Electronic and Magnetic Properties of the Kagome Systems $YBaCo_4O_7$ and $YBaCo_3MO_7$ (M=Al, Fe)

N. Hollmann[1], Z. Hu[1], M. Valldor[1], A. Maignan[2], A. Tanaka[3], H.-H. Hsieh (謝輝煌)[4], H.-J. Lin (林宏基)[5], C.-T. Chen (陳建德)[5], and L.-H. Tjeng[1]

[1]II. Physikalisches Institut, Universität zu Köln, Köln, Germany
[2]Laboratoire CRISMAT, CNRS/ENSICaen, Caen, France
[3]Department of Quantum Matter, Hiroshima University, Higashi-Hiroshima, Japan
[4]Chung Cheng Institute of Technology, National Defense University, Taoyuan, Taiwan
[5]National Synchrotron Radiation Research Center, Hsinchu, Taiwan

Recently, a class of cobaltates was introduced with the compound $YBaCo_4O_7$, which contains kagomé layers of tetrahedrally coordinated Co.[1] The first susceptibility measurements yielded the large number of 5.8μ_B per magnetic ion,[1] but later experiments gave only 2.2 μ_B.[2] One neutron study estimated μ_t =3.49 μ_B for the ordered moment in the triangular layer and μ_k =2.19 μ_B in the kagomé lattice,[2] while another reported μ_t =1.66 μ_B and μ_k =1.68 μ_B, respectively.[3] It is perhaps *a priori* also not very clear what moments to expect theoretically since it is known, for example, that a Co^{3+} ion has the so-called spin state degree of freedom: it can be low spin (LS, S=0, nonmagnetic), intermediate spin (IS, S=1) or high spin (HS, S=2), depending on the details of the local crystal field.

Here, we present a study of the local electronic properties of $YBaCo_4O_7$, and its variants $YBaCo_3AlO_7$ and $YBaCo_3FeO_7$, using soft x-ray absorption spectroscopy (XAS) at the Co and Fe $L_{2,3}$ edges. We critically examine the charge state of the ions as well as their separate orbital and spin contributions to the magnetic moment.

Figure 1 shows the Co $L_{2,3}$ edge spectra with the Ba $M_{4,5}$ signal subtracted. It can be seen that the spectra of $YBaCo_3AlO_7$ and $YBaCo_3FeO_7$ are practically identical, and that they are different from that of $YBaCo_4O_7$.

We first focus on the Co spectrum of $YBaCo_3AlO_7$. From the chemical formula, for which Al is known to be very stable as a nonmagnetic trivalent ion, one is expecting the Co to be in the divalent state. The experiment spectrum can be excellently reproduced by cluster calculations as shown in Fig. 1. We have also carried out the simulation of $YBaCo_4O_7$ with a relation of 3:1 for Co^{2+}:Co^{3+}. We found that the experimental spectrum of $YBaCo_4O_7$ can be well reproduced for both HS Co^{2+} (dotted line) and HS Co^{3+} (dash line) in T_d symmetry as shown in the bottom in Fig. 1.

The striking similarity between the $YBaCo_3FeO_7$ spectrum with that of $YBaCo_3AlO_7$ suggests that all the Co ions are also divalent in $YBaCo_3FeO_7$. From the Fe-$L_{2,3}$ XAS we found a Fe^{3+} state in $YBaCo_3FeO_7$, which fulfills the charge balance requirement.

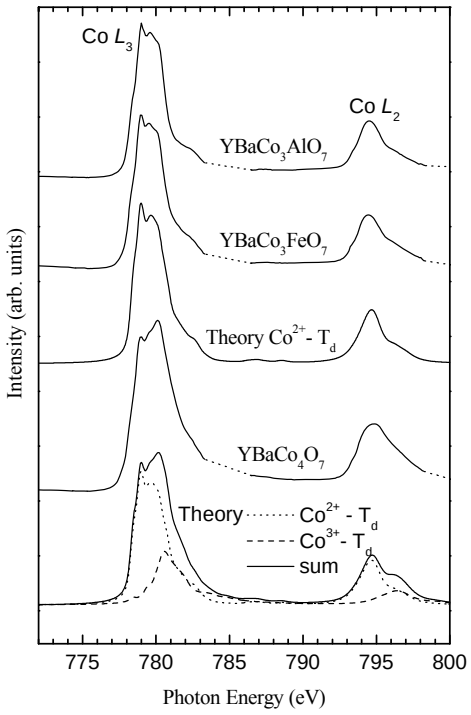

Fig. 1: Experimental Co $L_{2,3}$ XAS spectra of $YBaCo_3FeO_7$, $YBaCo_3AlO_7$, and $YBaCo_4O_7$ after subtraction of the Ba $M_{4,5}$ white lines and the theoretical calculation for Co^{2+} in tetrahedral symmetry (T_d). The bottom curve depicts the weighted sum of a calculation for Co^{2+} and Co^{3+} as a simulation for $YBaCo_4O_7$.

References

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