Studying Holon-Antiholon Continuum in $SrCuO_2$

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Introduction

The separation of spin and charge degrees of freedom is one of the most important and fascinating properties of electrons in strongly correlated systems in one dimension. In particular, it is well known that in the 1-D Hubbard model, the low-energy physics is dominated by collective excitations of decoupled charge and spin degrees of freedom called holons and spinons, respectively [1]. Experimentally, if one creates a hole by removing an electron, this hole is expected to decay into a spinon and a holon, which can be studied with angle-resolved photoemission spectroscopy (ARPES) [2]. The situation is different for so-called “particle-hole” probes, such as optical spectroscopy, resonant inelastic x-ray scattering (RIXS), and electron energy loss spectroscopy (EELS). In these experiments, total charge is conserved in the scattering process, so that an electron is simply moved from one site to another, creating a hole and a doubly occupied site. The decay of the hole creates a holon and a spinon, while the double occupancy decays into an antiholon and a spinon. Since photons and electrons strongly couple to the charge sector, the behavior of holon-antiholon pairs can be studied with these particle-hole probes [3, 4].

We report here on a RIXS study of charge excitations in the quasi-1-D Mott insulator $SrCuO_2$. We observe a continuum of excitations arising from the creation of particle and hole pairs. Within this continuum, a well-defined spectral feature with a large sinusoidal dispersion (~1.1 eV) is also observed. A comparison of the RIXS spectra with both the theoretical results and optical conductivity yields a consistent picture of charge excitations in this material: The excitation spectrum is described as a holon-antiholon continuum with a dispersive onset, together with a more dispersive broad resonance that is the remnant of the strong-coupling exciton.

Methods and Materials

In RIXS experiments, the incident x-ray energy is tuned to near the absorption edge of the particular element of interest, so that the inelastic scattering intensity of certain electronic excitations is resonantly enhanced. In addition, this gives an element specificity that is valuable for studying complex materials, such as the cuprate compounds [4-7]. The RIXS experiments were carried out at CMC-CAT beamline 9-ID at the APS with a double-bounce Si(333) monochromator. A spherical, diced, Ge(733) analyzer was used to obtain an overall energy resolution of ~0.4 eV full width at half-maximum (FWHM). The polarization of the incident x-ray was kept perpendicular to the Cu-O plaquette. Note that the corner-sharing CuO_2 chain runs along the c-direction. A single-crystal sample of $SrCuO_2$ was grown by using the traveling solvent floating zone method. The crystal was cleaved along the (0 1 0) plane and mounted on an aluminum sample holder at room temperature in an evacuated chamber. In our measurements, the incident photon energy was fixed at $E_i = 8982$ eV, while the final photon energy was varied to produce spectra as a function of energy transferred to the electron system.

Results

Our results are shown in Fig. 1, in which the RIXS intensity is plotted as a function of momentum and energy transfers. The most prominent feature of the RIXS spectra shown in Fig. 1 is the highly dispersive feature around $\omega = 3$ eV. The momentum dependence of the peak position of this feature exhibits a clear sinusoidal dispersion with a bandwidth of 1.1 eV. However, closer inspection of Fig. 1 reveals the presence of additional spectral weight on the low-energy side around the zone boundary ($q/2\pi = -0.5$) position. The best description of the observed spectra is that of a continuum of excitations, in which the dispersive feature resides. Note that the q-dependence of the onset energy of the continuum is smaller than that of the peak position. Our RIXS results suggest that the continuum starts at much lower energy (1.5 to ~2 eV) and that the particle-hole pairs do not form a bound exciton at $q = \pi$, in contrast to previous EELS and RIXS results [3, 4].

Discussion

The RIXS data are difficult to model because, at present, there is no completely satisfactory theory of RIXS. However, it is clear that the RIXS process involves charge fluctuations that may be represented by the low-energy excitations of the extended Hubbard model [8]. We have therefore compared our data with the dynamical density-density correlation function $N(q, \omega)$ calculated with numerical methods [9]. The parameters were

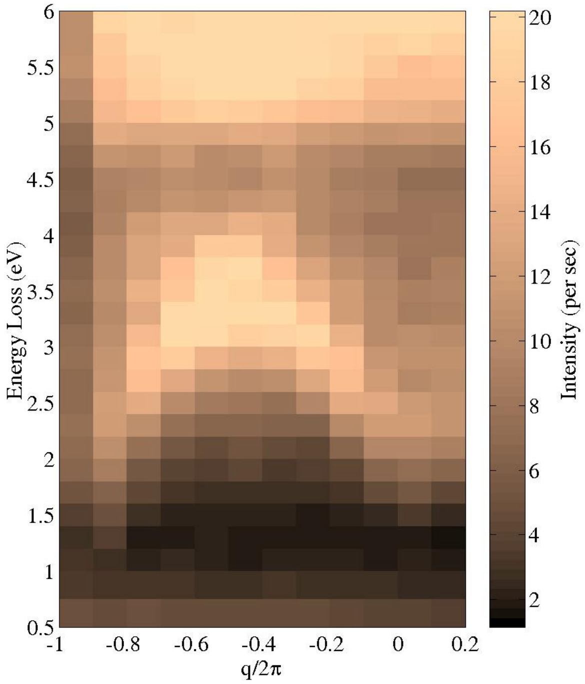

FIG. 1. The pseudocolor scale of the observed intensity is shown on the right side in units of counts per second. The momentum transfer is shown in reduced units of q/2π along the chain direction and corresponds to the (0 11 q) position.

independently determined from fitting the optical conductivity data. This realistic calculation suggests that the (strong-coupling) exciton acquires a finite lifetime and turns into a broad holon-antiholon resonance in $SrCuO_2$. The onset of the holon-antiholon continuum in N(q, ω) seems to lie in the area where the RIXS spectrum begins to show appreciable intensity, and the nonsinusoidal behavior of the onset energy of N(q, ω) seems to follow the momentum dependence of the RIXS onset energy (see Fig. 1). The dispersion of the peak positions from the calculation also seems to be very similar to that of the RIXS data, although this may well be coincidental. Clearly, further calculation of the full RIXS response function is necessary to draw any firm conclusions.

Acknowledgments

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