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Temperature and Pressure Induced Valence Transitions in YbCu$_{5-x}$Ga$_x$

Studied by Yb-L$_{III}$ XANES

R. Lübbers, J. Dumschat, G. Wortmann and E. Bauer* 

Fachbereich Physik, Universität-GH Paderborn, 33095 Paderborn, Germany 
* Institut für Experimentalphysik, Technische Universität Wien, 1040 Wien, Austria

Abstract: Employing the Yb-L$_{III}$ XANES, YbCu$_5$, YbCu$_4$Ga, YbCu$_5$Ga$_2$ and YbCu$_5$Ga$_{1.5}$ were studied in the temperature range 25 K to 300 K. YbCu$_5$ and YbCu$_4$Ga were studied at 300 K at pressures up to 175 kbar.

1. INTRODUCTION

The intermetallic series YbCu$_{5-x}$Ga$_x$, crystallizing in the hexagonal CaCu$_5$ structure, exhibit with increasing Ga content a transition of the Yb valence from (nearly) divalent to (nearly) trivalent. This valence transition is reflected by characteristic changes in the magnetic and other properties as measured already by a variety of methods [1]. We studied the temperature and pressure dependence of the Yb valence employing the Yb-L$_{III}$ edge. As reference compound for the near-edge structure the isomorphous GdCu$_5$ was used.

2. EXPERIMENTAL DETAILS

The preparation and characterization of the samples is described in Ref. [1]. The XAS measurements were performed at the ROEMO-II beamline (HASYLAB, Hamburg) employing a Si(3 11) double-crystal monochromator. The high-pressure studies were performed in a diamond-anvil cell with an axial transmission of the synchrotron radiation (SR) through the diamonds. The flux of SR was thereby reduced by a factor of about 10$^3$. The sample was placed in a 400 micron hole of a Ta$_{90}$W$_{10}$ gasket. The pressure was determined by the ruby fluorescence.

3. RESULTS AND DISCUSSION

Fig. 1 shows the Gd-L$_{III}$ near-edge XANES spectrum of GdCu$_5$. The white-line (WL) at the absorption edge exhibits a double structure, which could be adjusted by an arcus tangens and two Lorentzians with an intensity ratio of about 4 : 1 and separated by about 6 eV. This double structure can be explained either by a crystal-field splitting of the Gd-5d states or by multiple scattering effects, as indicated by preliminary calculations of the near-edge structure with the FEFF 6.01 program [4]. This double profile of the WL, characteristic for the hexagonal CaCu$_5$ structure, was used in the analysis of the Yb-L$_{III}$ edge spectra of the YbCu$_{5-x}$Ga$_x$ systems. Fig. 2 shows the spectra of YbCu$_5$, which could be fitted satisfactorily only with the WL profile derived from GdCu$_5$. It should be recalled that also the Yb-L$_{III}$ spectra of mixed-valent YbCu$_{5-x}$In could be only fitted with a double-peaked WL [5]. Already at ambient pressure a (temperature independent) trivalent component of about 25% intensity is observed. Such a behaviour is similar to Eu(II) systems near to a valence transition [2,3]. With increasing pressure, YbCu$_5$ exhibits a continuous transition towards trivalency reaching v(L$_{III}$) = 2.83 at 156 kbar.

Fig. 3 shows the corresponding spectra of YbCu$_4$Ga at various pressures and temperatures. As known from Ref. [1], this system exhibits a strong temperature and pressure dependence of the Yb valence. At ambient pressure, we derived from the spectra a variation of v(L$_{III}$) from 2.45 (25 K) to 2.57 (300 K), which agrees quite well with a simpler evaluation of the Yb-L$_{III}$ spectra. With increasing pressure, the valence reached with v(L$_{III}$) = 2.96 almost the trivalent state. It should be noted that the spectral shape is now very similar to that of trivalent GdCu$_5$.

With further increasing Ga content, we observed an increasing trivalent component connected with an almost linear temperature dependence of the Yb valence. We derived for YbCu$_{5-x}$Ga$_{1.5}$ v(L$_{III}$)-values from 2.61 (25 K) to 2.75 (300 K) and for YbCu$_5$Ga$_2$ from 2.71 (25 K) to 2.80 (300 K), which means the highest Yb valence at ambient pressure in the investigated series.

References:

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Figure 1: Gd-L\textsubscript{III} edge of GdCu\textsubscript{5} at T = 300 K.

Figure 2: Yb-L\textsubscript{III} edge of YbCu\textsubscript{5} at T = 300 K under atmospheric pressure (top) and at p = 156 kbar (bottom). The fit deconvolutions are marked with solid and dashed lines for Yb\textsuperscript{2+} and Yb\textsuperscript{3+}, respectively.

Figure 3: Yb-L\textsubscript{III} edge of YbCu\textsubscript{4}Ga at T = 25 K, T = 300 K and at p = 175 kbar. The fit deconvolutions are marked with solid and dashed lines for Yb\textsuperscript{2+} and Yb\textsuperscript{3+}, respectively.

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