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Electronic structure and magnetism of the "pseudo-ladder" compounds ACu_2O_3, A = Ca,Mg

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Abstract

The electronic structure of ACu_2O_3 compounds [A = Ca,Mg] has been calculated within the local density approximation (LDA) and compared with recent magnetic neutron scattering data. Both compounds deviate markedly from the usual $pd\sigma$ cuprate picture. Strong interlayer exchange is found to be responsible for the missing spin gap generic for ideal two-leg ladders (TLL). Hence, they can be modelled as weakly coupled anisotropic bilayers.
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Cuprate based spin-ladder compounds have been intensively studied in last years [1]. Two-leg pseudo-ladders (TLPL), such as $LaCuO_{2.5}$ and the title compounds, constitute a seemingly related group. However, their magnetic ordering at low temperatures T suggests a position in between highly anisotropic nearly 1D corner-shared antiferromagnetic (afm) CuO_3 chain compounds and less anisotropic materials with more or less comparable interacting strength of CuO_4 plaquettes in all directions. In contrast to weakly interacting TLL with a spin gap and $\mu = 0$ in a spin-liquid ground state, strongly interacting TLPL exhibit 3D Néel order at $T < T_N$ with $\mu \neq 0$, like unfrustrated single-chain compounds without spin-Peierls transition. Here, we will give some insight in the origin of the behavior of known TLPL which can be first characterized as: $LaCuO_{2.5}$ ($T_N \approx 117 \pm 17$ K, $\mu = 0.5 \pm 0.2\mu_B$ [2]), $MgCu_2O_3$ ($T_N \approx 95$ K, $\mu = 0.32 \pm 0.02\mu_B$ [3]), and $CaCu_2O_3$ ($T_N \approx 27$ to 25 K, $\mu = 0.2 \pm 0.07\mu_B$ [4,5]). Below T_N the isostructural title compounds (which can be viewed as stacked buckled Cu_2O_3 planes of the prototypical planar TLL $SrCu_2O_3$ system (see Fig. 1)) show besides common afm period doubling along the chain (*b*-axis), *different* magnetic structures in *c* and *a*-directions: ferromagnetic (fm) ordering in *c*-direction and on the kinked PL-rungs for A = Mg, and afm ordering for A = Ca. Along the *a*-axis afm period doubling and/or an incommensurate spiral-type wave near 3/7 commensurability, respectively, have been observed. Ongoing from $SrCu_2O_3$ with straight rungs to strongly "kinked" rungs in $MgCu_2O_3$ and beyond, with decreasing buckling (Cu–O–Cu bond) angle Θ there is a transition from a superexchange dominated afm to a fm rung exchange (governed by Hund's rule coupling at the intermediate O of the rung centre) approaching $\Theta = 90°$, provided the crystalline field caused splitting of onsite energies for differently oriented O 2p orbitals is small. Comparing the observed magnetic structures one concludes that this transition occurs at a critical Θ_c in between those of A = Ca and A = Mg. Near Θ_c the rung exchange J_r should be small.

In order to quantify the afm contributions we analyzed the dispersions of bands near the Fermi energy E_F

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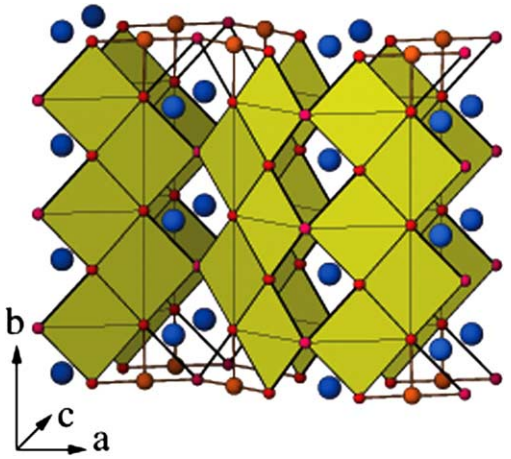

Fig. 1. The crystal structure of $CaCu_2O_3$ ($MgCu_2O_3$). CuO_2 zigzag chains (legs) running along the b-axis are alternatively tilted by nearly 28.6° (37.5°) forming up and down buckled TLPL with "kinked" rungs $\| a$, and stacking along c.

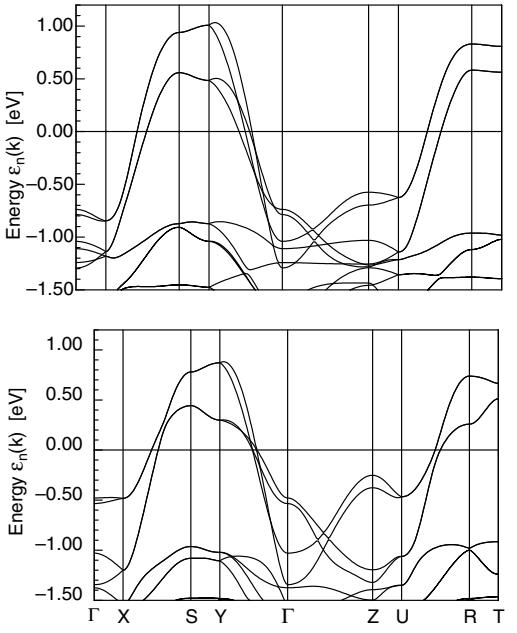

Fig. 2. LDA band structure of ACu_2O_3 near E_F. A = Ca (Mg) upper (lower) panel.

obtained from full potential LDA calculations with the FPLO code. For details see Ref. [6]. A justification for the LDA based electronic structure is the reasonable description of the unoccupied electronic states for A = Ca probed recently by polarization dependent X-ray absorption spectroscopy [6]. Since the exchange along a is frustrated due to the shift of the nearest neighbor TLPL by $0.5b$ along b, only a weak afm exchange J_a results (compare e.g. the weak dispersions along ΓX in both panels of Fig. 2) from the unshifted second neighbor pseudo-ladder. Ignoring that frustration as well as a weak rung exchange J_r as first suggested in Ref. [4], this leads to a spatially anisotropic Heisenberg problem with a hierarchy of exchange integrals $J_b \gg |J_c| \gg J_a$. Hence, at $T = 0$ the magnetic order can be described within an effective 2D-model for interacting chains in the b–c plane. With Sandvik's 2D mean-field result for μ (in units of μ_B) at $T = 0$ [7]

$$\mu = 0.273 g_L \sqrt{1/\gamma}(1 + 0.095/\gamma) \ln^{1/3}(1.3\gamma), \qquad (1)$$

we estimate from the mentioned μ-values the exchange anisotropy: $\gamma \equiv J_b/|J_c| \approx 6 \pm 0.9$ (A = Mg) and 20 (50 to 9) (A = Ca), where a Landé factor $g_L = 2.2$ has been adopted. With $J_b = 1540$ and 2000 K [3,4] we arrive at $J_c(\mathrm{Mg}) = -22.1$ meV and $J_c(\mathrm{Ca}) = +8.6$ meV with signs chosen according to the experimental magnetic structures. Note also the approximate relation $T_N \propto |J_c|$, typical for quantum spin chains [8,9]. We decompose the J_cs into afm and fm contributions. From effective transfer integrals t_z derived from the dispersions parallel to c (ΓZ in Fig. 2) and a typical Hubbard $U = 4$ eV, one arrives at $J_{c,\mathrm{afm}}(\mathrm{Ca}) = 4\, t_z^2/U = 17.6$ and 36.6 meV for A = Mg. Then markedly different empirical fm contributions result: $J_{c,\mathrm{fm}} = -58.7$ for A = Mg and −9.4 meV for A = Ca. A similar total fm interladder coupling of about −10 meV has been estimated for $LaCuO_{2.5}$ [10]. Mixed crystals $Ca_{1-x}Mg_xCu_2O_3$ allow a smooth transition between the two limiting crystal structures. In our view, at some critical concentration x_c (buckling angle) J_c changes its sign. Near x_c the ideal TLL picture with vanishing $T_N(x)$ and $\mu(x)$ might be retained, if $J_a \ll |J_r|$ holds. Otherwise, minima of $T_N(x)$ and $\mu(x)$ are expected. Notably, a shallow minimum of T_N has been reported for $x \approx 0.2$ [5].

To summarize, the magnetism of both title compounds is governed by sizable exchange in c-direction which destroys the usual TLL-picture.

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