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

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We report momentum resolved charge excitations in a one dimensional (1-D) Mott insulator studied using high resolution (\(\sim 325\) meV) 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 1-D 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 inelastic x-ray scattering in providing valuable information about electronic structure of strongly correlated insulators.

After several decades of research efforts, 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 are 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 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 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 [5,7-10]. One dimensional half-filled spin-1/2 quantum systems as realized in some copper oxide compounds (such as \(\text{Sr}_2\text{CuO}_3\) and \(\text{SrCuO}_2\)) 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 (2-D) spin-1/2 systems where charge motion is strongly coupled to the spin fluctuations and rather restricted [10,12,14-17]. In 1-D, charge excitations would be expected to be highly dispersive compared to analogous 2-D systems. 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 2-D cuprate using inelastic x-ray scattering [8,10,20]. In this Letter, we report study of 1-D 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 a 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 1-D as compared to 2-D parent cuprate insulator.

The experiment was performed using the high flux undulator beamline 12-ID (BESSRC-CAT) 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 crystalline \(\text{Sr}_2\text{CuO}_3\). Overall energy resolution of 325 meV was achieved for this experiment. This is an improvement over our earlier works on 2-D Mott systems by more than 100 meV [8,10,20]. This improvement in resolution (in combination with the high flux from the Advanced Photon Source) allowed us to resolve the Mott excitations in 1-D systems for the first time despite 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 (\(E_o = 8.996\text{ eV}\)) for resonant enhancement of excitation features. The scattered beam was reflected from a diced Ge-based analyzer and focused onto a solid-state detector. For \(\mathbf{q}\)-scans, the incident energy was kept fixed and \(\mathbf{q}\) was varied by rotating the entire spectrometer around the scattering center. The background, measured on the energy gain side, was about 2-3 counts per minute. \(\text{Sr}_2\text{CuO}_3\) crystals used for this experiment were grown and characterized by techniques described previously which confirmed its quasi-one dimensionality above 6 K (Néel transition due to 3-D coupling)[14,15]. Un-
like extensively studied 1-D cuprates such as CuGeO$_3$ or KCuF$_3$, Sr$_2$CuO$_3$ and SrCuO$_2$ show no spin-Peierls transitions hence provides a unique opportunity to study the charge fluctuations in a 1-D spin-1/2 quantum Heisenberg chain system [14,15].

Fig. 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 ($E_o = 8.996$ KeV). All the spectra in each panel were normalized to the intensity in a window between 8 and 9 eV energy-loss (not shown in the Fig.1). Each spectrum shows two features, one around 5.6 eV and another, lower in energy, appear in the range of 2.5 to 3.5 eV depending on different values of the scattering wave vector, $q$. The 5.6-eV feature can be assigned to be a charge transfer excitation from the groundstate to the antibonding-type excited states which is analogous to the 6 eV excitation observed in 2-D cuprate insulators [7,8,10]. The other prominent feature that appears at lower energies has a significant movement in changing $q$. The feature disperses upward in energy about 1 eV monotonically over the Brillouin zone in going from the zone center ($q \sim 2\pi$) to the edge of the zone ($q \sim \pi, 3\pi$). Inelastic features with similar intensities and dispersions were also seen for incident energy of 8.992 KeV.

Inelastic x-ray scattering measures the dynamical charge-charge correlation function (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. Near an absorption edge the measured response function gets modified but it can still be interpreted as composites of pair excitations [8-10,18-21]. 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. We interpret the dispersion of the low energy feature seen in our data as the dispersion of the effective Mott gap (charge-transfer type [22]) in the system. The particle-hole pair formed in the process absorbs the energy lost from the incident x-ray beam and propagates with momentum $hq$. The propagation of this pair would depend on the charge and spin distributions in the system. Sr$_2$CuO$_3$ and SrCuO$_2$ are believed to be quasi-1-D half-filled Mott insulators with short-range antiferromagnetic spin correlations (quantum Heisenberg systems) [14,15] and has a Cu-O bond length (lattice constant of 3.90 Å) much similar (less than 1% difference) to the Cu-O bond length of 2-D parent cuprate such as Ca$_2$CuO$_2$Cl$_2$ (lattice constant of 3.87 Å) studied earlier [10,14].

Spectroscopic studies interpreted based on model calculations suggest that these 1-D cuprates exhibit spin-charge separation [14-16]. Angle-Resolved Photoemission Spectroscopy (ARPES) shows that the hole bandwidth is much larger in 1-D than 2-D contrary to the expectation of LDA-type model for electronic structure [15]. The quasiparticle decay modes, probed by ARPES, are interpreted as spinons and holons [15]. Since holon

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**FIG. 1.** Inelastic x-ray scattering spectra along the chain direction (the Cu-O bond direction) are shown as a function of $q$ scanning over the entire Brillouin zone. The values of $q$ for the spectra bottom to top are 1.10$\pi$, 1.55$\pi$, 2.05$\pi$, 2.45$\pi$, 2.75$\pi$ and 3.05$\pi$ respectively. The spectrum for $q \sim 1.1\pi$ was taken with a resolution of about 490 meV whereas the rest were taken with 325 meV energy resolution. The right panel focuses on the lower energy feature (elastic scattering removed by fitting). $E_o = 8.996$ KeV.

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**FIG. 2.** (Left panel) Momentum dependence of the experimental inelastic features are compared with different types of numerical calculations [21,16,19] based on the Hubbard model. Data points with square symbols are from an independent experimental run not shown in Fig.1. (Right panel) Comparison of $q$-dependence of charge excitations along the Cu-O bond direction in 1-D and 2-D [10] is shown. All the $q$ points are plotted in their equivalent positions within 0 to $\pi$ for comparison. Charge excitations are found to be more dispersive in 1-D than 2-D.
is a collective charge mode it would couple to the x-rays strongly and exhibit its characteristic $q$-dependence. It is interesting to note that $q$-dependence of the Mott feature in 1-D is larger than it is along the bond direction in 2-D we studied earlier [10]. We compare this behavior in Fig. 2(Right panel). 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 Hubbard model [19,21]. In Fig. 2(Left panel) we compare our experimental results to the expectations from 1-D half-filled spin-1/2 Hubbard model describing charge fluctuation spectrum at finite-$q$ [16,19,21]. Within the level of energy resolutions the results are qualitatively described by (or at least consistent with) excitations involving holons in 1-D Hubbard model. It is interesting to note that these results are qualitatively consistent with electron scattering (nonresonant) studies of 1-D cuprates mainly in the low-energy regime [16].

$q$-dependent charge fluctuations in 1-D Mott insulators indicate that in 1-D Mott gap is of direct nature and excitations at the gap edge are more dispersive as compared to 2-D. The dispersions are also consistent with models describing the motion of holons in 1-D spin-1/2 Mott insulators. These results suggest that inelastic x-ray scattering can be used to study electronic structure of complex insulators and correlated electron systems in general. Higher resolution experiments would be necessary to extract quantitative details about the fundamental electronic parameters using such spectroscopies. Development of high brightness fourth generation synchrotrons can potentially make such experiments feasible with energy resolution in the millivolt regime.

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