

Resonant inelastic x-ray scattering in one-dimensional copper oxides

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(Received 23 September 1999)

The Cu *K*-edge resonant inelastic x-ray scattering (RIXS) spectrum in one-dimensional insulating cuprates is theoretically examined by using the exact diagonalization technique for the extended one-dimensional Hubbard model with nearest-neighbor Coulomb interaction. We find the following characteristic features that can be detectable by RIXS experiments: (i) The spectrum with large momentum transfer indicates the formation of excitons, i.e., bound states of *holon* and *doublon*. (ii) The spectrum with small momentum transfer depends on the incident photon energy. We propose that the RIXS provides a unique opportunity to study the upper Hubbard band in one-dimensional cuprates.

Insulating copper oxides, when mapped onto a single-band component,¹ have their occupied lower Hubbard band (LHB) and the unoccupied upper Hubbard band (UHB) separated by a Mott gap. Angle-resolved photoemission spectroscopy (ARPES) has succeeded in observing the momentum dependence of the LHB for two-dimensional (2D) (Ref. 2) and 1D (Ref. 3) cuprates. In contrast to the LHB, the UHB is far from our understanding.

The information about the UHB can be extracted from excitations from the LHB to UHB across the Mott gap. A representative technique for this is electron energy-loss spectroscopy (EELS), which has been applied to both 2D (Ref. 4) and 1D (Ref. 5) insulating cuprates. Another technique is resonant inelastic x-ray scattering (RIXS). When the incident photon energy ω_i is tuned through the Cu *K* absorption edge, the momentum dependence of the excitations from the LHB to UHB has been observed for insulating 2D cuprates, $\text{Sr}_2\text{CuO}_2\text{Cl}_2$ (Ref. 6) and $\text{Ca}_2\text{CuO}_2\text{Cl}_2$ (Ref. 7). On the theoretical side, the present authors predicted the dependence based on a 2D Hubbard model with second- and third-neighbor hopping terms.⁸

Typical examples of 1D cuprates are Sr_2CuO_3 and SrCuO_2 . Their magnetic susceptibilities are explained by the 1D Heisenberg model.⁹ ARPES measurements^{3,10} showed that a photodoped hole cannot exist as a quasiparticle, but changes into two collective excitations, a *spinon* and a *holon*, as expected from the 1D single-band Hubbard and *t*-*J* models. From EELS experiments,⁵ it has been pointed out that an extended version of the Hubbard model with a moderate nearest-neighbor Coulomb interaction *V* is necessary to understand excitations across the Mott gap. It is interesting to know what we obtain in the RIXS spectrum prior to the experiments.

In this study, the momentum dependence of the RIXS spectrum in 1D cuprates is demonstrated by using the exact-diagonalization technique for the extended Hubbard model. We find the following characteristic features that can be detectable by RIXS experiments: The spectral weight concentrates on the narrow energy region due to the formation of excitons. This is also seen in the dynamical charge-correlation function, although the dependence of the intensity on the momentum transfer is different. We also find that the spectral weight distribution for a small momentum trans-

fer depends on the incident photon energy. This is associated with the intermediate state that is characteristic of the RIXS process.

A minimal model that can describe the LHB and UHB is the extended Hubbard model as used in the analyses of the EELS spectra.⁵ Hereafter, we use the term “3*d*-electron system” to stand for the LHB and UHB. The Hamiltonian is given by

$$H_{3d} = -t \sum_{i,\sigma} (d_{i,\sigma}^\dagger d_{i+1,\sigma} + \text{H.c.}) + U \sum_i n_{i,\uparrow}^d n_{i,\downarrow}^d + V \sum_i n_i^d n_{i+1}^d, \quad (1)$$

where $d_{i,\sigma}^\dagger$ is the creation operator of 3*d* electron with spin σ at site *i*, $n_{i,\sigma}^d = d_{i,\sigma}^\dagger d_{i,\sigma}$, and $n_i^d = n_{i,\uparrow}^d + n_{i,\downarrow}^d$. The on-site Coulomb energy *U* corresponds to the charge-transfer energy of cuprates.⁸

In the intermediate states of the Cu *K*-edge RIXS process, 3*d* electrons interact with a 1*s*-core hole created by the dipole transition of an 1*s* electron to 4*p* orbital due to absorption of an incident photon with energy ω_i and momentum $\mathbf{K}_i$. This interaction is written as

$$H_{1s-3d} = -V_c \sum_{i,\sigma,\sigma'} n_{i,\sigma}^d n_{i,\sigma'}^s, \quad (2)$$

where $n_{i,\sigma}^s$ is the number operator of the 1*s*-core hole with spin σ at site *i*, and V_c is taken to be positive. This interaction V_c causes excitations of 3*d* electrons across the gap. The photoexcited 4*p* electron is assumed to go into the bottom of the 4*p* band with momentum $\mathbf{k}_0$ and not to interact with either the 3*d* electrons or 1*s*-core hole due to the delocalized nature of the 4*p* orbital.⁸ In the final state, the 4*p* electron goes back to the 1*s* orbital, emitting a photon with energy ω_f and momentum $\mathbf{K}_f$. The RIXS spectrum is then given by⁸

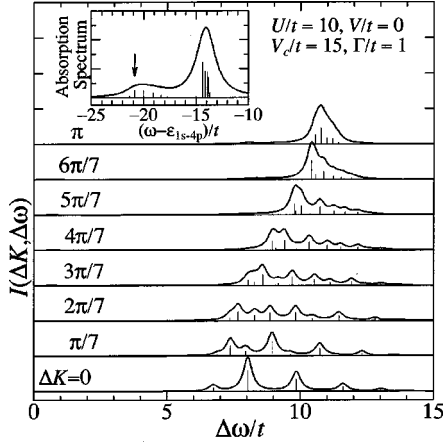


FIG. 1. Resonant inelastic x-ray scattering spectra for the Cu K edge in the 1D half-filled Hubbard model. The spectra of the elastic-scattering process at $\Delta K=0$ are not shown. The parameters used are $U/t=10$, $V/t=0$, $V_c/t=15$, and $\Gamma/t=1$. The δ functions (the vertical thin solid lines) are convoluted with a Lorentzian broadening of $0.2t$. The inset is the Cu 1s absorption spectra with a broadening of $1.0t$, and the incident photon energy ω_i is set to the value denoted by the arrow.

$$I(\Delta K, \Delta \omega) = \sum_{\alpha} \left| \langle \alpha | \sum_{\sigma} s_{\mathbf{k}_0 - \mathbf{K}_f, \sigma} p_{\mathbf{k}_0, \sigma} \right. \\ \times \frac{1}{H + \varepsilon_{1s-4p} - E_0 - \omega_i - i\Gamma} p_{\mathbf{k}_0, \sigma}^{\dagger} s_{\mathbf{k}_0 - \mathbf{K}_i, \sigma}^{\dagger} | 0 \rangle \left. \right|^2 \\ \times \delta(\Delta \omega - E_{\alpha} + E_0), \quad (3)$$

where $H = H_{3d} + H_{1s-3d}$, $\Delta \mathbf{K} = \mathbf{K}_i - \mathbf{K}_f$, $\Delta \omega = \omega_i - \omega_f$, $s_{\mathbf{k}, \sigma}^{\dagger}$ ($p_{\mathbf{k}, \sigma}^{\dagger}$) is the creation operator of the 1s-core hole (4p electron) with momentum $\mathbf{k}$ and spin σ , $|0\rangle$ is the ground state of the half-filled system with energy E_0 , $|\alpha\rangle$ is the final state of the RIXS process with energy E_{α} , Γ is the inverse of the relaxation time in the intermediate state, and ε_{1s-4p} is the energy difference between the 1s level and the bottom of the 4p_z band. The momentum component parallel to the 1D Cu chain is represented by ΔK hereafter. The values of the parameters are set to be $U/t=10$, $V_c/t=15$, and $\Gamma/t=1$ as for the 2D cuprates.⁸ The RIXS spectrum in Eq. (3) is calculated on a 14-site ring by using a modified version of the conjugate-gradient method together with the Lanczö's technique.

Figure 1 shows the momentum dependence of the RIXS spectrum for the case of $V/t=0$. The inset shows the absorption spectrum of the 1s electron to the 4p orbital.⁸ The incident photon energy ω_i that appears in Eq. (3) is set to be the value denoted by the arrow. This means that the states near the arrow satisfy the resonance conditions of Eq. (3). For small ΔK , the spectra spread over about $8t$. The energy region, however, shrinks with increasing ΔK , and the spectral weight concentrates on a narrow energy region at $\Delta K=\pi$. This momentum dependence is similar to that of the dynamical charge-correlation function $N(q, \omega)$, shown by dashed lines in Fig. 2, which is explained by the particle-hole model with only a charge degree of freedom.^{5,11} In the particle-hole model, *holon* and *doublon* bands separated by U describe the LHB and UHB, respectively. We note that the spin degree of

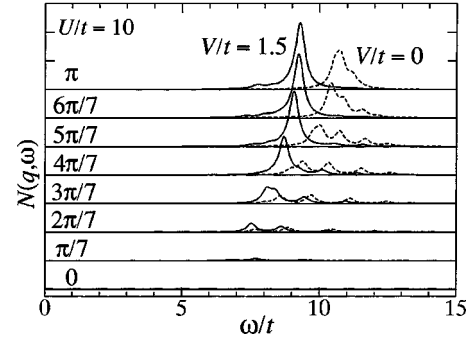


FIG. 2. Dynamical charge correlation function $N(q, \omega)$ in the 1D half-filled Hubbard model. The parameters used are $U/t=10$, $V/t=0$ (broken lines), and $V/t=1.5$ (solid lines). The δ functions are convoluted, with a Lorentzian broadening of $0.2t$.

freedom is less effective on $N(q, \omega)$ because of the wavefunction factorization¹² in the large- U limit. At the zone center of the interband transitions, one obtains a continuum with a width of $8t$ above the gap of $U-4t$. The width of the continuum decreases with an increase of the momentum transfer, and finally becomes an excitation with energy U at the zone boundary. This momentum dependence is qualitatively consistent with that shown in Fig. 1 and also with $N(q, \omega)$ shown in Fig. 2. It is interesting to note that the integrated weight of $I(\Delta K, \Delta \omega)$, with respect to $\Delta \omega$, is almost independent of ΔK in contrast to $N(q, \omega)$ where the integrated weight is proportional to $\sin^2(q/2)$ for the large- U region.¹¹

The effect of the nearest-neighbor Coulomb interaction V on $I(\Delta K, \Delta \omega)$ is shown in Fig. 3, where $V/t=1.5$ is taken according to the analyses of the EELS data.⁵ From the comparison between Figs. 1 and 3, we find that the shape of the spectrum changes remarkably for $\Delta K > \pi/2$, accompanied by the formation of sharp peaks together with the shift of the spectral weight to the lower-energy region. This is due to excitons that appear at the momenta satisfying a condition that $V > 2t \cos(\Delta K/2)$.¹¹ A similar change is also seen in $N(q, \omega)$ (see Fig. 2). For $\Delta K < \pi/2$, the intensity of the spectrum near the lower edge is enhanced by the presence of excitonic effects, which is again similar to the feature of $N(q, \omega)$.¹¹ We note that the presence of V also affects the

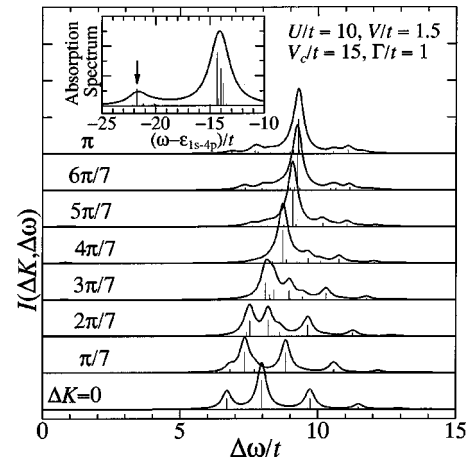


FIG. 3. The same as Fig. 1, but for $V/t=1.5$.

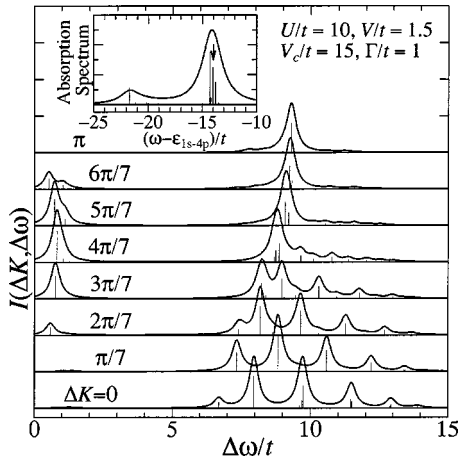


FIG. 4. The same as Fig. 3, but with the incident photon energy ω_i set to the value denoted by the arrow in the inset.

RIXS spectrum in the 2D case, although the edge structure, which is a fingerprint of the shape of the UHB,⁸ does not depend on V .

The spectral weight depends on the incident photon energy ω_i . By changing $\omega_i - \varepsilon_{1s-4p}$ from about $-21t$ to $-14t$, the distribution of intensity in the lower-momentum region shifts to the higher-energy region, as shown in Fig. 4. This can be attributed to the nature of the intermediate state around the resonance condition, denoted by the arrows in the insets of Figs. 1 and 3. The peaks at $\omega - \varepsilon_{1s-4p} \sim -21t$ ($\sim U - 2V_c$) in the absorption spectrum are characterized by

states having configurations with the core-hole site doubly occupied by $3d$ electrons together with an empty site of $3d$ electrons. On the other hand, in states near $\omega - \varepsilon_{1s-4p} = -14t$ ($\sim -V_c$), the core-hole site is singly occupied by $3d$ electrons and there is no empty site. Since the different intermediate states are used depending on the incident photon energy, the RIXS spectrum depends on the energy. The spectrum in the low-energy region of $\Delta\omega/t \lesssim 1$ in Fig. 4 corresponds to singlet excitations of the spinon pair, because the intermediate state has singly occupied configurations.

In summary, we have examined the momentum dependence of the RIXS spectrum in 1D cuprates, which provides a unique opportunity to study the upper Hubbard band, using the exact diagonalization technique for the extended Hubbard model. We have found the following characteristic features of the RIXS spectrum: The spectrum with large momentum transfer, $\Delta K > \pi/2$, indicates the formation of excitons, i.e., bound states of a *holon* and a *doublon*. We have also found that the spectral weight distribution for small momentum transfer, $\Delta K < \pi/2$, depends on the incident photon energy. The dependence is associated with the intermediate state that is characteristic of the RIXS process. These findings will be good guides for RIXS experiments on 1D insulating cuprates that will be performed in the future.

This work was supported by Priority-Area Grants from the Ministry of Education, Science, Culture and Sport of Japan, CREST, and NEDO. Computations were carried out in ISSP, University of Tokyo; IMR, Tohoku University; and Tohoku University.

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