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To cite this version:
R. Lübbers, J. Dumschat, O. Wortmann, E. Bauer. Temperature and Pressure Induced Valence Transitions in YbCu5-xGax Studied by Yb-LIII XANES. Journal de Physique IV Colloque, 1997, 7 (C2), pp.C2-1021-C2-1022. 10.1051/jp4:19972125. jpa-00255184

HAL Id: jpa-00255184
https://hal.archives-ouvertes.fr/jpa-00255184
Submitted on 1 Jan 1997

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Temperature and Pressure Induced Valence Transitions in YbCu_{5-x}Ga_{x} Studied by Yb-L_{III} XANES

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Abstract: Employing the Yb-L_{III} XANES, YbCu_{5}, YbCu_{4}Ga, YbCu_{5}Ga_{2} and YbCu_{5.5}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(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 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_{4}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_{5.5}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:
Figure 1: Gd-L$_{III}$ edge of GdCu$_5$ at $T = 300$ K.

Figure 2: Yb-L$_{III}$ edge of YbCu$_5$ 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$^{2+}$ and Yb$^{3+}$, respectively.

Figure 3: Yb-L$_{III}$ edge of YbCu$_4$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$^{2+}$ and Yb$^{3+}$, respectively.

Acknowledgement: This work was supported by the BMBF (Projects 05-5PPACB and 05-643PPa-5).