Pressure Induced Valence Changes in YbAl₃ Studied by Resonant Inelastic X-ray Emission

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Introduction

Yb based intermetallic compounds exhibit interesting properties such as heavy fermion behavior, Kondo effect, superconductivity and electronic instabilities due to the intrinsic nature of the 4f electrons [1]. The number of 4f holes is directly related to the Yb valence and it is sensitive to external temperature and pressure applied on the system. In the Yb kondo systems temperature increases the population of the low lying excited states and a continuous valence change has been reported due to change in the 4f electronic configuration. Valence changes are also observed by applying pressure in several Yb compounds [2, 3]. Any technique which can probe the 4f population can be used for investigating such valence changes and high resolution x-ray emission spectroscopy (XES) is one among them. We report here the valence changes observed in YbAl₃ compound by XES and partial fluorescence yield absorption (PFY-XAS) experiments performed under pressures up to 38 GPA.

Methods and Materials

YbAl₃ has been prepared by arc melting of the appropriate metals in the stoichiometric ratio. Single phase poly crystalline powder resulted from the reaction was well ground and loaded in a 150 µm Be gasket pre-indented to 65 µm with few ruby chips in a panoramic type diamond anvil cell for XES measurements. For x-ray diffraction experiments the sample has been loaded with ruby grains into 135 micron hole of a Re gasket pre-indented to 60 microns in a Mao-Bell type diamond anvil cell with silicone fluid pressure medium. The XES experiments were performed at 16IDD at the Advanced Photon Source. The incident beam energy was tuned to the Yb L3 edge and focused to 130 x 50 µm at the sample by using K-B mirrors. The fluorescence signal was collected using a 1m Rowland circle spectrometer in which a spherically bent Si (333) single crystal was used to refocus the signal onto a Si detector. The xray diffraction experiments were performed at 16IDB in the angle dispersive geometry.

Results

The PFY – XAS spectrum collected for YbAl₃ at diffent pressures is shown in Fig.1 with the 2+ and 3+ components. The resonant XES spectra were collected up to 38 GPa with 2 eV steps for the incident energy.



Fig. 1. X-ray absorption intensity at various pressures for YbAl₃

Discussion

On analyzing the PFY-XAS, resonant XES spectra at ambient and high pressures, we have clearly noticed changes in the divalent and trivalent spectral distributions. The divalent contribution is found to be dominant at low pressures and it decreases continuously on application of pressure. It can be noticed that PFY-XAS spectra obtained at the highest pressure 38 GPa showed only the trivalent contribution. This indicates pressure induces a valence change in Yb from a mixed valence state to a trivalent state. Similar continuous valence change has also been observed in YbAl₃ by temperature increase as predicted by Andreson's impurity model [4]. Diffraction and magnetization measurements performed under pressure also support the valence changes.

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