Holon-Doublon (Anti-holon) Pair Excitations in 1-D Mott Insulators Studied by Resonant Inelastic X-ray Scattering

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Introduction

After several decades of research, a comprehensive understanding of the electronic structure of late transition metal oxides is still lacking. The existence of exotic electronic, magnetic, and optical properties — high T_c superconductivity as exhibited by the cuprates, 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]. Recent experimental and theoretical investigations have shown that one can measure the momentum dependence of electronic pair excitations by working near a resonance [5, 7-10].

Methods and Materials

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, charge fluctuations in these systems propagate rather freely and independently of the spin fluctuations [11-15]. Because of the insulating gap, charge fluctuations are at relatively high energies in Mott insulators [1, 2]. This report discusses the first study of the 1-D Mott insulator's charge fluctuation spectrum by varying q over the entire Brillouin zone by using high-resolution inelastic x-ray scattering. The experiment was performed using the BESSRC high-flux undulator beamline 12-ID and CMC-CAT beamline 9-ID at the APS. An overall energy resolution of 325 meV was achieved for this experiment. This is an improvement over our earlier work on 2-D Mott systems by more than 100 meV [8, 10, 20]. 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 about 2-3 counts per minute. Sr_2CuO_3 and $SrCuO_2$ crystals used for this experiment were grown and characterized by techniques described previously, which confirmed its quasi-one-dimensionality above 6K (Neel transition due to 3-D coupling) from using different methods [14, 15].

Results

Figure 1 shows inelastic x-ray scattering spectra with varying momentum transfers along the chain direction, with the incident energy fixed near the Cu K edge (E_o = 8.996 keV). Each spectrum shows two features, one around 5.6 eV and another, lower in energy, in a 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 ground state to the anti-bonding-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 when q is changed. The feature disperses upward in energy by about 1 eV monotonically over the Brillouin zone while going from the zone center ($q \sim 2\pi$) to the edge of the zone ($q \sim \pi, 3\pi$).

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]. The dispersion of the low-energy feature seen in our data is the dispersion of the

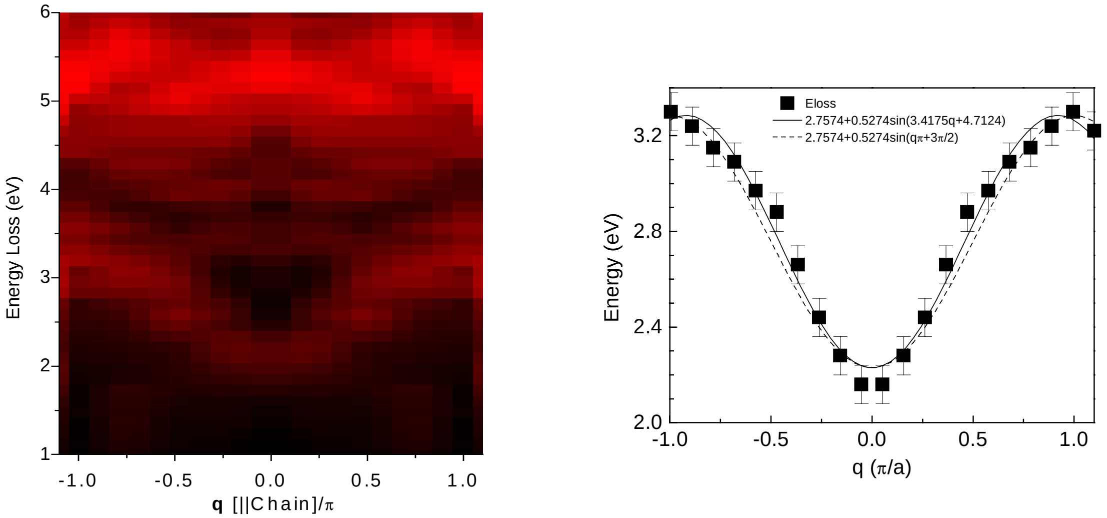

FIG. 1. Image graph (left) of inelastic x-ray scattering spectra near the Cu K edge along the chain direction (the Cu-O bond direction) (red is high intensity, black is low intensity). The corresponding dispersion plot is shown on the right. Data were taken at CMC-9-ID.

effective Mott gap in the system. The propagation of a particle-hole pair would depend on the charge and spin distributions in the system. Sr_2CuO_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] that have a Cu-O bond length very similar to the bond length of $Ca_2CuO_2Cl_2$ studied earlier [10, 14]. Spectroscopic interpretations 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 the local density approximation model for electronic structure [15]. It is interesting to note that q-dependence of the Mott feature is larger in 1-D than it is along the bond direction in 2-D. We compare this behavior in Fig. 2. Such 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. This is also seen from numerical studies of the Hubbard model [19, 21]. In Fig. 2, we compare our experimental results to the expectations from a 1-D half-filled spin-1/2 Hubbard model describing charge fluctuation spectrum at finite q [16, 19, 21]. Within the level of error-bars, the results are qualitatively described by, or at least consistent with, excitations involving holon pairs (holon-doublon) in the 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].

Discussion

Our results show that the charge fluctuations in a 1-D Mott insulator consist of holon-doublon (anti-holon) pairs. These results suggest that inelastic x-ray scattering can be used to study the electronic structure of complex insulators and correlated electron systems in general.

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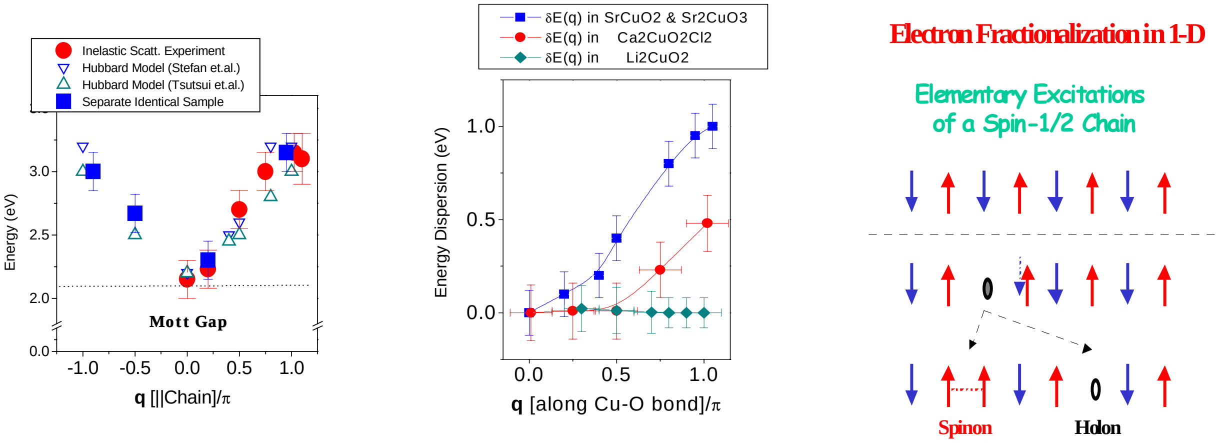

FIG. 2. In the left graph (© 2003 World Scientific Publishing Corp.), the momentum dependencies of the experimental inelastic features are compared with different types of numerical calculations [16, 19, 21] based on the Hubbard model. Data points with symbols of nested circles are from an independent experimental run not shown in Fig. 1. The middle graph (© 2002 by the American Physical Society) shows a comparison of dispersions (bond direction) in 1-D with 0-D and 2-D [10]. In the figure on the far right (© 2003 World Scientific Publishing Corp. and © 2002 by the American Physical Society) is a depiction of low-lying excitations in a 1-D spin-1/2 lattice.

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