Resonant Inelastic x-rays Scattering Measurement on the Momentum-Structure of Remnant Mott-gap in Hole Doped Cuprates

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Introduction

High resolution resonant inelastic x-ray scattering (RIXS) is a recently developed x-ray spectroscopy[1, 2] used to measure the momentum resolved charge excitation over the Mott gap in the Mott insulater[3] and it's doped offsprings. In the RIXS experiment, the incident energy is tuned near and absorption edge so that the inelastic signal can be drastically enhanced. Besides, the knowledge of absorption feature where the incident energy is selected helps in figuring out the nature of the excitation corresponding to the enhanced inelastic signal. In 2D cuprate, in particular, the Mott-gap is of charge-transfer type[4] and the resonant energy of the excitation is located around the Cu *K* absorption feature which is characterized by the well screened final state. We performed RIXS measurement on both undoped insulator single crystal La_2CuO_4 and hole doped single crystal compounds $La_{2-x}Sr_xCuO_4$ (x=0, 3%, and 14%) and a systematic doping evolution of the Mott gap structure is revealed.

Methods and Materials

In our RIXS measurement on hole-doped $La_{2-x}Sr_xCuO_4$ systems, the resonant incident energy was selected near the Cu *K* absorption edge. Before searching for the resonant incident energy, an absorption curve at the Cu *K* edge is obtained by measureing the fluorescence from the sample. Among different inelastic signals enhanced with different incident energy, the charge transter excitation is identified by finding the resonant energy near the absorption feature which is associated with the well screened final state. With resonant incident energy identified, the momentum dependent measurments were then performed and dispersion relation of the charge transfer excitations were obtained.

The measurement were carried out at BESSRC beamline 11-ID and CMC beamline 9-ID. The instruments are typical triple-axis spectometers. With the incident energy fixed at the resonant value, the scattered photon energy is scanned by Braggly reflecting the scattered photons from a Ge (733) crystal analyzer. The measurements were performed along [100] direction at room temperature. The overall energy resolution is about 370meV.

Results

Our RIXS measurements on hole doped cuprates were performed at Cu *K* edge. Fig. 1 shows the Cu *K* absorption edge for the undoped insulator La_2CuO_4 obtained by measuring the fluorescence with incident polarization along and perpendicular to *c*-axis. The absorption structure has strong polarization dependence. When the polarization along *c*-axis, the absorption structure is dominated by the transition from Cu 1*s* to $4p_\sigma$ level with higher energies. In the case of polarization perpendicular to *c*-axis, it is dominated by the 1*s* to $4p_\pi$ transition with lower energies. In each polarization case, the structure is consisted of two major features, the higher energy feature is believed to come from transition to the poorly sceened final state in which the Cu has a $3d^9$ configuration and the lower energy feature is from the transition to the well screened final state which as a $3d^{10}$ configuration[5]. The well screened final state appears as a result of transfering an electron from the neighboring oxygen atom to the Cu site, and consequently the charge transfer excitation measured in the RIXS is expected to be enhanced around the energy value of this feature.

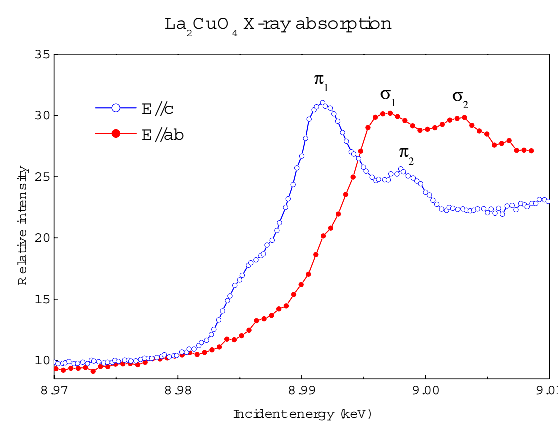

Fig. 1: Cu *K*-edge x-ray absorption curves for La_2CuO_4 single crystall measured wthi incident photon polarization along *c*-axis (empty circle) and perpendicular to the *c*-axis (solid circle).

This is indeed the case, as can be seen in Fig. 2. The image plot of Fig. 2 shows the incident energy dependence of the intensity of the inelastic signals. With increased incident energy, the lowest energy feature around 2 to 4eV energy-loss, which should correspond to the Mott gap excitation, first appears around 8.990keV, which is the energy value of the absorption feature with the well screened final state. This double peak inelastic feature dies off when the incident energy goes past this absorption feature. Then at the incident energy corresponding to the poorly screened final state, a higher energy feature around 7eV energy-loss is maximized. To investigate the charge transfer excitation, we choose 8.990keV as the incident energy in the momentum dependent measurement. The 7eV feature, which has no charge transfer included, could be the excitation from the real lower Hubbard band to the upper Hubbard band. The absorption structure for the doped systems are quite similar to the undoped one especially when the peak position only is concerned. The lowest energy excitation in the doped system appears at the same incident energy as undoped insulator.

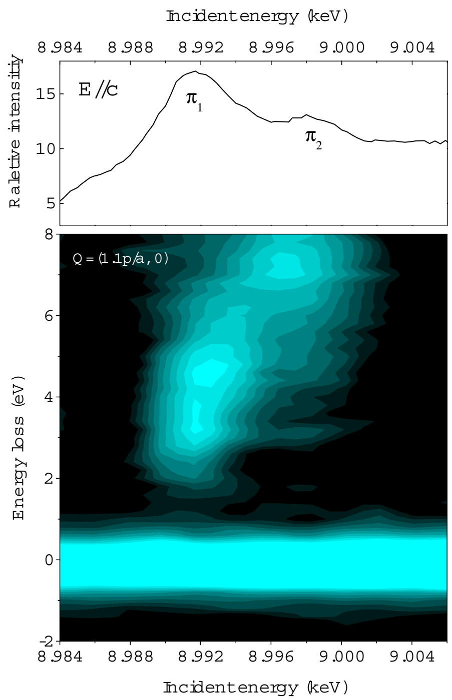

Fig. 2: Resonance profiles of inelastic spectra plotted in images for single crystal La_2CuO_4. Incident polarization is along *c* axis. The absorption curve is plotted on top of the image for La_2CuO_4 for comparison. π_1 and π_2 on the absorption curve labels the features with well screened and poorly screened final state, respectively.

With the resonant energy determined, we carried out the momentum-dependent measurement of the charge transfer excitation over Mott gap in both undoped insulate La_2CuO_4 and hole doped compounds, all along [100] direction and in room temperature. Fig. 3 (left) shows energy loss curves for both undoped insulator La_2CuO_4 and 14% hole doped $La_{1.86}Sr_{0.14}CuO_4$ that are selected near the high symmetry points along [100] direction. For the undoped sample, a double peak feature appear between 2 and 5eV energy-loss and they monotonically disperse within the whole Brillouin zone with different dispersion amount, the lower energy feature dispersing more than the higher energy feature. Upon doping, the excitation feature evolve in two ways. Firstly the lower energy feature starts melting but it continues to exist till the 14% doped system, showing a remnant gap structure even in the highly doped metallic phace. The higher energy peak, on the other hand, does not show significant change both in intensity and in position. Secondly, accompanying with the melting of the Mott gap, a continuum feature builds up in the gap as evident from the appearing and increase of the spectrum weight within the excitation gap (Fig. 3 right).

The doping evolution of the dispersion of the charge transfer excitation is illustrated in Fig. 4 by plotting the leading-edges of lower-energy feature as a function of the momentum. The leading-edge points were determined from the first maximum of the derivative of the energy loss curves. It is seen that the undoped insulator has a maximum dispersion of about 500 meV. The dispersion then decrease significantly with doping to about 100 to 150 meV at 14% doping. The dispersion reduction with doping is a result of the lifting of the bottom of the excitation band located at the zone center.

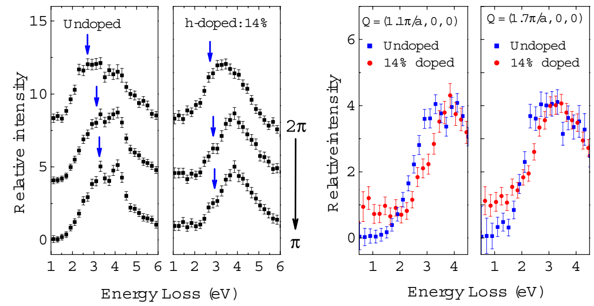

Fig. 3 Momentum dependence of gap excitations. Top: Image plots showing the excitation band in *Q* vs *E* plane along [100] direction for various doping levels. The images are made from the *Q*-dependent energy-loss data with elastic signal removed by fitting. The brightness represents the spectrum intensity. The dotted line indicates the dispersion of the leading edge. Bottom: Comparison of selected energy-loss curves at high symmetry points between undoped and highly doped compounds.

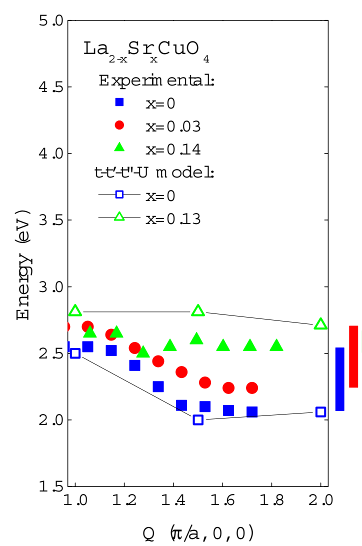

Fig. 4: Leading-edge dispersion for various hope-doping and comparison with *t-t′-t″-U* model. The bars on right side illustrate the excitation bandwidths.

Discussion

The RIXS process at *K*-edge involves the transfer of a Cu 1*s* electron to the 4*p* level by absorbing an incident photon with energy ω_1 and momentum $\boldsymbol{k}_1$. Then in an intermediate state, due to the interaction with the 1*s* core hole and 4*p* excited electron, an excitation in the 3*d* electron system occurs. Finally the 4*p* electron transfers back to the 1*s* level in an excited state, emitting a photon of energy ω_2 and momentum $\boldsymbol{k}_2$ [6]. The energy loss and the momentum transfer of the photon measures the energy and the momentum of the excitation of the 3*d* electron system. An important feature of the RIXS is the correlation between the excitation in the 3*d* electron system and the Cu *K*-edge absorption structure, the intermediate state of the RIXS being the final state of the latter. This relation between the K absorption edge and the RIXS process helps to identify the nature of an observed excitation of the $3d$ electron system. In Fig. 1, the absorption curve for each polarization consists of two features. The higher energy feature comes from the transition to poorly screened final state in which the Cu d electrons have a configration of $3d^9$. If in the transition final state, an electron from the neighboring oxygen atom transfered to the Cu site and results in a configration of $3d^{10}$, the state is then well screened and has a lower energy. Consequently an absorption feature with lower energy appears (π_1 and σ_1 in Fig. 1). The excitation enhanced at this energy then is expected to be of charge transfer type. In the energy dependent scans as shown in Fig. 2, the lowest energy excitation, which should be the Mott gap excitation in the Mott insulator, does appear around the well screened absorption feature, and this actually confirms that the insulater gap of cuprate system is indeed of charge transfer type.

The doping evolution of the Mott gap excitation as observed here involves a melting of the Mott gap with doping but a remnant Mott gap charactor persists until the highly doped metallic phase. This observation combined with the ARPES result showing a remnant Fermi surface in the undoped insulator[7] may shine a light on future theoretical consideration. The continuum feature appears in the doped system may comes from the excitation originated from the conduction electrons. We also look for possible correlation of excitation bandwidth with magnetic structure of the system. We noticed that the decreasing of the excitation band with doping has a direct scaling to the doping dependence of the Neel temperature. This is an indication that the propagation of the excited particle-hole pair is closly related to the antiferromagnetic charactor of the lattice structure. Finally we compare the experimental result with the t-t'-t''-U model[8]. As shown in Fig. 4, the calucated result is roughly in agreement with the experimental result. For highly doped sample, however, there is deviation in simulation from the experimental result especially in the middle of the Brillouin zone.This could be due to other instabilities such as charge stripes in this area in reciprocal space [9]. More detailed work is necessary to establish such connections.

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