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# Two polarization dependent Auger decays observed by resonant photoelectron spectroscopy at the Cu2p-edge of superconducting BiSrCu-oxides

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## ABSTRACT

We report on a resonant photoemission (resPES) study of  $\text{Pb}_x\text{Bi}_{2-x}\text{Sr}_{2-y}\text{La}_y\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+4+\delta}$  (BISCO) single crystals to unravel the resonant decay mechanisms at the Cu2p absorption edge. The resPES studies on superconducting BISCO are performed at Bessy U49/2. We investigated BISCO single crystals with different hole doping concentrations at temperatures between 300 K and 17 K. CuO films have been used as a reference. We focus on the resPES data recorded at the Cu2p absorption edge. We find a strong polarization dependence for in-plane and out-of-plane geometries (with respect to the Cu-O plane) in our data which are caused by two independent Auger processes. We also report on the influence of different doping concentrations. Our resPES data enable us to sort the individual resonant mechanisms at the Cu2p edge in more detail than recently anticipated (Brookes et al., 2015). We discuss our data in the context of the recent discussion about the applicability of the Zhang-Rice model (Chen et al., 2013).

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## 1. Introduction

Early on the relevance of individual orbitals for the electronic structure of cuprates has been discussed [15,16]. It was argued that the basic features of the cuprates can be derived from a one band model incorporating exclusively hybridized  $\text{Cu}3d_{x^2-y^2}$  and  $\text{O}2p_{x,y}$  orbitals. This culminated in the view that one initial hole at the Cu combines with a doped hole residing at the ligand oxygen to form a Zhang-Rice singlet (ZRS) [15] in the  $\text{CuO}_2$  plane. Also the hole concentration in p-type cuprates has been known as a fundamental parameter in superconductivity as shown from the correlation between the superconducting transition temperature and the hole concentration. Both topics are adjunct and will be discussed in this publication. By performing resonant photoemission (resPES) at the  $\text{CuL}_3$  edge we will relativize X-ray absorption (XAS) as a reliable experimental tool to estimate the hole content of high- $T_c$  superconductors in a straightforward manner. In single crystals of the BISCO family we observed an out of-plane polarization dependence of resPES spectra by varying the angle between the electrical field vector  $E$  of the synchrotron light and the crystallographic  $a/b$  ( $\text{CuO}$ ) plane. This polarization dependence reveals the

existence of two different Auger processes – in plane and out of plane. The results lead to a modified scenario for the Zhang Rice construction in the  $\text{CuO}$  plane and to the necessity to incorporate hybridization with out of plane orbitals especially from Sr- and Bi-atoms.

## 2. Experimental

Single crystals of double-layer ( $n = 2$ ) optimum doped  $\text{Pb}_{0.4}\text{Bi}_{1.6}\text{Sr}_{2.0}\text{CaCu}_2\text{O}_8$  ((Pb,Bi)-2212-opt) ( $T_c = 92$  K,  $n_H \sim 0.16$ ) and of single-layer ( $n = 1$ ) slightly underdoped  $\text{Pb}_{0.6}\text{Bi}_{1.4}\text{Sr}_{1.5}\text{La}_{0.5}\text{CuO}_6$  ((Pb,Bi)-2201-ud) ( $T_c = 24$  K,  $n_H \sim 0.14$ ) and heavily underdoped, non superconducting  $\text{Pb}_{0.27}\text{Bi}_{1.6}\text{Sr}_{1.2}\text{La}_{0.8}\text{CuO}_6$  ((Pb,Bi)-2201-hud) ( $n_H \sim 0.06$ ) were investigated. They were grown out of the melt and well characterized by Laue diffraction and magnetic ac susceptibility. The elemental composition was checked by energy dispersive fluorescence. Also insulating, antiferromagnetic  $\text{CuO}$  films were taken as reference, obtained from oxidized Cu.

Photoemission spectroscopy (PES), resonant PES (resPES) and X-ray absorption measurements were performed at the storage ring BESSYII at beamline U49/2-PGM2 [4–6]. Samples were cleaved in UHV and transferred in-situ to the manipulator-cryostat with five degrees of freedom. Sample positioning was done to obtain

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in-plane and out-of-plane geometry with respect to the linearly polarized light.

Constant initial state (CIS) data are recorded at a fixed initial state energy. XAS data of the selected initial states were deduced by summing over all resPES data (partial Integrated Yield (pIY) spectra). The XAS-pIY data show the resonant contributions at the Cu2p edge and do not contain contributions of lower energy transitions or secondary electrons.

### 3. Results

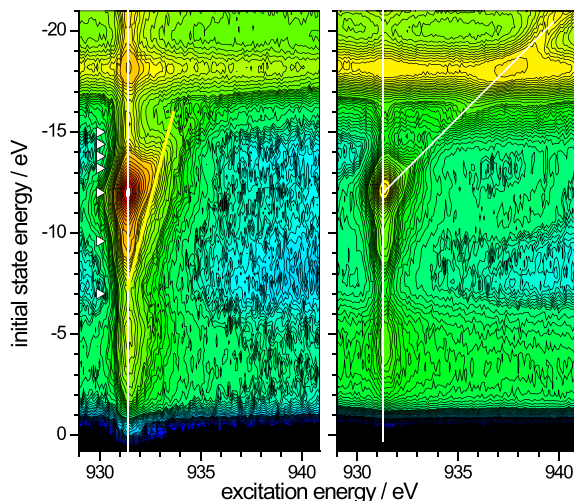
In Fig. 1 we show the resonant photoemission profiles at the Cu- $L_3$  edge recorded with in-plane (left) and out-of-plane (right) polarization. Here the resonance energy  $E_0$  (931.2 eV) is marked by the vertical white line in the two panels.

At the Cu2p absorption edge the most pronounced intensity is due to the Cu3d<sup>8</sup> satellite (initial state energy around -12 eV in Fig. 1). The intensity of the valence band states (around -4 eV initial state energy) with a 3d<sup>9</sup> configuration is much weaker. Very remarkable is the polarization dependence which is shown in Fig. 1 for a polarization geometry parallel to the Cu-O plane (in-plane, left panel) and perpendicular to it (out-of-plane, right panel).

In these data we identify two Auger processes. One is assigned to involve an in-plane ligand-to-metal CT state (yellow line, left panel). It originates right at the strong Cu3d<sup>8</sup> satellite and yields to a combined participator and spectator Auger decay with a four-hole (4h) final state [1]. The second Auger process is indicative of a Cu-LMM decay (white line, right panel). It appears only in out-of-plane geometry and involves Sr valence states as well.

In Fig. 2 the pIY for three BISCO samples with different doping levels (upper three panels) and the corresponding CIS curves for initial energies (states) in the valence band near the  $E_0$  peak at 12.5 eV (lower three panels) are depicted.

Fig. 2(middle panel) gives an example for the strong difference for the in plane and the out-of-plane polarization geometries. Here we use the integrated intensities (pIY) of the resPES profiles shown in Fig. 1. In the upper panels we show the corresponding data for the undoped ((Pb,Bi)-2201-hud) (Fig. 2(right panel)) and the optimum doped ((Pb,Bi)-2212-opt) (Fig. 2(left panel)) samples.



**Fig. 1.** Cu- $L_3$  edge resonant photoemission (resPES) profiles of ((Pb,Bi)-2201-ud) recorded at 17 K for in-plane (left) and out-of-plane (right) polarization. The thick yellow and the thin white lines mark the 4h-Auger and the Cu-LMM processes, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

We discuss these resPES data in the context of previous assignments of the doping dependence where the shoulder in the main Cu2p absorption signal is used to determine the hole doping concentration [2]. In Fig. 2(middle panel) we have also displayed the CIS spectra as taken from the in-plane data in Fig. 1. For these data we have used selected initial state energies around the Cu3d-sat emission (marked by the yellow line in Fig. 1). These detailed resPES data demonstrate the existence of an Auger emission which originates from  $E_0$ . In particular, there is no additional absorption signal separated from the main line at one particular photon energy, i.e. no “satellite”. The asymmetric broadening is caused instead by an Auger feature which emerges next to the main line and the individual curves shift depending on the energetic position as indicated by the small bars. The Auger contributions indeed cause a shoulder in the integrated pIY curves as shown in Fig. 2. The shoulder may be used for a qualitative assignment of the hole doping concentration, however, its quantification seems to be rather ambiguous. Insofar our results point to a problem by employing XAS data in TEY mode which are often used to analyze the doping behavior [2,7–11]. In these studies it was argued that the doping process would cause a chemical shift in the empty Cu3d states which would result in the formation of an extra peak showing up as the shoulder in the Cu2p XAS-TEY data. Here we demonstrate that this assumption definitely is wrong. Rather the broadening of the Cu2p XAS signal reflects a life time broadening of the KH intermediate state and this shows up as a combined Auger decay in our resPES data.

The CIS data of the three BISCO samples appear to be quite similar. The most important observation is that in none of these systems there is evidence for a second peak. All CIS data show a broadening caused by the Auger decays originating from the Cu3d8-sat emission. The observed broadening is strongest for the ((Pb,Bi)-2212-opt) sample, for the ((Pb,Bi)-2201-ud) it appears weaker, and for the undoped sample ((Pb,Bi)-2201-hud) it is still noticeable but very weak only.

In Fig. 3 we display the corresponding resPES data obtained for a CuO film. Here we notice that the resonance profile (top) is very narrow and the respective CIS data – again taken around the Cu3d8-sat emission – are almost symmetric. There is no evidence for an Auger process.

In total we have presented detailed resPES data for BISCO for different doping levels, and for CuO all recorded at the Cu2p resonance. We have investigated ((Pb,Bi)-2212-opt) and ((Pb,Bi)-2201-ud) samples which according to the EDX analysis and under the assumption of the universality of the superconducting dome have a doping concentration at or close to the optimum value of 0.16 holes per unit cell. These resPES data (Fig. 2(left and right panel)) appear to be rather similar and are indicative of an Auger process emerging out of the Cu3d8-sat emission. We compared these results to a BISCO sample which from the chemical analysis is undoped as well as to a CuO sample as a reference. These resPES data (Figs. 2(right panel) and 3) show a very sharp resonance profile, in contrast.

### 4. Discussion

Based on our resPES data we find that the resonant behavior at the Cu2p edge is dominated by two very different Auger processes.

#### 4.1. In-plane-Auger (IPA)

The IPA shows up only for in-plane polarization, it appears very close to the Cu3d-sat feature, and has a characteristic slope of 67°. This indicates a combined (P + S) decay as recently reported for organic and oxidic systems [1,12–14].

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