High Field X-ray Magnetoabsorption Spectroscopy of Valence Transition in $YbInCu_4$

The correlation between itinerant electrons and localized electrons induces a variety of exotic properties in solids such as high-T_c superconductivity, the formation of the heavy fermion state and valence fluctuation phenomena. The materials that show these phenomena are so-called strongly correlated materials. Since the localized electrons (*d*- or *f*-electrons) can have large magnetic moments, the investigation of the interplay between the magnetic and electronic properties is crucial for the study of strongly correlated materials. The magnetic field is thus one of the most important external fields in experiments. It is particularly intriguing to study the magnetic-field-induced phase transitions found at low temperatures. At low temperatures, thermal excitation is suppressed and only the change in the ground state due to the quantum mechanical effects can be observed.

$YbInCu_4$ is a typical valence fluctuation material. A sharp first-order valence transition from $Yb^{\sim 3+}$ to $Yb^{\sim 2.85+}$ occurs at about 40 K with decreasing temperature. The valence fluctuates at low temperatures and the localized 4*f* magnetic moments are compensated by the conduction electrons. The ground state is thus non-magnetic below ~ 40 K. As mentioned above, the magnetic-field-induced valence transition at a low temperature is also very interesting. The field-induced valence transition in $YbInCu_4$ was observed by magnetization and magnetostriction measurements [1]. The metamagnetism and the anomaly in the striction were observed and the transition field was found to be around 30 T (Tesla) at 4.2 K.

Synchrotron X-ray spectroscopy plays an important role for the study of the valence fluctuation. The X-ray absorption due to the inner-shell transition of rare-earth ions with different valence states occurs at different energies because the Coulomb repulsive energy between the 4*f* electrons and the core holes (created by the incident X-rays) considerably depends on the number of 4*f* electrons in the shell. The valence transition in $YbInCu_4$ induced by changing the temperature has been investigated intensively by several X-ray spectroscopic techniques. Although it would be interesting to perform an X-ray absorption experiment on $YbInCu_4$ in high magnetic fields exceeding 30 T, such an experiment has not previously been performed because of technical difficulties.

In this article, we present our recent results of a pulsed-high-magnetic-field X-ray absorption experiment on the valence transition in $YbInCu_4$. The portable pulsed field generator allows us to measure X-ray absorption spectra in magnetic fields up to 41 T at 5 K [2]. The significant field dependence of the Yb valence was directly observed for the first time.

The experiment was carried out at beamline **BL22XU**. A single crystal of $YbInCu_4$ was powdered and brushed onto a piece of Scotch tape for the transmission measurement. A very small pulsed magnet (*D*, *L*, *d* = 30 mm, 30 mm, 3 mm, where *D* is the outer diameter, *L* is the length and *d* is the inner diameter) was used to generate a high magnetic field. Since this small magnet can be readily installed into conventional apparatus such as a cryostat or a diffractometer, various experiments can be performed easily in high magnetic fields over 30 T [2,3]. In this study the magnet is installed into a He-flow cryostat and is cooled to the lowest temperature together with the sample. Figure 1 shows a photograph of the small magnet attached on the bottom of the inner rod of the cryostat.

Figure 2 shows the X-ray absorption spectra near the L_{III} edge of Yb at 0 and 41 T. The absorption band at ~ 8.94 keV is due to the transition of Yb^{2+} and that at ~ 8.95 keV is due to Yb^{3+} [4]. Although the Yb^{2+} absorption band is clearly observed at 0 T, it is weaker at 41 T. The Yb^{3+} band is correspondingly stronger at

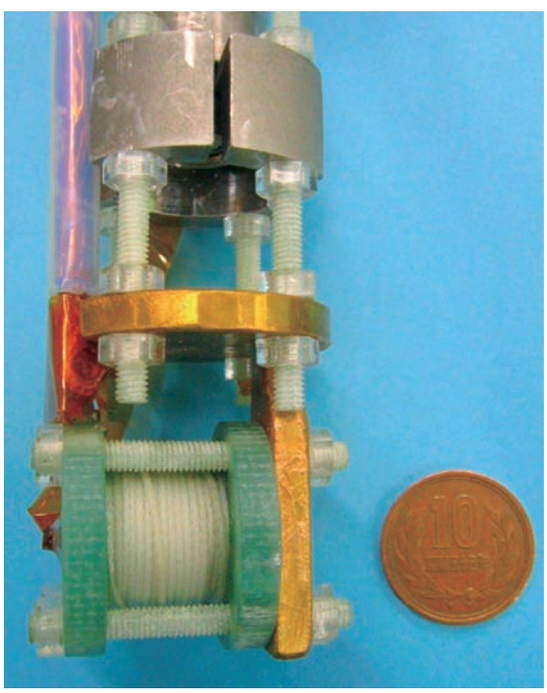

Fig. 1. Photograph of the solenoid-type miniature magnet used for the X-ray absorption measurement.

41 T. Since it is generally accepted that the absorption intensity corresponds to the probability of the existence of each valence state, the valence v^* can be deduced by $v^* = 2I_2/(I_2+I_3)+3I_3/(I_2+I_3)$, where I_2 and I_3 are the absorption intensities of Yb^{2+} and Yb^{3+}, respectively. The solid, dashed and dotted curves in Fig. 2 are the results of the curve fitting using the Lorentzian and the arctangent. It was found that the fitted solid curve accurately reproduces the experimental result. From the fitting, I_2 and I_3 are deduced and the valence v^* can be obtained.

The obtained valences are plotted as a function of the magnetic field in Fig. 3. The temperature variation at 0 T deduced from the X-ray absorption spectra is also shown in the inset. It was found that the valence significantly increases beyond 30 T and becomes saturated at approximately 40 T. Note that the saturated valence ($v^* = 2.96\pm0.02$) is nearly the same value as that at high temperatures ($v^* = 2.97\pm0.02$), as shown in the inset. This fact suggests that the compensated 4*f* moments recover to be localized completely at 40 T. Since the thermal excitation can be disregarded at 5 K, the small deviation of the valence from 3.0 may be due to the effect of final state (the effect of the core-hole). However, further studies are required to allow further discussion.

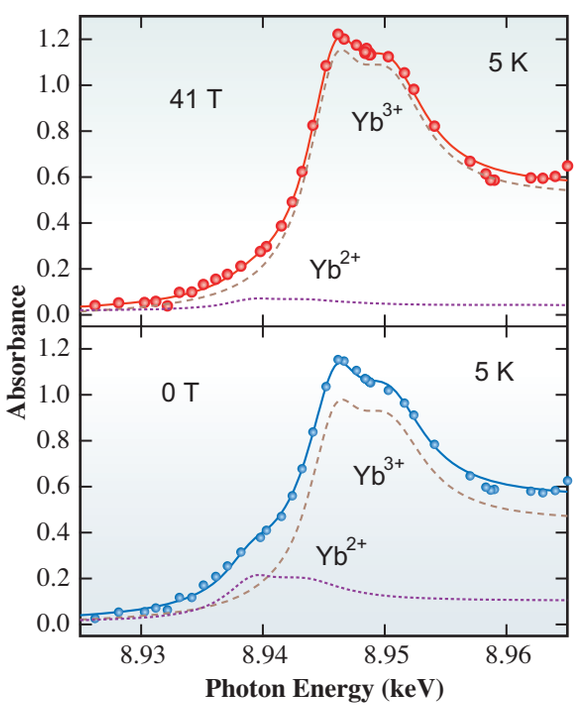

Fig. 2. X-ray absorption spectra at 0 and 41 T. The solid, dashed and dotted curves show the results of the curve fitting.

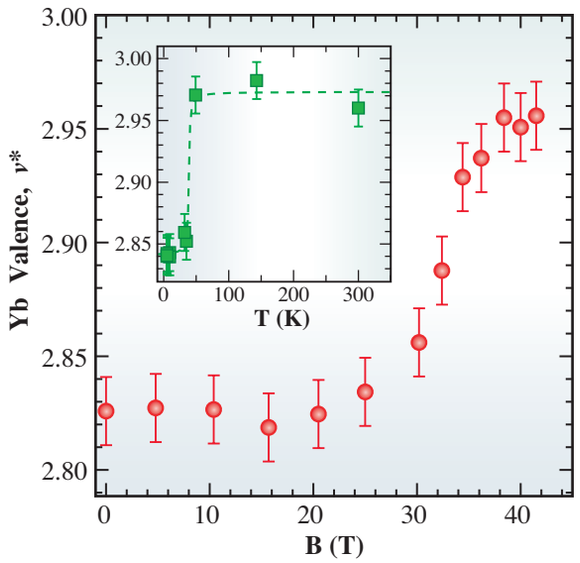

Fig. 3. Magnetic field dependence of the valence at 5 K. The inset shows the temperature dependence of the valence at zero magnetic field.

In the present work, high-magnetic-field X-ray absorption spectroscopy has been established as a unique tool for examining the valence state in very high magnetic fields. This technique can also be applied to various types of magnetic-field-induced phase transitions found in strongly correlated materials.

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