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X-Ray diffraction study of $YbInCu_4$ at high pressure

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Abstract

The lattice constants of cubic $YbInCu_4$ have been measured at high pressure up to 13 GPa at room temperature in order to examine the phase transition and the valence state of Yb at high pressure. It is found that the cubic structure is stable up to 13 GPa and the bulk modulus B_0 and its derivative B_0' are 112 GPa and 4.0, respectively. By comparing the present result with that of the concentrated Kondo compound $YbAgCu_4$, the trivalent valence state of Yb in $YbInCu_4$ is suggested to be stable up to 13 GPa at room temperature.

1. Introduction

It has been well established that the physical properties of intermetallic compounds having unstable f-electrons are very sensitive to external forces such as temperature, pressure and magnetic field [1]. Ce compounds, particularly the so-called concentrated Kondo (CK) compounds, show a wide variety of electrical and magnetic properties by application of pressure [2]. It has been reported that the CK state in the Ce compounds tends to disappear at high pressure and then enter a new electronic state, the intermediate valence (IV) state [3]. The crossover in the electronic state is due to the strong hybridization between 4f-electron and conduction band states.

On the other hand, Yb compounds, which may be a counterpart of Ce compounds, are also known to show a lot of anomalous properties at high pressure [4–6]. The response of the physical properties of Yb compounds to applied pressure is in sharp contrast to that of Ce compounds [4]. For example, the temperature of the resistance maximum T_{max} which roughly corresponds to the Kondo temperature T_K, increases with pressure for almost all Ce compounds [7] but decreases for Yb compounds [4]. In some Yb compounds, the CK state is induced by applying pressure [4].

The intermetallic compound $YbInCu_4$ crystallizes in the $AuBe_5$ type (C15b) structure with the lattice constant, $a = 7.156$ Å [8,9]. The most interesting feature of $YbInCu_4$ is that it shows a valence transition from high temperature stable trivalent Yb^{3+} to low temperature mixed valence Yb around 50 K ($= T_v$). T_v is found to be strongly dependent on pressure with $\partial T_v/\partial P \approx -2$ K/kbar [5,6]. Nowik *et al.* [5] reported an anomaly near 6 kbar in the pressure dependence of T_v. However, no anomaly in the T_v–P curve was observed by Kojima *et al.* [6] and Matsumoto *et al.* [10].

In the present work, we made an attempt to measure the pressure dependence of the lattice constant a (Å) of $YbInCu_4$ in order to examine the effect of the stability of the electronic state on the volume of this compound at high pressure. The result obtained here is compared with that of the heavy fermion compound $CeInCu_2$ showing pressure-induced crossover.

2. Experimental procedure

The details of sample preparation were reported previously [6]. The ingot was crushed into fine powder in alcohol to avoid oxidation. Hydrostatic pressure was generated by using a WC Bridgman-type anvil having a face 3 mm in diameter [11]. The powdered sample and NaCl were placed in a 0.3-mm hole at the center of a beryllium disk 0.5 mm thick, which was used as a gasket. A 4:1 methanol/ethanol mixture was used as a pressure transmitting medium, in which the pressure is expected to be hydrostatic up to 10 GPa. The pressure was determined by using Decker's equation of state of NaCl [12]. A Guinier type focusing camera was used to obtain high accuracy in the determination of the lattice constants. The diffraction lines were recorded on highly sensitive film.

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TABLE 1. Summary of the present results and the data of $YbAgCu_4$ [13] and $CeInCu_2$ [3, 11]

	a_0 (Å)	γ (mJ/mol K^2)	B_0 (GPa)	B_0'	T_{max} (K)	$\partial T_{max}/\partial P$ (K/kbar)
$YbInCu_4$	7.16	22	112	4.0	–	–
$YbAgCu_4$	7.07	245	108	3.3	100	−1.4
$CeInCu_2$	6.78	1200	90	3.9	27	4

3. Results and discussion

Figure 1 shows the relative change in the lattice constant a/a_0 as a function of pressure at room temperature, where a and a_0 are the lattice constants at high and ambient pressure, respectively. Since there are no new diffraction lines up to 13 GPa, the $AuBe_5$ structure is stable at room temperature at least to 13 GPa. A discontinuous change in the value of a/a_0, similar to the γ–α transition in Ce metal, is not observed in the present pressure range within an experimental error. This result indicates that a discontinuous valence transition as observed at T_v is not induced by pressure up to 13 GPa at room temperature. Further, there is no anomaly in the lattice compression around 1 GPa.

In order to estimate the bulk modulus, we attempted a least-squares fit of the data in Fig. 1 to the following first-order Murnaghan's equation of state:

$$P = \frac{B_0}{B_0'}\left[\left(\frac{V}{V_0}\right)^{-B_0'} - 1\right] \quad (1)$$

where V ($\approx a^3$) and V_0 ($=a_0^3$) are the volume at high and ambient pressure, respectively. The result of the least-square fit is shown by the solid curve in Fig. 1. The values of B_0 and B_0' were found to be $B_0 = (112 \pm 2)$ GPa and $B_0' = 4.0 \pm 0.2$, respectively. B_0 of

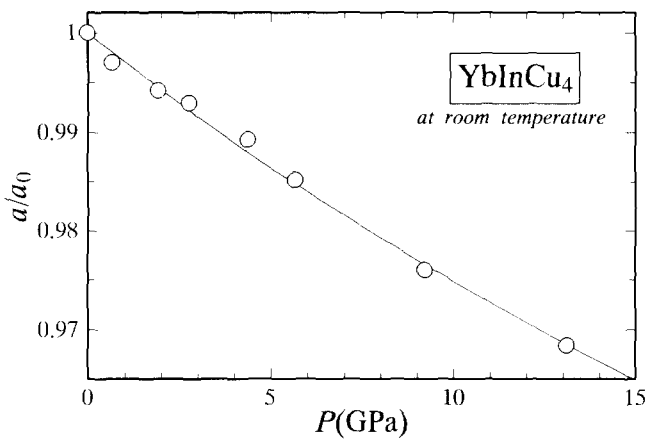

Fig. 1. Pressure dependence of the relative change in volume V/V_0 of $YbInCu_4$ at room temperature, where V and V_0 are the volume at high and ambient pressure, respectively.

$YbInCu_4$ is slightly larger than that of the CK compound $YbAgCu_4$, 108 GPa [13].

The results obtained in the present work are summarized in Table 1 together with those of the related CK or heavy fermion compounds, $YbAgCu_4$ and $CeInCu_2$ for comparison [3,13]. There seems to be a relationship between B_0, the electronic specific heat coefficient γ and the lattice constant a_0. To clarify this, however, we need to accumulate more data about Yb or Ce compounds.

It has been pointed out that the electronic state of $YbInCu_4$ changes from the intermediate valence (IV) state to the CK state by substituting In by Ag [14,15]. This is considered to be due to a decrease in the hybridization between the localized f-electron and the conduction band caused by an addition of Ag. Since the application of pressure essentially results in an increase in the hybridization, the effect of Ag addition is opposite to that of pressure. Taking account of these facts and the pressure dependence of T_v, $YbInCu_4$ might correspond to a high pressure phase or a kind of compressed state of $YbAgCu_4$ to have a larger bulk modulus than $YbAgCu_4$. This consideration explains qualitatively the present results for the bulk modulus. Furthermore, from the theoretical calculation by Lavagna *et al.* [16], it was revealed that the bulk modulus in the Kondo state is always lower than that in the non-Kondo or normal state. If we consider that $YbAgCu_4$ is in the Kondo state and $YbInCu_4$ in the normal trivalent state, the bulk modulus of $YbInCu_4$ would be larger than that of $YbAgCu_4$. This is also consistent with the present results.

Finally, we comment on the value of $B_0' = (\partial B/\partial P)_{P=0} - 1$. In Table 1, B_0' of $YbInCu_4$ is found to be larger than that of $YbAgCu_4$. The small value of B_0' indicates a relatively slow stiffening of the lattice with increasing pressure. For the IV compound $YbAl_2$ [17], the value of B_0' was reported to be 5.0 ($B_0 = 43.1$ GPa), which is about half of that of the stable trivalent compound $PrAl_2$ ($B_0' = 7.7$ and $B_0 = 74.8$ GPa). This fact indicates that the electronic state of the compounds having unstable f-electrons has a strong effect not only on the value of B_0 but also on B_0'. The small value of B_0' of $YbAgCu_4$ suggests a large change in the electronic state due to increasing hybridization by pressure. On

the other hand, for $YbInCu_4$, the effect of the electronic state on the lattice stiffening may be relatively small, which indicates that the Yb^{3+} state is stable up to 13 GPa. This result is consistent with that of the electrical resistivity in which the $\rho(T)$ curve of $YbInCu_4$ shows normal properties above T_v [8].

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