A study of lamellar copper oxides using the resonant inelastic X-ray scattering (RIXS) technique

J. N. Hancock, [1] L. Lu, [1] G. Chabot-Couture[1] O. Vajk[1], M. Greven[1], T. Gog[2], D. Casa[2], K. Ishii[3], J. Mizuki[3]
[1]*Department of Physics, Applied Physics and Stanford Synchrotron Radiation Laboratory, Stanford University*
[2]*Argonne National Laboratory, Argonne, IL U.S.A.*
[3] *JAEA (formerly JAERI), Spring8 Hyogo Prefecture, Japan*

Introduction

With the advent of 3rd generation synchrotron sources, inelastic X-ray scattering has emerged as a powerful probe of momentum and frequency-dependent charge and phonon dynamics. This development has allowed direct inferences regarding the two-particle response function in many contexts, including the Mott physics of correlated electron systems such as the lamellar copper oxides [1-5] and the manganites [6]. Resonant inelastic X-ray scattering (RIXS) provides a considerable advantage over ordinary inelastic scattering since, at resonance, the inelastic signal is significantly enhanced. In the lamellar copper oxides, this resonance condition can be met by tuning the incoming photon energy to the vicinity of the Cu *K* edge. The RIXS cross section sensitively depends on the incident photon energy [5] and on the nature of the intermediate states. Accordingly, the spectra simultaneously depend on both the incident and final photon energies and polarization [7].

In this work, we carefully investigate several systematic aspects of the RIXS cross section in order to identify and characterize the two-photon spectroscopic signature of the excitation spectra of the Mott insulator La_2CuO_4.

Methods and Materials

We have prepared a large single crystalline rod of the Mott insulator La_2CuO_4. To remove excess interstitial oxygen embedded during the growth, we annealed the sample at 950ºC for 24 hours. The crystal is of pristine quality for materials of this type, and the Neel transition, monitored through dc magnetometry, was found to be 320 K. A large piece of the crystal was then cut and then faceted by mechanical polishing to create ~1x1mm^2 facets along the 100, 001, 441, and 301 directions (real space) to remove all limitations of sample self-absorption on access to all of reciprocal space.

Here we use the tetragonal index convention for this crystal, which corresponds to the reciprocal lattice of an ideal CuO2 plane, with a~b=3.85, and c=13.1A.

RIXS is mathematically identical to Resonance Raman spectroscopy, which can be used to identify the symmetries of excitations using polarization analysis of the incident and scattered beam of usually near-IR, visible, or UV radiation. In this work, we use a similar, although less definite, polarization analysis by using the naturally polarized synchrotron source and carefully choosing distinct scattering geometries to isolate the symmetries of the different electronic excitations. To do this, we exploit two distinct and complementary scattering geometries, one with a vertical scattering plane, available at the Advanced Photon Source (CAT-9-ID-D), and second with a horizontal scattering plane, at Spring8 in Japan (JAEA-BL11XU). This represents the first real attempt to make a connection between the well-developed principles of Raman spectroscopy, and the burgeoning technique of RIXS.

Results and Discussion

Figure 1a shows a contour plot of line scans in a geometry (APS), that picks out the Raman-active symmetry channels A_{1g} $+E_g$, similar to previous published work[4,5,7]. These line scans are taken at a fine step size, ~500 meV, in order to aid in the identification of fine overlapping structures in the spectra. Using this method, in addition to the 2.2 eV and 4.3 eV features, we have identified a third feature nested between them with an energy of 3.0 eV.

While the origin of the 4.2 and 3.0 eV features is generally not understood, the 2.2 eV feature is commonly identified with the charge transfer gap, sometimes called the Mott gap, which is believed to reside at this energy scale in a host of transition metal oxide systems[3-7].

To investigate further the possible symmetries of these excitations, we have followed up on these measurements with subsequent measurements at Spring8, where a 90º condition and horizontal scattering geometry can be used to suppress the A_{1g} channel. Figure 1b shows the same contour plot as in 1a, but with a suppressed A_{1g} channel. We find that the 3 eV feature, clearly resolvable in the A_{1g} $+E_g$ channel, is strongly suppressed. This is the first demonstration of Raman principles and hard X-ray science to investigate the symmetry of charge excitations in a correlated electron system.

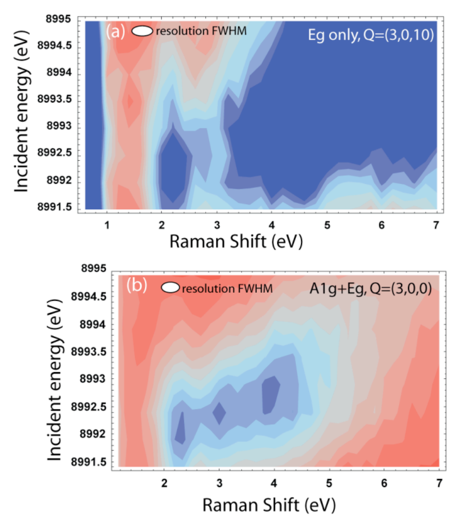

Fig. 1. Contour plot of the RIXS signal in 2 geometries. Scans are taken every 500meV in incident energy in both (a) and (b).

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