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Temperature and Pressure Induced Valence Transitions in $\text{YbCu}_{5-x}\text{Ga}_x$ Studied by Yb-L_{III} XANES

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Abstract: Employing the Yb-L_{III} XANES, YbCu_5 , YbCu_4Ga , YbCu_3Ga_2 and $\text{YbCu}_{3.5}\text{Ga}_{1.5}$ were studied in the temperature range 25 K to 300 K. YbCu_5 and YbCu_4Ga were studied at 300 K at pressures up to 175 kbar.

1. INTRODUCTION

The intermetallic series $\text{YbCu}_{5-x}\text{Ga}_x$, 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 $\text{Si}(311)$ 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 $\text{Ta}_{90}\text{W}_{10}$ 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 $\text{YbCu}_{5-x}\text{Ga}_x$ 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_4In 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 $v(\text{L}_{\text{III}}) = 2.83$ at 156 kbar.

Fig. 3 shows the corresponding spectra of YbCu_4Ga 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 $v(\text{L}_{\text{III}})$ 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 $v(\text{L}_{\text{III}}) = 2.96$ 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 $\text{YbCu}_{3.5}\text{Ga}_{1.5}$ $v(\text{L}_{\text{III}})$ -values from 2.61 (25 K) to 2.75 (300 K) and for YbCu_3Ga_2 from 2.71 (25 K) to 2.80 (300 K), which means the highest Yb valence at ambient pressure in the investigated series.

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Figure 1: Gd-L_{III} edge of GdCu₅ at T = 300 K.

Figure 2: Yb-L_{III} edge of YbCu₅ 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 Yb²⁺ and Yb³⁺, respectively.

Figure 3: Yb-L_{III} edge of YbCu₄Ga at T = 25 K, T = 300 K and at p = 175 kbar. The fit deconvolutions are marked with solid and dashed lines for Yb²⁺ and Yb³⁺, respectively.

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