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DIRECT SPECTROSCOPIC EVIDENCE OF HOLONS IN A QUANTUM ANTIFERROMAGNETIC SPIN-1/2 CHAIN

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We report charge dynamics of a quantum spin-1/2 chain ($SrCuO_2$) over the complete Brillouin zone using the momentum tunability of inelastic X-ray scattering. Excitations at the insulating gap edge are found to be highly dispersive. Observed dispersions are consistent with charge fluctuations involving holons unique to 1-D spin-1/2 quantum systems.

Keywords: Resonant; inelastic x-ray scattering; Holon; correlated electrons; Mott insulators.

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 synchotron facilities, inelastic X-ray scattering has been developing as a tool to study the BULK electronic structure (unlike electron based techniques) of condensed matter systems.[4–7] 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 experiment quite difficult. We have recently demonstrated 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 in a momentum-resolved manner.[6,7]

One-dimensional (1-D) 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.[8–11] 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.[9,12] 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.[1–3] The momentum dependence of the Mott gap (effective charge-transfer gap) has been reported recently in a parent 2-D cuprate using inelastic X-ray scattering over the full Brillouin zone.[6] We report study of 1-D Mott insulator's charge fluctuation spectrum by varying 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 $q \sim 0$) within the level of experimental resolution and the charge fluctuations at the gap edge are highly dispersive along the Cu-O-Cu bond direction.

The experiment was performed using the high flux undulator beamlines 12-ID (BESSRC-CAT) and 9-ID (CMC-CAT) at the Advanced Photon Source of Argonne National Laboratory reflecting improvement in signal-to-noise ratio of the data over our earlier results. Inelastic scattering was measured by varying q along the chain direction of single crystalline $SrCuO_2$. Overall energy resolution of 350 meV was achieved for this experiment. Incident beam was set near the Cu K-edge as obtained from the fluorescence spectrum for gaining resonant enhancement of excitation features. The scattered beam was reflected from a diced Ge-based analyzer and focused onto a solid-state detector. For q-scans, incident energy was kept fixed and q was varied by rotating the entire spectrometer around the scattering center. Background was measured by keeping track of scattering intensities on the energy gain side which was less than 5 counts per minute. Crystals used for this experiment were grown and characterized by techniques described previously which confirmed its quasi-one dimensionality above 6 K using different methods.[11,12] Figure 1 (Left panel) 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 ($E_o = 8.997$ KeV). Each spectrum shows two features, one around 5.6 eV (reported elsewhere[7]) and another, lower in energy, appears in the range of 2.5 to 3.5 eV depending on different values of the scattering wave vector q. In this paper we focus on the lower energy feature which 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 ($q \sim 0$) to the edge of the zone ($q \sim \pi$). For the q-scans incident energy was set fixed at 8.997 KeV. Inelastic features were also seen for other incident energies within 10 eV above and below 8.997 KeV.

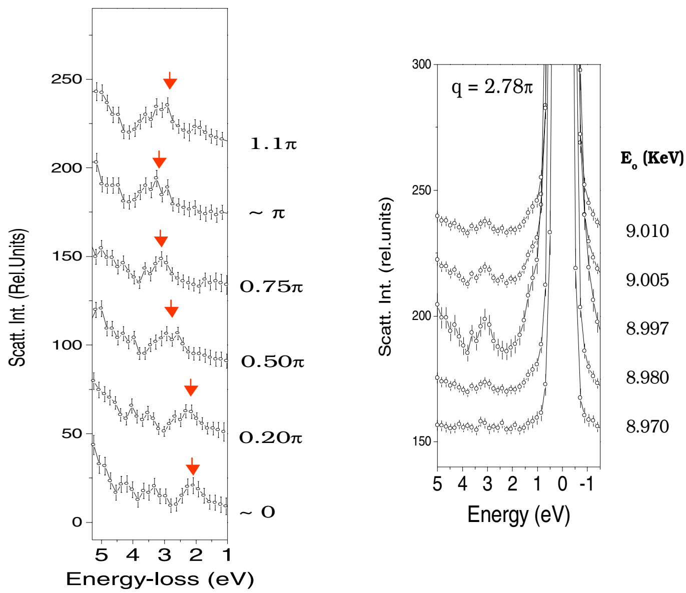

Fig. 1. (Left) Inelastic X-ray scattering spectra along the chain direction. The approximate values of q are noted right to each spectrum where 0 is taken from 2π. Quasi-elastic scattering has been removed by standard procedure.[6] (Right) Plot shows the energy dependence of inelastic excitations at fixed $q = 2.78\pi$. Best inelastic enhancement is observed around 8.997 KeV which was used for the q-scans.

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.[4–7] 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 states and an electron in the unoccupied states bound by the Coulomb interaction. We interpret the dispersion of the low energy feature seen in our data as the dispersion of the effective Mott gap (charge-transfer type[13]) in the system. The particle-hole pair formed in the process absorbs the energy and momentum lost from the incident X-ray beam and propagates with momentum $hq/2\pi$. The propagation of this pair would depend on the charge and spin distributions in the system. $SrCuO_2$ is believed to be quasi-1-D half-filled spin-1/2 Mott insulator with short-range antiferromagnetic spin correlations (quantum Heisenberg systems)[11] and has a Cu-O bond length very much similar to the analogous 2-D system ($Ca_2CuO_2Cl_2$) we studied earlier.[6,18] Spectroscopic studies interpreted based on model calculations suggest that these 1-D cuprates exhibit spin-charge separation.[8–12] Angle-Resolved Photoemission Spectrocopy shows that the hole bandwidth is much larger in 1-D than 2-D contrary to the expectation of LDA-type

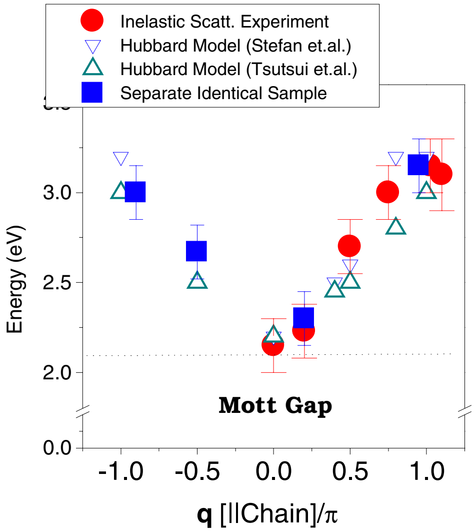

Fig. 2. The momentum dependence of the experimental inelastic features are compared with different types of numerical calculations[16,17] based on the Hubbard model. The full spectrum corresponding to some of the data points (solid rectangles) are not shown in Fig. 1.

model for electronic structure.[12,14] We find that q-dependence of the Mott feature in 1-D is larger than it is along the bond direction in 2-D reported earlier.[6] Such a behavior would be qualitatively expected from a system with spin-charge separation in the sense that charge fluctuations are free to move when they decouple from the spin and exhibit more dispersion. Furthermore, this is seen from numerical studies of Hubbard model.[15–17] In Fig. 2 we compare our experimental results to the expectations from 1-D half-filled spin-1/2 Hubbard model describing charge fluctuations at finite-q. Within the level of error-bars 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.[15]

The q-dependent charge fluctuations in a 1-D Mott insulator indicates that in 1-D Mott gap is of direct nature and excitations at the gap edge are more dispersive and consistent with models describing the motion of holons in 1-D. A cartoon view of low-energy excitations is shown in Fig. 3. Inelastic X-ray scattering couples directly to the charge component of the excitations (holons) but extremely weakly to the spin component and hence not visible in our data (besides the fact that it appears a much lower energies where electronic excitations are musked by the quasielastic tail of the present analyzers). Higher resolution experiments would be necessary to extract quantitative details about the fundamental electronic parameters using such spectroscopies. Developments of high brightness synchrotrons and efficient X-ray optics can potentially make such experiments feasible with resolution in the millivolt regime.

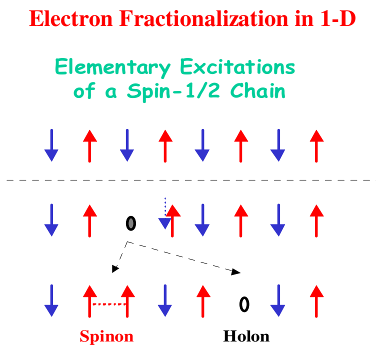

Fig. 3. A cartoon view of low-energy excitations in spin-1/2 antiferromagnetic chains. X-rays couple directly to the charge excitations (holons).[7,12]

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