Orbital distribution of holes in the linear chain cuprate Sr$_2$CuO$_3$

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The electronic structure of the prototypical linear chain cuprate Sr$_2$CuO$_3$ is studied theoretically by exact diagonalizations (ED) for clusters within the framework of generalized pd-models as well as by bandstructure calculations. From the 4- and 7-band analysis of the Sr$_2$CuO$_3$ band structure, enhanced values of Cu-O transfer integrals have been derived. The results are compared with polarization dependent O 1s x-ray absorption spectroscopy measurements (XAS) [1]. From the observed anisotropy of the XAS intensities significantly enhanced values of the intersite Coulomb interactions $V_{pd}$ and $V_{dd}$ compared with typical layered cuprates can be deduced. Keywords: Electronic structure, Intersite Coulomb interaction, One-dimensional cuprates,

1. INTRODUCTION

Sr$_2$CuO$_3$ is the best known realization of a spin-$\frac{1}{2}$ antiferromagnetic Heisenberg chain, [2] showing what is possibly a record value for the nearest neighbor (n.n.) exchange parameter $\tilde{J}$ 200 to 260 meV, which significantly exceeds the corresponding values in the 2D cuprates of 130 to 150 meV. The origin for this unexpected high $\tilde{J}$ value remains unclear and its elucidation requires a detailed knowledge of the underlying electronic structure. It is the aim of our analysis to contribute to the understanding of these chains and to propose a possible scenario of the large $\tilde{J}$.

2. MODEL

Starting from a detailed multi-band analysis of LDA-LCAO band structure calculations, we show which orbitals are mainly involved in low energy excitations and yield insight in the magnitude of the corresponding transfer integrals. We shall describe the chain cuprates under consideration by the extended 4-band $dp$ Hubbard-model written within standard notation (see also Fig. 1):

$$H = \sum_i \epsilon_i \hat{n}_i + \sum_{<i,j>,s} t_{ij}(c_{i,s}^\dagger c_{j,s} + \text{H.C.}) +$$

$$\sum_i U_{ii} \hat{n}_{i\uparrow} \hat{n}_{i\downarrow} + \sum_{<i,j>} V_{ij} \hat{n}_{i\uparrow} \hat{n}_{j\downarrow}. \quad (1)$$

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Figure 1. Sketch of the cluster modeling the CuO$_3$ chains in Sr$_2$CuO$_3$ within the pd-Hamiltonian (Eq. (1)) adopted. Upper half: The periodic clusters considered here. Lower half: On-site energies for the two inequivalent oxygen sites.
3. RESULTS AND DISCUSSION

According to the orbital analysis [3] three orbitals contribute more than 94\% to the total density of states of the antibonding band, namely the O(1) 2p\_z and O(2) 2p\_y as well as the Cu 3d_{2z^2-r^2} states. Hence, the usual pd-model with one orbital per site but extended to two nonequivalent oxygen sites per unit cell is a good first approximation. Our LDA-LCAO transfer integrals derived from a 7-band fit [3] read: \( t_{\text{pd}} = 1.57 \text{eV}, t_{\text{pd}} = 1.8 \text{eV}, t_{\text{pp}} = 0.62 \text{eV} \). We regard them as a reasonable estimate (from above) of the hopping parameters which enter Eq. (1).

Calculating with the aid of exact diagonalizations and Monte Carlo calculations the occupation numbers \( n_{\text{pi}} \) at the O(i) sites changing systematically the most unknown Hamiltonian parameters \( \Delta_{\text{pp}} \) and \( V_{\text{pd}} \), we reproduced the experimental anisotropy of the XAS-intensities [1] \( R = 2n_{\text{p2}}/n_{\text{p1}} = 1.22 \),

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as shown in Fig. 2. If reasonable values for \( \Delta_{\text{pp}} \approx 0.5 \) to 0.75 eV are adopted, Eq. (2) can be satisfied, only if a significantly enhanced intersite Coulomb interaction \( V_{\text{pd}} \) (i.e. nearly twice as large as for typical layered cuprates) is admitted. From the strong excitonic effects seen in the EELS data [4], analyzed within an effective one-band extended Hubbard model, a large interplaquette Coulomb repulsion \( V \approx 0.8 \) eV has been deduced. Within our approach this points to the relevance of further n.n.n. interactions such as a weak Cu-Cu \( V_{\text{dd}} \approx 0.85 \) eV and \( V_{\text{pp}} \approx 1.35 \) to 1.5 eV.

To summarize, our analysis reveals strong support for the suggested enhanced intersite Coulomb interaction \( V_{\text{pd}} \) in Sr\(_2\)CuO\(_3\) derived from the relatively small value \( \Delta_{\text{pd}} \leq 3 \text{eV} \) to explain the Cu XPS data [5] although at somewhat larger transfer integrals in our case. The latter result might explain together with a sizeable ferromagnetic contribution to the total exchange parameter \( J \approx 260 \) meV its unusual large value.

The Deutsche Forschungsgemeinschaft is gratefully acknowledged for financial support.

Figure 2. The obtained empirical relationship between the oxygen on-site energy difference \( \Delta_{\text{pp}} \) and the intersite Coulomb interaction \( V_{\text{pd}} \). The curve denoted by 'NNN' has been obtained including next-nearest-neighbor Coulomb interaction. The circle and the box indicate the most favored parameter ranges for Sr\(_2\)CuO\(_3\) and the 2D cuprates, respectively.

REFERENCES