Exciton Dynamics in Insulating Cuprates and Nickelates

Beamline

SP12U1 Inelastic X-ray Scattering beamline

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In this work we use high resolution resonant inelastic X-ray scattering (RIXS) to investigate the dynamics of low energy excitations in correlated insulators. We obtain excellent overall resolution of 300 meV with a spherically bent Si analyzer crystal and a 2m diameter Rowland circle spectrometer. We measure the energy dispersion of the charge transfer exciton in two prototype charge transfer insulators, La_2CuO_4 and La_2NiO_4. The first is a parent compound for a high T_c superconductor while the second remains insulating for high levels of doping. From the measured dispersion of these excitations we find that the exciton is mobile in the cuprate while it is well-localized in the nickelate reflecting strong polaronic effects in the latter.

Electronic excitations in materials with strong electronic correlations are an important field of study because they provide information about the energy and dispersion of empty electronic levels. This information is complementary to that about occupied levels provided by spectroscopies such as photoemission and combining both, a reasonable idea about the electronic structure of these materials can be formed. Resonant inelastic X-ray scattering (RIXS) has been used in the last few years with this aim. In the resonant method the incident energy is chosen in the region of an absorption edge. This choice also determines the intermediate state of the scattering process which involves a core hole generated by the incident photon. Electronic decay follows which fills the core hole but may leave the system in an excited final state, resulting in inelastic features. Since the photon can exchange sizeable momentum with excitations in the system, their dispersion can be measured. The method is bulk sensitive, with an enhanced cross section due to the core-hole which however is absent from the final state, so that information relevant to the physics of the materials in question is directly obtained. However, the core hole selectively emphasizes localized processes.

In this work we study RIXS at the Cu-K absorption edge in La_2CuO_4 and at the Ni-K absorption edge in La_2NiO_4. The cuprate and the nickelate are almost completely iso-structural but with remarkably different physical properties. No superconductivity has been reported in the nickelate and metallic behavior arises at very high doping. Ni ions are embedded in O octahedra with a nominally $3d^8$ electronic configuration. It is known that Ni ions are high spin (HS), with both e_g orbitals $3d_{z^2-r^2}$ and $3d_{x^2-y^2}$ occupied by one electron and a total spin S=1 per Ni ion as opposed to S=1/2 in the cuprates.

The measurements were carried out at SPring-8 on the undulator beamline SP12U1 (NSRRC ID). Spherically bent 2m radius Si analyzers (Si 553 for the Cu K-edge and Si 551 for the Ni K-edge) were used to reach a

total resolution of 300 meV for the experiments. Figures 1(a) and (b) show RIXS scans with the momentum transfer parallel to the a/b axis in samples of La_2CuO_4 and La_2NiO_4 respectively. Each scan corresponds to a point in the BZ, between [4.5 0 0] and [5 0 0]. The scans have been displaced vertically for clarity and from top to bottom the momentum transfer varies from the center of the zone to the edge. The three point smoothed spectrum taken at the zone edge (dashed line) is used as a base-line for highlighting the behaviour of the lowest energy inelastic feature. In both compounds, between the elastic line (zero energy transfer) and a given energy (about 2 eV in the cuprate and about 4 eV in the nickelate) no inelastic features are present corresponding to the measured optical gaps of the charge-transfer type. This charge transfer and other processes across the gap result in the hump constituting the inelastic signal.

In Fig. 1(a) at an energy loss of 2.2 eV, a peak is seen in the scans for the cuprate (arrow) near the center of the BZ, which we assign to the fundamental charge transfer exciton. As the momentum transfer increases from the zone center to the zone edge, the peak disperses to higher energy loss and its intensity simultaneously vanishes. By subtracting the 3 point smoothed spectrum taken at the BZ edge from the spectra taken at various other points in the BZ the dispersion of the low energy peak is calculated and shown in Fig. 2(a). The exciton disperses quadratically (dashed line) in a free particle fashion by about 0.5 eV.

In the nickelate, though no clearly resolved peak emerges in Fig. 1(b), close inspection reveals that the scans nearer the zone center show a low energy shoulder (arrow) which vanishes near the zone edge. Following the analysis for La_2CuO_4, differences of the RIXS scans with respect to the zone edge scan (dashed lines) allow us to identify an excitonic peak at about 3.6 eV which loses intensity in going from zone center to edge. However no dispersion is seen (Fig. (2b)) indicating complete localization of the charge transfer exciton in La_2NiO_4.

In La_2CuO_4, the CuO_2 unit cell in the plane contains one hole on the Cu site. Consider a plaquette consisting of a central Cu atom and four surrounding O atoms. With the transfer of an electron from the O 2p orbitals on the plaquette, a neighbouring Cu site assumes the Cu $3d^{10}$ configuration. The O 2p hole and the transferred electron can then form a bound exciton. The dynamics of this exciton in the CuO_2 plane also depends on the interaction of the O 2p hole and the $3d^9$ hole on the central Cu of the plaquette. These two holes can interact to form a singlet state or a triplet state. An important consequence of singlet formation on the plaquette is that the exciton which consists of two spinless Cu sites moves through the CuO_2 plane without upsetting the antiferromagnetic (AF) S=1/2 background. In the nickelate charge transfer can also potentially result in the formation of a bound exciton and to a spin zero Ni site for a singlet coupling between the spins of the Ni $3d^9$

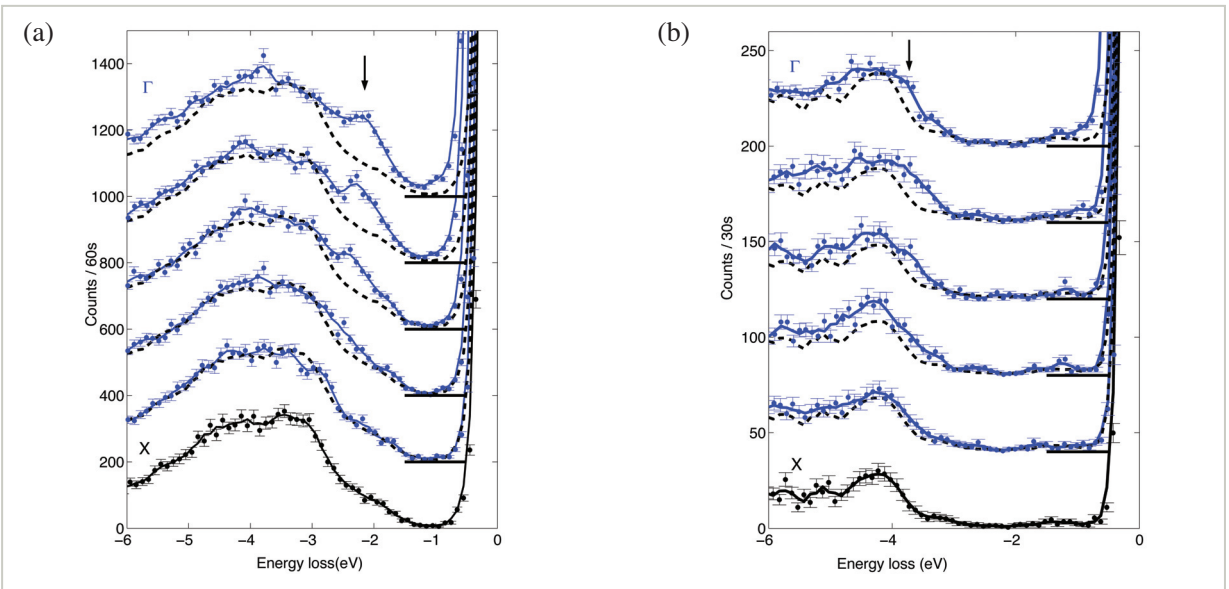

Fig. 1: RIXS in (a) La_2CuO_4 and (b) La_2NiO_4 along the [100/010] direction. Spectra have been shifted vertically, and the baseline for each spectrum is indicated. The excitonic features are indicated by arrows.

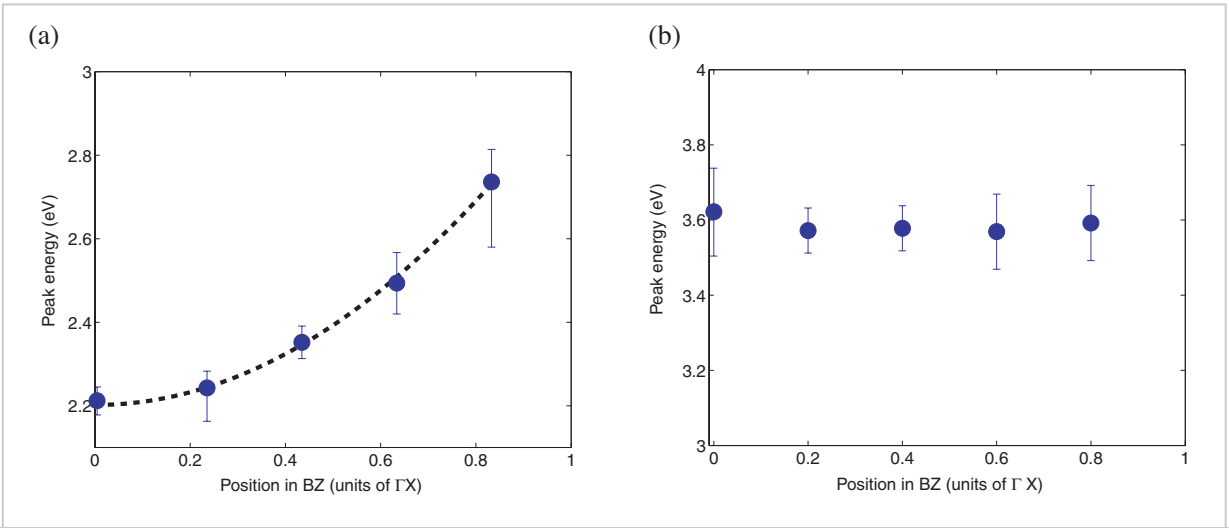

Fig. 2: Exciton dispersion in (a) La_2CuO_4 and (b) La_2NiO_4 as determined from the data in Figure 1.

(S=1/2) hole and the (S=1/2) O 2p hole. But, in contrast to the cuprate, the central Ni site with a $3d^8$ configuration is high spin (S=1). The direct consequence of this difference is the observed change in exciton dynamics: the single spin zero site in the S=1 NiO_2 AF lattice can only hop by disrupting the AF order and thus should have reduced mobility with respect to the twin spin zero Cu sites in the S=1/2 CuO_2 AF lattice.

Experimental Station

IXS Spectrometer

References

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