approx 2, n_p \approx 2$, and $m_p \approx 1.5$, see Fig. 1.

First, we investigate the onset of the CDW phase in a single ladder of Fig. 1 by assuming $U_p = 0$. In the charge transfer regime (for $\Delta = 3t$ following Ref. 17) the CDW is stable already for $U \geq t$ with periods: $\lambda = 5$ for $n_h = 1.20$, $\lambda = 4$ for $n_h = 1.25$, and $\lambda = 3$ for $n_h = 1.33$ [Fig. 2(a)]. For higher values of the on-site Coulomb repulsion $U$, $p$ first increases quite fast irrespectively of the actual
FIG. 2: (Color online) Characterization of the CDW ground states obtained with $U_p = 0$ for increasing $U$ (left, $\Delta = 3t$) and $\Delta$ (right, $U = 8t$): (a), (b) CDW order parameter $p$, and (c), (d) ZR singlet dispersion $\sigma^2$, for $\lambda = 5, 4, 3$ shown by solid, dashed, and dotted lines, respectively; (e), (f) for $\lambda = 5$ charge (magnetization) in the rung on Cu sites shown by solid (dotted) line and on O sites shown by dashed (dashed-dotted) line, see Eqs. (4)–(6). The realistic values (Ref. 17) of $U/t = 5$ is indeed very small for all periods [Fig. 2(a)]. In general the dependence of $\Delta_{CDW}$ on $U$ qualitatively mimics the relation between $p$ and $U$ which suggests that the CDW gains stability when an insulating state is formed. Indeed, the electronic density of states $N(E)$ (inset of Fig. 3) shows well developed lower and upper Hubbard bands (LHB and UHB) separated by an oxygen band, with a small CDW gap in the latter band. Altogether, one finds that: (i) the Coulomb interaction $U$ can stabilize the CDW in the Cu$_2$O$_2$ ladders, (ii) the CDW phase can be viewed as an equidistant distribution of the ZR singlet states in the relevant parameter regime, and (iii) all of the stable periods (even and odd) behave similarly.

Next, we investigate the influence of the interladder coupling. At finite $U_p$ the "external" fields $\rho_{j\alpha} = \langle n_{j,\alpha} \rangle$ in Eq. (1) contribute and were self-consistently determined by iterating the HF equations. Thereby, the symmetry of the CDW state was chosen in such a way that the rungs were translated by $\lambda$ Cu-O lattice constants ($\lambda$ odd) in the neighboring ladders to maximize the distance between them (Fig. 1), which minimizes the HF energy. For even $\lambda = 4$ the numerical calculations performed with the realistic parameters for Cu$_2$O$_2$ ladder ($U = 8t$ and $\Delta = 3t$) confirmed that two states shown by dotted ovals in Fig. 1 are degenerate, as expected. The effect of the interladder interaction $U_p$ was identified by comparing the ground states derived separately in two cases: (A) with $\rho_{j\alpha} = 0$, i.e., using only the (intraorbital) repulsion between oxygen holes on the considered ladder; (B) by implementing the "external" fields $\{\rho_{j\alpha}\}$ calculated self-consistently, i.e., including both the intraorbital and interorbital Coulomb repulsion between...
FIG. 4: (Color online) The CDW ground state for increasing $U_p$: (a) CDW order parameter $p$ and (b) ZR singlet dispersion $\sigma^2$, for $\lambda = 5, 4, 3$ shown by solid, dashed, and dotted lines (squares, triangles, and circles) in case A (B), see text; (c) for $\lambda = 3$ charge on Cu (O) sites in the rung by solid (dashed) line in case A and by squares (circles) in case B, see Eq. (4); (d) for $\lambda = 4$ charge in different $y$ orbitals ($n_y$, $n_{y1}$, and $n_{y2}$, shown by diamonds, triangles down, and up) in the rung in case (B), see text. Vertical lines mark the realistic value (Ref. 17) of $U_p = 3t$. Parameters: $\Delta = 3t$, $U = 8t$.

holes on oxygen sites.

One finds that in case A the CDW order parameter $p$ decreases in a similar way for all periods, cf. Fig. 4(a), as well as for even period ($\lambda = 4$) when the interladder coupling is switched on (case B). Remarkably, a qualitatively distinct behavior is found for odd periods – here the interladder coupling supports the onset of the CDW phase and the order parameter either saturates or even increases with increasing strength of the on-site repulsion $U_p$ (as for $\lambda = 3$), see also Fig. 4(c). In fact, the interladder coupling enhances the hole density in the rungs.

Another striking effect is the qualitatively distinct behavior of the ZR dispersion $\sigma^2$ for odd and even periods, see Fig. 4(b). While for period $\lambda = 4$ switching on the interladder coupling (B) drastically increases $\sigma^2$ with respect to the single ladder case (A), the results are precisely opposite for odd periods $\lambda = 3, 5$. Furthermore, this increase of $\sigma^2$ with $U_p$ in case (B) is large for even period – its value $\sim 0.1$ found for large (but still realistic) $U_p \sim 3.5t$ is comparable to the value of the ZR dispersion for a single ladder with $\Delta \sim t$ [Fig. 2(d)], where we do not expect stable ZR singlets. This large increase of $\sigma^2$ in this case follows from the geometrical frustration of the CDW state, as for even periods the two $y$ orbitals in the same rung are not equivalent [one of them (say $y1$) is closer than the other one (say $y2$) to the rung in the neighboring ladder], as shown in Fig. 1(d). We have also verified that the mean hole density $n_y = \frac{1}{2}(n_{y1} + n_{y2})$ almost does not change when the interladder coupling is switched off (not shown).

Thus, we conclude that the interladder interaction: (i) supports the CDW states with odd periods $\lambda = 3, 5$ and slightly disfavors the frustrated CDW state with even period $\lambda = 4$, (ii) destabilizes the homogeneous ZR-like distribution of holes in the rungs for period $\lambda = 4$. In contrast, experimentally one finds that in SCCO with $x = 4$ ($n_h \sim 1.25$) the holes are distributed isotropically over O sites in the rung, but the CDW is unstable. We suggest that, since in reality the ZR singlets are much more rigid than the present classical ZR states (as the energy gain due to quantum fluctuations and phase coherence are not captured in these states) and in reality the system is less prone to order than in the HF approximation, the interladder interactions in the model Eq. (11) would indeed destabilize the CDW with even period.

In summary, we have shown that the CDW combined with the SDW can be stabilized in the spin ladders of SCCO merely due to on-site Coulomb repulsion on Cu sites. The presented results explain the experimentally observed CDW states with odd periods for $x = 0$ and $x = 11$, and provide a theoretical explanation why the CDW states with even period could not be observed. In addition our results suggest that an extension to the two leg ladder $t$-$J$ model plus an interladder Coulomb repulsion to represent the physics described in our study. We are looking forward to future studies of this kind using the much more sophisticated DMRG-like methods.

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Note added in proof: In contrast to the present charge transfer model, Roux et al. have found that the CDW state is unstable for other hole densities than $n_h = 1.25$ and $n_h = 1.50$ in their recent DMRG and bosonization studies of the $t$-$J$ model for the single ladder with $1.0 < n_h < 1.5$.

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