Resonant Inelastic X-ray Scattering (RIXS) Spectra for Ladder Cuprates

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The ladder compound Sr$_{14}$Cu$_{24}$O$_{41}$ is of interest both as a quasi-one-dimensional analog of the superconducting cuprates and as a superconductor in its own right when Sr is substituted by Ca. In order to model resonant inelastic x-ray scattering (RIXS) spectra for this compound, we investigate the simpler SrCu$_2$O$_3$ system in which the crystal structure contains very similar ladder planes. We approximate the LDA dispersion of SrCu$_2$O$_3$ by a Cu only two-band tight-binding model. Strong correlation effects are incorporated by assuming an anti-ferromagnetic ground state. The available angle-resolved photoemission (ARPES) and RIXS data on the ladder compound are found to be in reasonable accord with our theoretical predictions.

I. INTRODUCTION

Resonant inelastic x-ray scattering (RIXS) is a second-order optical process in which there is a coherent absorption and emission of X-rays in resonance with electronic excitations. RIXS can probe charge excitations extending to fairly high energies of up to ~ 8 eV. This allows the analysis of electronic states over a wide energy range, including electron correlation effects originating from strong electron-electron Coulomb repulsion, providing thus a powerful tool for investigating Mott physics in solids.

The chain-ladder compound Sr$_{14}$Cu$_{24}$O$_{41}$ exhibits very interesting magnetic, transport and properties. It has attracted wide attention due to the discovery of a superconducting phase in highly Ca-doped samples at high pressure and charge order of the doped ladders. The compound possesses an incommensurate layered structure consisting of alternating layers of sublattices involving CuO$_2$ chains and Cu$_2$O$_3$ ladders. The superconductivity arises on the ladders, making them a quasi-one-dimensional analog of the cuprates. Very recently, K-edge RIXS data on the ladder compound has been reported, providing motivation for undertaking corresponding theoretical modeling of the spectra. Here, we attempt to do so by considering the simpler analog compound SrCu$_2$O$_3$. This should be a good approximation since interlayer coupling in Sr$_{14}$Cu$_{24}$O$_{41}$ is negligible, and both compounds have very similar ladder planes with similar hopping parameters. Specifically, we obtain K-edge RIXS spectra within a mean field approach for momentum transfer along as well as perpendicular to the direction of the ladders. A two-band Cu-only tight-binding model is used in which strong correlation effects are incorporated by treating an antiferromagnetic (AFM) ground state.

II. ELECTRONIC STRUCTURE AND THE TWO BAND MODEL

The spin-ladder compound SrCu$_2$O$_3$ possesses the orthorhombic structure with space group Cmmm in which Cu$_2$O$_3$ planes are stacked with Sr atoms sandwiched between these planes. Fig. 1 shows the detailed arrangement of Cu and O atoms in the Cu$_2$O$_3$ planes. This so-called ‘trellis structure’ involves Cu-O ladders where successive ladders are seen to be offset by half a unit cell. We obtained the band structure of SrCu$_2$O$_3$ self-consistently using a full-potential, all electron scheme within the local density approximation (LDA). The first principles bands were fitted by a 2-band tight-binding (TB) model in the vicinity of the Fermi energy, and provided the basis for RIXS computations presented in this study. Fig. 2 shows the first-principles as well as the TB bands along several high symmetry lines in the Brillouin zone (BZ). There are seen to be only two bands around the Fermi energy, which display large dispersion along the ladder direction Γ-Z, and a relatively smaller dispersion along the perpendicular Γ-X direction. In the first-principles band structure, both these bands are dominated by states of Cu $d_{x^2-y^2}$ character whose weight is given by the color bar on the right hand side of Fig. 2.

In making a TB fit to the two aforementioned LDA bands near the Fermi energy, we have adapted a Cu-only 2-band model suggested in Ref. 7. The detailed form of the TB Hamiltonian is discussed in the Appendix. The TB bands are seen from Fig. 2 to provide a good fit to the LDA bands near the Fermi energy. Our values of various parameters, i.e. the on-site energy $\epsilon_0$ and the hopping parameters $t_1-t_9$, are seen from Table 1 to be in reasonable accord with those of Ref. 7. The meaning of specific overlap terms involved in defining $t_1-t_9$ is clarified by the red arrows in Fig. 1. The present TB model includes not only the nearest neighbor hopping terms $t_1-t_3$, but also

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FIG. 1: (color online) Arrangement of Cu and O atoms in the Cu$_2$O$_3$ planes in the form of a series of ladders offset by half a unit cell along the c-axis. Unfilled circles give the location of out-of-plane Sr atoms. The green parallelepiped marks a primitive unit cell around which the orientation of Cu $d_{x^2-y^2}$, O $p_x$, and O $p_y$ orbitals is shown. Red arrows give the specific hopping parameters used in two-band model fits to the band structure. The AFM ordering of spins in the ladders is depicted by the green arrows.

the longer range hoppings $t_4 - t_9$. Interestingly, we find that the inter-ladder dispersion (i.e. along Z-A in Fig. 2) cannot be fitted well using only a nearest-neighbor hopping model. Table 1 shows that the intra-ladder hopping parameters ($t_1$, $t_2$ and $t_5$) are generally larger than the inter-ladder terms such as $t_3$, $t_4$ and $t_6$. This can be understood with reference to Fig. 1 where orientation of the Cu-$d_{x^2-y^2}$ and O-$p_x$ and $p_y$ orbitals is sketched on a few sites. An intra-ladder Cu-O-Cu path with a bond angle of 180° (e.g. Cu$_2$-O$_1$-Cu$_1$) will be expected to provide a larger orbital overlap than an inter-ladder path with a 90° bond (e.g. Cu$_4$-O$_3$-Cu$_1$). The glide symmetry of SrCu$_2$O$_3$ leads to some dispersion anomalies, including extra degeneracies at the zone boundaries and an apparent $4\pi$ periodicity of the dispersion along the ladder. A $2\pi$ symmetry can be effectively restored by including different cuts along $k_z$ as shown in Fig. 3(a). Similar anomalies in c-axis dispersion due to a glide symmetry are also found in Bi$_2$Sr$_2$CaCu$_2$O$_8$ (Bi2212). Due to the 180° Cu$_1$-O$_1$-Cu$_2$ bonds, the spins are strongly coupled antiferromagnetically on the legs and the rungs of the ladders as indicated by green arrows in Fig. 1. However, the displacement of successive ladders with respect to each other frustrates the development of long range AFM order. One nevertheless expects the electronic system to experience significant AFM fluctuations, which are presumably sufficient to impose an underlying dispersion characteristic of the AFM order. In this spirit, we have approximated the correlation effects within a Hartree-Fock model of an itinerant AFM.

FIG. 2: (color online) First-principles LDA band structure of SrCu$_2$O$_3$ along several high symmetry lines in the irreducible Brillouin zone is shown by dots of various colors. The colors of the dots code the weight of Cu $d_{x^2-y^2}$ character in the associated wavefunctions as indicated by the color-bar on the right hand side of the figure. Solid red lines give the 2-band tight-binding model fit to the LDA bands near the Fermi energy.

TABLE I: TB parameters for 2-band model.

| Parameter | This work  | Ref. 7 |
|-----------|-----------|--------|
| $\epsilon_0$ | -0.0350(eV) | -0.0450(eV) |
| $t_1$ | 0.5650 | 0.5650 |
| $t_2$ | 0.3800 | 0.3950 |
| $t_3$ | 0.0400 | 0.0400 |
| $t_4$ | 0.0520 | 0.0500 |
| $t_5$ | -0.1200 | -0.1150 |
| $t_6$ | 0.0700 | 0.0400 |
| $t_7$ | 0.0750 | 0.0750 |
| $t_8$ | 0.0057 | 0.0050 |
| $t_9$ | -0.0115 | -0.0200 |
as in the planar cuprates\textsuperscript{14}. Taking the on-site energy to be $U=3.3\text{ eV} \sim 6t$, the magnetization was computed self-consistently to be $m=0.43$. The AFM Hamiltonian is given in the Appendix and the resulting dispersions are shown in Fig. 3(b). Comparison with the paramagnetic solution shows that a large gap of $\sim 2.3\text{ eV}$ opens up between the upper (UMB) and the lower magnetic bands (LMB). The theoretical LMBs display the characteristic backfolding near $k_z = \pi/2c$, which is in accord with the experimentally observed dispersion (green dots in Fig. 3(b)) via ARPES\textsuperscript{15}, and is reminiscent of a similar effect in the insulating planar cuprates.

III. RIXS SPECTRA

Our computations of the K-edge RIXS cross section for the Cu $1s \rightarrow 4p$ core level excitation are based on the expression\textsuperscript{16,17}

$$ I(q, \omega, \omega_i) = (2\pi)^3 N |W(\omega, \omega_i)|^2 \times \sum_{j,j',k} \chi_{jj'}^\omega(q, k, \omega) |M_{ij}(k)|^2 $$

where

$$ \chi_{jj'}^\omega(q, k, \omega) = \delta(\omega + E_j(k) - E_{j'}(k+q)) \times n_j(k)[1 - n_{j'}(q + k)], $$

$n_j(k)$ is the electron occupation of the $j^{th}$ band and $E_j(k)$ is the corresponding energy dispersion obtained by self-consistently solving the two-band AFM Hamiltonian (see Appendix), and

$$ W(\omega_f, \omega_i) = |\gamma| \Sigma_k \frac{V_f}{D(\omega, k_1) D(\omega_f, k_2)}. $$

Here, $D(\omega, k) = \omega + \varepsilon_{1s} - \varepsilon_{4p}(k) + i\Gamma_{1s}$, $\gamma$ is the matrix element for scattering from $1s$ to $4p$, and $V_f$ is the core-hole potential in $3d$ level. $\omega_f(\omega_i)$ and $q_i(q_f)$ denote the initial (final) energy and momentum, respectively, of the photon, and $\omega = \omega_i - \omega_f$ and $q = q_i - q_f$ give the energy and momentum transferred in the scattering process. Since Cu $1s$ is a core state, the associated energy band $\varepsilon_{1s}(k)$ is assumed dispersionless. The Cu $4p$ band dispersion $\varepsilon_{4p}(k)$ is modeled by a 2D-TB model with nearest neighbor hopping. $\Gamma_{1s}$ is the decay rate of core hole taken to be $0.8\text{ eV}$. The matrix element $M_{ij}$ associated with the interaction between the core hole and $3d$ levels around the Fermi energy is

$$ M_{ij}(k) = \sum_{l,\sigma,\sigma'} e^{i\mathbf{q} \cdot \mathbf{R}_l} \alpha_{l\sigma} X_{\sigma}^j(k) \Lambda_{\sigma,\sigma'}^{ij}(\omega, \mathbf{q}) X_{\sigma'}^{i'}(k+\mathbf{q}), $$

in terms of the eigenvectors $X_{\sigma}^j$ of the AFM Hamiltonian, where $\sigma$ denotes electron spin and $l$ an orbital index. $\alpha_l = V_l/V_{4s}$, where $V_l$ is the Coulomb interaction between a core hole and an electron on atom $l$ separated by a distance $R_l$. Here we approximate the vertex correction $\Lambda \rightarrow \delta_{\sigma,\sigma'}$.

Fig. 4 shows RIXS spectra computed within the 2-band AFM model in the form of a color plot for mo-
FIG. 5: (color online) (a) Lowest and uppermost tight-binding AFM bands in SrCu₂O₄ at kᵢ = 0. (b) Red line gives partial contribution to the RIXS spectrum at qᵢ = π/2c (marked by yellow vertical line in Fig. 4(a)) from transitions between the two bands in (a). Blue line gives the corresponding total RIXS spectrum. Features A-C are discussed in the text.

FIG. 6: (color online) Comparison of unbroadened (blue curves) and broadened (black curves) RIXS spectra with the corresponding experimental data (red dots) taken from Ref.4. Left hand side panels are for qᵢ∥c at various qᵢ values, while the right hand side panels are for qᵢ⊥c at various qᵢ values as indicated. Spectra are normalized as shown.
for \( q_x c \) with \( q_z \) varying over the range \( 0-\pi/a \). The unbroadened theoretical spectra (blue lines) require a substantial broadening for a meaningful comparison with the data (red dots). Accordingly, we have applied a combined Gaussian and Lorentzian broadening to the computed spectra to obtain the broadened theoretical spectra in Fig. 6 (black lines). The Gaussian broadening is taken as the nominal experimental resolution of 120 meV\(^2\). The residual broadening, which reflects presumably lifetime effects not accounted for in our computations, is modeled via a Lorentzian with half-width-at-half-maximum of \( \Gamma_d=1.1 \) eV for \( q \parallel c \) and of \( \Gamma_d=1.4 \) eV for \( q \perp c \).\(^{18,19} \) Although Figs. 4-6 show that the RIXS spectra intrinsically contain considerable information concerning the charge excitations and their momentum dependencies in the ladder compound, much of this structure is seen to be lost in the broadened spectra. Nevertheless, for both \( q \parallel c \) as well as \( q \perp c \), the experimental spectra are in reasonable accord with the broadened theoretical, some discrepancies in shape and fine structure in the spectra are in reasonable accord with the broadened theoretical spectra. Broadened theoretical spectra (blue lines) require a sub-

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**Appendix: AFM and Paramagnetic Hamiltonians and Dispersions**

The AFM Hamiltonian for the two band model can be written as

\[
H = \sum_{j} \varepsilon_0 d_j^+ d_j + \sum_{<i,j>} t_{ij} (d_i^+ d_j + d_j^+ d_i) + \sum_{j} U n_{d_j^+} n_{d_j} + \sum_{j} \Delta n_{d_j^+} n_{d_j},
\]

where \( n_{d_j} = d_j^+ d_j \). \( \varepsilon_0 \) is the on site energy, \( t_{ij} \)'s are the hopping parameters (see Fig. 1), and \( U \) is Hubbard \( U \). The AFM ordering of spins is shown in Fig. 1 by green arrows. The Hartree-Fock decomposition of the Hubbard term in the Hamiltonian is given by

\[
H_{11} = H_{44} = \Delta + U m, \quad H_{22} = H_{33} = \Delta - U m
\]

For the the paramagnetic case \( U_m \) is equal to zero and the 4 \times 4 AFM Hamiltonian is reduced to a 2 \times 2 form with matrix elements

\[
H_{11} = H_{22} = \varepsilon_0 - 2t_1 \cos(k_z) - 2t_7 \cos(2k_z) - 4t_4 \cos(k_z/2) \cos(k_z/2) - 2t_9 \cos(3k_z/2) \cos(3k_z/2),\quad (8)
\]

where

\[
\Delta = \varepsilon_0 - 2t_7 \cos(2k_z) - 2t_4 \cos((k_z + k_x)/2) - 2t_9 \cos((3k_z - k_x)/2)).
\]

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\]
The resulting dispersion is

\[ \varepsilon_{\pm}(k) = \varepsilon_0 + \varepsilon_{\parallel}(k) \cos(k_x/2) + (\varepsilon_{\perp,1}(k) \cos(k + x/2) - \varepsilon_{\perp,3}(k) \cos(k_x/2)^3 + 2\varepsilon_{\perp,3}(k) \varepsilon_{\perp,3}(k) \cos(k_x/2))^{1/2} \] (9)

where

\[ \varepsilon_{\parallel}(k) = -2t_1 \cos(k_x) - 2t_7 \cos(2k_x), \] (10)
\[ \varepsilon_{\perp,1}(k) = -4t_4 \cos(k_x/2) - 4t_9 \cos(3k_x/2), \]
\[ \varepsilon_{\perp,3}(k) = t_2 + 2t_5 \cos(k_x) + 2t_8 \cos(2k_x), \]
\[ \varepsilon_{\perp,4}(k) = 2t_3 \cos(k_x/2) + 2t_6 \cos(3k_x/2). \]