Momentum-Resolved Charge Excitations in a Prototype One-Dimensional Mott Insulator

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We report momentum-resolved charge excitations in a one-dimensional (1D) Mott insulator studied using high resolution inelastic x-ray scattering over the entire Brillouin zone for the first time. Excitations at the insulating gap edge are found to be highly dispersive (momentum dependent) compared to excitations observed in two-dimensional Mott insulators. The observed dispersion in 1D cuprates ($SrCuO_2$ and Sr_2CuO_3) is consistent with charge excitations involving holons which is unique to spin-$1/2$ quantum chain systems. These results point to the potential utility of momentum-resolved inelastic x-ray scattering in providing valuable information about electronic structure of strongly correlated insulators.

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After several decades of research efforts, the electronic structure of late transition metal oxides lacks comprehensive understanding. The existence of exotic electronic, magnetic, and optical properties such as high T_c superconductivity as exhibited by the cuprates or colossal magnetoresistance as in the manganites or highly nonlinear optical responses as observed in the nickelates is believed to be related to the strong electron-electron Coulomb correlations in these systems [1–4]. This suggests the necessity of studying their correlated charge dynamics. In the last several years, with the advent of high brightness synchrotron facilities, inelastic x-ray scattering has been developing as a tool to study the *bulk* electronic structure of condensed matter systems [4–10]. X-ray scattering from the valence charge distribution is fairly weak and thus difficult to distinguish from the total scattering signal especially in high-Z materials making such experiments quite difficult to perform. Recent experimental and theoretical investigations have shown that by tuning the incident energy near an x-ray absorption edge a large enhancement can be achieved, making the study of valence excitations feasible in high-Z systems such as low-dimensional cuprates [5,7–10].

One-dimensional half-filled spin-$1/2$ quantum systems as realized in some copper oxide compounds (such as Sr_2CuO_3 and $SrCuO_2$) are believed to exhibit spin-charge separation. As a consequence, in these systems charge fluctuations propagate rather freely and independently of the spin fluctuations [11–15]. This is in contrast to the two-dimensional (2D) spin-$1/2$ systems where charge motion is strongly coupled to the spin fluctuations and rather restricted [10,12,14–17]. Recently, it has been argued that the giant optical nonlinearities observed in 1D cuprates are due to spin-charge separation and the consequent unusual charge propagation behavior in these systems [18]. In 1D, charge excitations would be expected to be highly dispersive compared to analogous 2D systems. This behavior contrasts qualitatively to that of the band structure calculations with no interactions considered, where the 1D system is expected to have only half of the dispersion as that of the 2D. Because of the insulating gap, charge fluctuations are at relatively high energies in Mott insulators [1,2]. The momentum dependence of the effective Mott gap (charge-transfer gap) has been reported recently in a parent 2D cuprate using inelastic x-ray scattering [8,10,19]. In this Letter, we report a study of 1D Mott insulator's charge fluctuation spectrum by varying $\mathbf{q}$ (the scattering vector) over the entire Brillouin zone using high resolution inelastic x-ray scattering. The Mott gap is found to be of direct nature (minimum of the gap appears at $\mathbf{q} \sim 0$) within the level of experimental resolution and the charge fluctuations at the gap edge are more dispersive along the Cu-O bond direction in a 1D as compared to a 2D parent cuprate insulator.

The experiment was performed using the high flux undulator beam line 12-ID (Basic Eenergy Sciences Synchrotron Radiation Center Sector-12) at the Advanced Photon Source of Argonne National Laboratory. Inelastic scattering was measured by varying $\mathbf{q}$ along the chain direction (Cu-O bond direction) of single crystallines Sr_2CuO_3 and $SrCuO_2$. The experiment was performed at room temperature where the Cu-O-Cu bonds in the sample effectively behave as one dimensional [14]. Overall energy resolution of 325 meV was achieved for this experiment. This is an improvement over our earlier works on 2D Mott systems by more than 100 meV [8,10,19]. This improvement in resolution (in combination with the high flux from the Advanced Photon Source) allowed us to resolve the Mott excitations in 1D systems despite the high level of x-ray absorption due to Sr in the system under study. The energy of the incident beam was set near the Cu K edge ($E_0 = 8.996$ keV) for the resonant enhancement of excitation features. The scattering was performed in a standard triple-axis arrangement [5,6]. The scattered beam was reflected from a diced Ge-based analyzer [Ge(733)] and focused onto a solid-state (Cd-Zn-Te based) detector. The detector was thermoelectrically cooled to achieve a low level of random background which is necessary to detect small signals from the sample under study. For $\mathbf{q}$ scans, the incident energy was kept fixed and $\mathbf{q}$ was varied by rotating the entire spectrometer around the scattering center. For the geometry employed, beam polarization had a nonvanishing component along the direction of the momentum transfer ($\mathbf{q}$). The scattering geometry is described in Fig. 1 (top left panel). The background, measured on the energy gain side, was about 2–3 counts per minute similar to the study reported in Ref. [10]. The Sr_2CuO_3 and $SrCuO_2$ crystals used for this experiment were grown and characterized by techniques described previously which confirmed their quasi one dimensionality above 6 K. The sample undergoes a Néel transition around 6 K due to three-dimensional magnetic coupling [14,15]. In Sr_2CuO_3 or $SrCuO_2$ the overlap integral along Cu-O-Cu is much larger than it is in other extensively studied quasi-one-dimensional cuprates such as $CuGeO_3$. In addition, Sr_2CuO_3 and $SrCuO_2$ show no spin-Peierls transition and hence provide a unique opportunity to study the charge fluctuations in a 1D spin-$1/2$ quantum Heisenberg system [14,15].

Figure 1 shows inelastic x-ray scattering spectra with varying momentum transfers along the chain direction (the Cu-O bond direction) with incident energy fixed near Cu K edge (left bottom panel of Fig. 1 shows the location of the incident energy, $E_0 = 8.996$ KeV, with respect to the absorption edge). Each spectrum shows two features, one around 5.6 eV and another, lower in energy, appear in the range of 2.5–3.5 eV depending on different values of the scattering wave vector, $\mathbf{q}$. The 5.6 eV feature can be assigned to be a charge transfer excitation from the ground state to the antibonding-type excited state which is analogous to the 6 eV excitation observed in 2D cuprate insulators [7,8,10]. The other prominent feature that appears at lower energies has a significant movement in changing $\mathbf{q}$. The feature disperses upward in energy about 1 eV monotonically over the Brillouin zone in going from the zone center ($\mathbf{q} \sim 2\pi$) to the edge of the zone ($\mathbf{q} \sim \pi, 3\pi$). We subtract the quasielastic scattering centered around zero energy loss from all the spectra shown in Fig. 1 and present them in Fig. 2 (left panel) for clarity. The $\mathbf{q}$ dependence of the low-energy feature is evident and the feature is broad for all momenta over the entire Brillouin zone and not resolution limited.

Inelastic x-ray scattering measures the dynamical charge-charge correlation function, $N(\mathbf{q}, \omega)$ (charge fluctuations), which can be interpreted as particle-hole pair excitations in the range of momentum transfers comparable to the size of the Brillouin zone of the system.

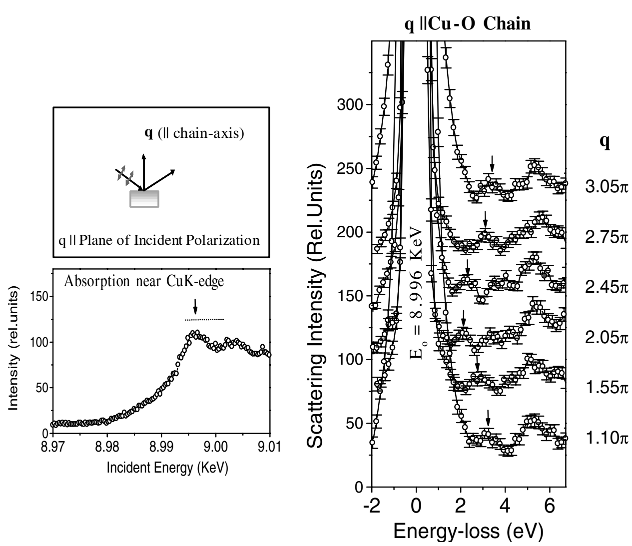

FIG. 1. Right panel: inelastic x-ray scattering spectra along the chain direction are shown as a function of $\mathbf{q}$ scanning over the entire Brillouin zone. The values of $\mathbf{q}$ for the spectra bottom to top are 1.10π, 1.55π, 2.05π, 2.45π, 2.75π, and 3.05π, respectively. The spectrum for $\mathbf{q} \sim 1.1\pi$ was taken with a resolution of about 490 meV, whereas the rest were taken with 325 meV energy resolution. Left panel: the top panel shows a schematic of the scattering geometry which had $\mathbf{q}$ in the plane of polarization. The bottom panel shows the location of the incident energy with respect to the Cu K absorption edge.

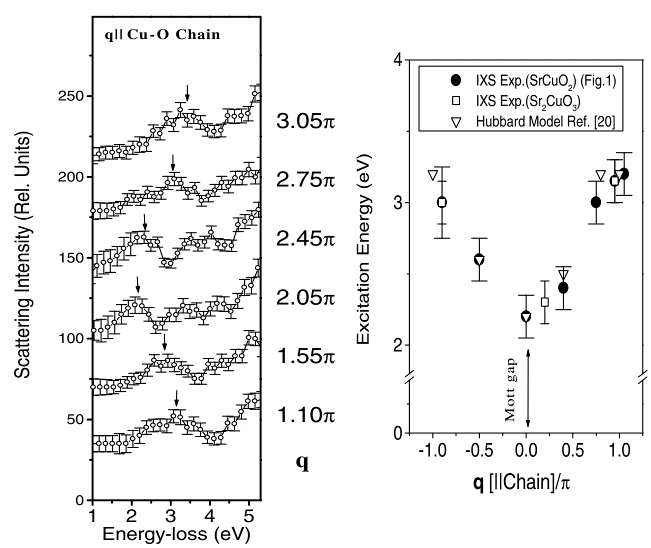

FIG. 2. Left panel: momentum dependence of the lower energy inelastic feature is shown. The quasielastic scattering centered at the zero energy loss has been removed by fitting [10]. Right panel: a dispersion plot (energy as a function of momentum) is shown based on data from both samples ($SrCuO_2$ and Sr_2CuO_3) and a comparison with the Hubbard model is made [20]. All the data points are plotted in their equivalent $\mathbf{q}$ position within $-\pi$ to π.

Near an absorption edge the measured dynamical response function gets modified due to the presence of a core hole and an excited electron, but it can still be interpreted as composites of pair excitations [8–10,19–22]. 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 having 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. The propagation of this pair would depend on the charge and spin distributions in the system.

Sr_2CuO_3 and $SrCuO_2$ are believed to be quasi-one-dimensional half-filled Mott insulators with short-range antiferromagnetic spin correlations (quantum Heisenberg systems) [14,15] and have a Cu-O bond length much more similar (less than 1% difference) to the Cu-O bond length of a 2D parent cuprate such as $Ca_2CuO_2Cl_2$ that we studied earlier [10,14].

Excitations of 1D Mott insulators are believed to be quite different from those of the 2D Mott insulators. In 1D (spin-1/2 with local antiferromagnetic coupling) there is no well defined quasiparticle where charge degrees of freedom dynamically decouple from the spin degrees of freedom and propagate freely. So it would be interesting to compare charge fluctuation spectra in 1D to the results from our earlier work on 2D ($Ca_2CuO_2Cl_2$). We plot a comparison of **q**-resolved charge fluctuation spectra in 1D and 2D in Fig. 3. It is evident from Fig. 3 that the **q** dependence of the Mott feature in 1D is larger than it is along the bond direction in 2D which is consistent with the view that charge motion is easier in 1D than in 2D [10]. This behavior contrasts qualitatively to that of the band structure calculations with no (or weak) interactions considered, where the 1D system is expected to have only half of the dispersion as that in 2D [15]. Such behavior would be qualitatively expected when charge fluctuations are free to move because of decoupling from the spin degrees of freedom. This is also seen from numerical studies of the Hubbard model assuming spin-charge separation [20,22]. In Fig. 2 (right panel) we compare our experimental results to the expectations from the 1D half-filled spin-1/2 Hubbard model describing the charge fluctuation spectrum at finite **q** [20]. The model calculation in Ref. [20] was performed under resonant conditions (in the presence of a Cu 1*s* core hole), which is similar to the current experimental condition. It is interesting to note that dispersions predicted in Ref. [16], calculated nonresonantly (in the absence of a core hole), are also consistent with our experimental data within the energy and momentum resolution of the experiment. For comparison with model calculations, we chose the onset of the feature which is a better measure of the charge gap rather than the center of gravity of the feature. The level of energy and momentum resolution of the current experiment is inadequate for linewidth analysis of the **q**-dependent features. Otherwise, the momentum dependence of the edge of the gap is qualitatively described by excitations involving holons [20,22] in the 1D Hubbard model.

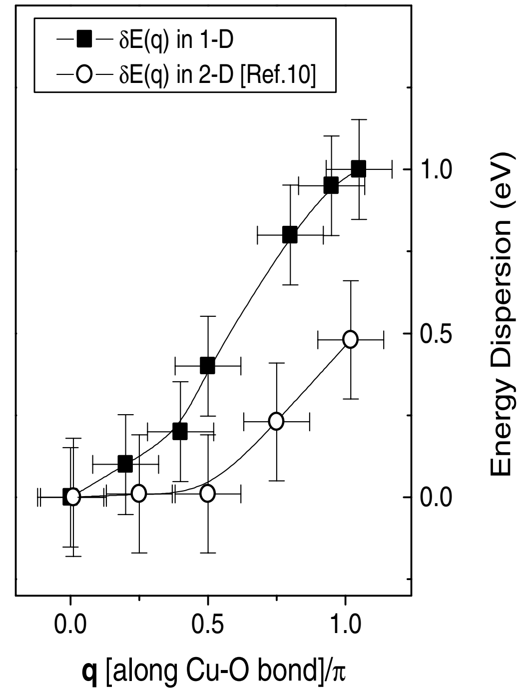

FIG. 3. Comparison of the **q** dependence of charge excitations along the Cu-O bond direction in 1D and 2D [10] is shown. All the **q** points are plotted in their equivalent positions within 0 to π for comparison. (Not all the experimental spectra used to make this plot are shown in Fig. 1.) Charge excitations are found to be more dispersive in 1D than in 2D along the Cu-O bond direction.

Spectroscopic studies interpreted based on model calculations suggest that these 1D cuprates exhibit spin-charge separation [14–16]. Angle-resolved photoemission spectroscopy (ARPES) shows that the hole bandwidth is much larger in 1D than in 2D, contrary to the expectation of LDA (local density approximation)-type models for electronic structure [15]. The quasiparticle decay modes, probed by ARPES, are interpreted as spinons and holons. (A cartoon view of the decay of a single hole into two topological excitations is shown in Fig. 4. Such excitations are described in detail in Ref. [15].) Since a holon is a collective charge mode it would couple to the x rays directly and strongly and exhibit its characteristic **q** dependence; hence inelastic x-ray scattering is a natural probe for studying such excitations. The inelastic x-ray scattering (IXS) results, in the low-**q** regime, are also qualitatively consistent with electron scattering (which is limited to the low-**q** regime) studies of 1D cuprates in the low-energy regime [16]. Present x-ray scattering results complement the electron scattering results in the high-**q** regime providing access to study dispersions over the complete Brillouin zone. In addition, having the incident x-ray energy tuned near the Cu *K* edge,

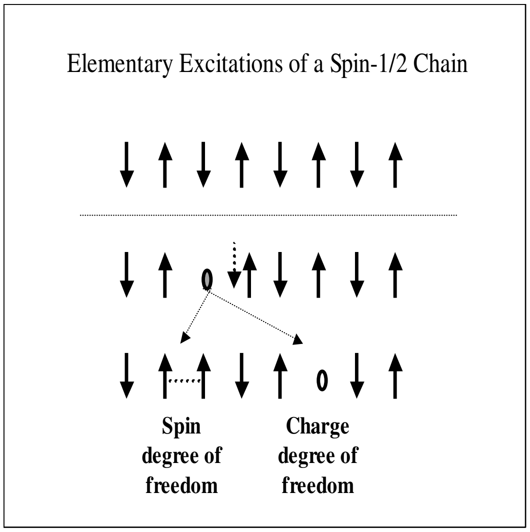

FIG. 4. A schematic (cartoon) view is presented to describe the elementary excitations in a spin chain with short-range antiferromagnetic order. The middle row of spins shows a particle-hole pair excitation. The bottom row shows that the hole decays into two topological defects; one carries the spin degrees of freedom and the other carries the charge. These topological objects can propagate easily in 1D [13,15]. Although difficult to visualize, these collective quantum processes are involved in the excitations created by the x rays. For the current IXS experiment, scattering intensities reflect only the charge components of these excitations. The nature of these topological excitations is described in Ref. [15].

present experiments provide a natural way to identify excitations originating from the Cu-O plane; hence they can easily be separated from excitations involving states originating from atoms that are out of the Cu-O plane [7–10].

Momentum-resolved charge fluctuations in 1D Mott insulators indicate that in 1D Mott gap is of direct nature and excitations at the gap edge are more dispersive as compared to the equivalent direction in 2D. The dispersions are also consistent with models describing the motion of holons in 1D spin-$1/2$ Mott insulators. Similar experiments could be performed when more than one orbital plays a role (as in many other transition metal oxides unlike cuprates) and study the dispersions of collective orbital excitations which is an unexplored degree of freedom in a strongly correlated electron system. The experiment can be improved by using brighter synchrotron sources and by developing more efficient crystal optics. Nonetheless, the results reported in this Letter suggest that momentum-resolved high resolution inelastic x-ray scattering can be used to study the electronic structure of complex insulators and strongly correlated electron systems in general.

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