Mott Gap and Orbital Excitations in Quasi-low-dimensional Cuprates Studied by Resonant Inelastic X-ray Scattering

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Introduction

The discovery of a host of unusual transport properties, such as high-temperature superconductivity and colossal magnetoresistance, in doped transition metal oxides has led to extensive interest in research on their parent compounds: the Mott insulators. The parent compounds of such oxides are half-filled systems (one electron per site of the lattice, where a large on-site Coulomb interaction dominates the physics). As a consequence, these systems exhibit a low-temperature insulating state characterized by a charge-excitation gap known as the Mott gap. The gap is due to strong electron-electron Coulomb interaction, which dominates over the conventional one-electron bandwidth. The existence of exotic electronic, magnetic, and optical properties is believed to be related to the strong electron-electron Coulomb correlations in these systems [1-4]. This suggests the necessity of studying their momentum-resolved charge dynamics.

Methods and Materials

We report here on a high-resolution inelastic x-ray scattering study of an edge-sharing compound ($CuGeO_3$ or Ge cuprate) and compare it with corner-sharing $SrCuO_2$ (Sr cuprate). These compounds are charge transfer systems exhibiting effective Mott gaps. We studied the charge fluctuation spectrum by varying q (the scattering vector) over the second and third Brillouin zones. We found that a better signal-to-noise ratio is achieved for the low-energy features in the higher Brillouin zones than in the first zone. We found that the charge fluctuations, consisting of a particle-hole pair at the gap edge, are only weakly dispersive in the Ge cuprate system but are largely dispersive in the Sr cuprate system. The experiment was performed by using the high-flux undulator beamlines 12-ID (BESSRC) and 9-ID (CMC-CAT) at the APS and beamline X-21 at the National Synchrotron Light Source at Brookhaven National Laboratory. Inelastic scattering was measured by varying q along the Cu-chain direction of single-crystalline $CuGeO_3$ and $SrCuO_2$. The basic structural differences between these compounds are shown in Fig. 1. $GeCuO_3$ is an edge-sharing system, and Cu-O-Cu bonds are about 99°, giving rise to small Cu-to-Cu hopping. In contrast, the Cu-O-Cu bond in $SrCuO_2$, a corner-sharing system, is almost 180°, leading to a large hopping integral and quasi-one-dimensionality. Scattering was performed in a standard triple-axis arrangement. The scattered beam was reflected from a diced Ge-based analyzer for energy analysis and focused onto a solid-state (AmpTek) detector. Overall energy resolution varied from 350 to 400 meV for this experiment.

Results

Figure 2 (right panel) shows inelastic x-ray scattering spectra in $CuGeO_3$ with varying momentum transfers

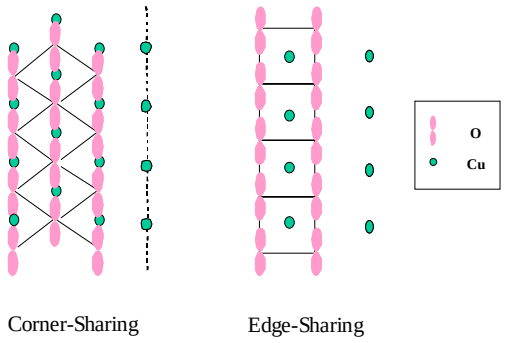

FIG. 1. Topology of the lattice determines the nature of the electronic excitations. In case of $CuGeO_3$, an edge-sharing system (right), Cu-O-Cu bonds are almost 99°, giving rise to small Cu-to-Cu hopping and quasi-zero-dimensionality. In contrast, in $SrCuO_2$, a corner-sharing system (left), Cu-O-Cu bonds are almost 180°, leading to large hopping integral and quasi-one dimensionality.

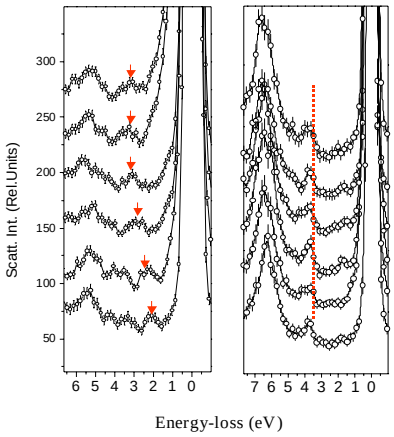

FIG. 2. q-Dependence of charge excitations in $SrCuO_2$ (left) and $CuGeO_3$ (right). For $CuGeO_3$, the approximate value of q (bottom to top) is 1, 1.2, 1.3, 1.5, 1.6, and 1.9 (in units of pi/a), whereas for $SrCuO_2$, the value of q (bottom to top) is 2.05, 2.4, 2.6, 2.8, 3.02, and 3.2 (in units of pi/a, with a being the lattice constant along the chain).

along the chain direction with the incident energy fixed near the Cu K edge (E_o = 9 KeV), where the largest resonance enhancement was observed. Each spectrum shows a strong feature around 6.4-eV excitations to the antibonding states, which are analogous to the 6-eV excitations observed in 2-D compounds [9, 10]. The second prominent feature is around 3.8 eV. At lower momentum transfers, near 2pi, the feature at 3.8 eV coincides with the excitations seen in optical studies that signal transitions across the insulating gap. Thus, we associate it with particle-hole pair excitation defining the effective Mott gap in this system [12]. Although the line shape of the excitation has some q-dependence, this excitation has virtually no energy dispersion within the level of our energy resolution. This strongly suggests the localized character of this pair, involving a hole of Zhang-Rice character and an electron with upper Hubbard band character [12]. Figure 2 (left panel) shows inelastic excitation spectra in the Sr cuprate along the Cu-O-Cu direction. These results are improvements on our earlier work [11]. These spectra were taken with the incident x-ray energy set to 8997.6 eV, where large enhancements were observed. At low-q near 2pi, it shows an excitation feature near 2 eV, which increases in energy as one goes from 2pi to 3pi. The total dispersion is about 1 eV. Beyond 3pi, the feature tends to appear at lower energies again. At low q values, there may be some weak multiple features around energy losses of 3–4 eV. These features could be due to excitations from the nonbonding bands to the upper Hubbard bands [12]. In comparison with optical data [12] and the x-ray spectrum at q of ~2pi, the 2-eV feature we observed is again a particle-hole pair excitation defining the effective Mott gap in the system. For comparison, the q-dependence of the Mott excitations in Ge cuprate and the Sr cuprate is plotted in Fig. 3, which shows the dramatic difference in dispersions due to effective dimensionality. Pair excitations at the edge of the gap are more highly propagative or dispersive in the quasi-1-D system than in the quasi-0-D system. In addition, because the incident x-ray energy tuned near the Cu K edge, these experiments provided a natural way to identify excitations originating from the Cu-O chain, so they can easily be separated from excitations involving states originating from atoms that are out of the Cu-O chains.

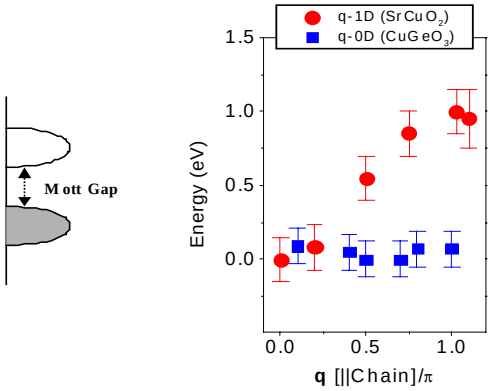

FIG. 3. Dispersions: The left side illustrates that the Mott gap is an energy-gap in the charge excitation spectrum in the presence of strong electron-electron interaction not described in a conventional band picture. The graph on the right shows the momentum-structure (q-dependence) of the Mott-gap excitations in quasi-0-D ($GeCuO_3$) and quasi-1-D ($SrCuO_2$) cuprates. Momentum dependence is largely determined by the dimensionality of the Cu-O network and the nature of electron-electron Coulomb interaction.

Discussion

In a simplistic view, the core-hole created by the x-ray photon near the absorption edge causes electronic excitations in the valence band, which can be composed of a hole in the occupied band and an electron in the unoccupied band across the gap in an insulator. The particle-hole pair formed in the process absorbs the energy and momentum lost from the incident x-ray beam. Pair excitations in insulating cuprates consist of a hole with Cu-3*d* (hybridized with O-2*p* states) character and an electron with upper Hubbard band character. The detail momentum dependence of such a complex provides information about upper Hubbard bands. In the case of $CuGeO_3$, effective Coulomb interaction is large, resulting in a larger Mott gap, but Cu-O-Cu interaction is weak, resulting in a weak dispersion. In the case of $SrCuO_2$, the

insulating gap again scales with electron-electron Coulomb interaction, but a larger dispersion results from larger Cu-O-Cu interactions. A comparison of momentum-resolved charge excitations between a quasi-0-D compound and quasi-1-D compound shows that the momentum structure of charge excitations over the Brillouin zone dramatically reflects the effective dimensionality of the lattice and the strength of electron-electron Coulomb interaction. A comparison with model many-body Hamiltonian functions would allow us to extract the fundamental electronic parameters, which will be addressed in a separate paper. This novel type of experiment can be improved by using brighter synchrotron sources and by developing more efficient crystal optics. The experimental results reported here demonstrate the power and versatility of such x-ray spectroscopy in addressing some fundamental issues of charge excitations in Mott insulators that cannot be directly addressed by other well-developed spectroscopic techniques.

Acknowledgments

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