Pressure effects on magnetic susceptibility of intermediate-valence compound YbAl₃

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Abstract. Recently, there have been considerable interests in a slow crossover from Fermi liquid behaviour to local moment behaviour for YbAl₃. We have investigated the pressure effects on the magnetic susceptibility of single-crystalline YbAl₃ as a function of temperature (2-300 K) and hydrostatic pressure (up to 1 GPa). At ambient pressure the susceptibility shows a broad maximum of χmax at Tmax ~ 116 K, consistent with a high Kondo temperature TK ~ 500 K. Below coherent temperature Tcoh ~ 40 K, the susceptibility shows anomalous enhancement with a small maximum at TmL ~ 17 K, and reaches constant value of χ₀ below 5 K. The application of pressure causes an increase in the susceptibility over the entire temperature range. Both the characteristic temperatures Tmax and TmL decrease with increasing pressure. We find that above Tcoh the temperature and pressure dependence of χ/χmax obey a universal curve scaled with reduced temperature T/Tmax. On the other hand, below Tcoh, χ/χmax deviates from the universal curve and is increased by applied pressure. The Grüneisen parameter for TK and Tcoh are –2.2 and –4.6~12, respectively. These results indicate that Tcoh/TK is decreased by applied pressure in YbAl₃.

1. Introduction
YbAl₃ is an intermediate-valence compound, which has attracted much attention for the last few decades [1-4]. YbAl₃ crystallizes in the simple cubic AuCu₃-type structure (a = 0.4213 nm, space group Pm3m) [5]. The magnetic susceptibility follows the Curie-Weiss law above 250 K with an effective moment 4.2 μ₀ and shows a well-known bell-shaped curve with a maximum of χmax at Tmax ~ 120 K, consistent with a high Kondo temperature TK ~ 500 K [3]. The Yb valence is 2.8 at 200 K and decreases to 2.65 as the temperature is reduced [6]. Below 40 K, the Fermi liquid behavior of T²-dependence of the electrical resistivity is observed [7, 8]. The susceptibility increases below 40 K to a peak at ~15 K [8-11]. The specific heat coefficient also displays an upturn below 30 K and saturates at ~45 mJ/molK² at zero temperature [1, 8, 10]. These features mean an enhancement of the effective mass in YbAl₃ associated with a coherent temperature of low-temperature energy scale Tcoh ~ 40 K. Thus intermediate-valence compound YbAl₃ is characterized by a high Kondo temperature TK ~ 500 K and a coherent temperature Tcoh ~ 40 K.

Recently, the interest in YbAl₃ has been stimulated by two energy scales and a slow crossover behavior from the Fermi liquid state to the local moment state [8, 10-12]. The crossover behavior for YbAl₃ has been studied by comparison between the prediction of the Anderson impurity model and the experimental results of susceptibility, specific heat, Hall coefficient and x-ray absorption at ambient...
pressure [8, 10, 11]. However, to extract information about the energy scales associated with the Kondo coupling, high-pressure studies are powerful techniques [13]. Since the Kondo coupling strength will be varied by pressure, we can examine the validity of the single energy scaling law to find the pressure-dependent scaling temperature $T_0(P)$ in YbAl$_3$.

In this paper, we investigated the pressure dependences of the magnetic susceptibility of high-quality YbAl$_3$ single crystals. Above $T_{coh}$ the temperature and pressure dependences of $\chi/\chi_{max}$ obey a universal curve scaled with reduced temperature $T/T_{max}$ within the framework of the Anderson impurity model, whereas below $T_{coh}$, $\chi/\chi_{max}$ deviates from the universal curve, being increased by applied pressure. We also obtained the Grüneisen parameters of $T_K$ and $T_{coh}$. We found that $T_{coh}/T_K$ is decreased by applied pressure in YbAl$_3$.

2. Experimental

Single crystals of YbAl$_3$ and LuAl$_3$ were grown by an Al flux-method; the details were reported elsewhere [7]. The phase purity for the resultant products was proved from an x-ray powder diffraction (CuK$_\alpha$) measurement. The magnetic susceptibility were measured by a SQUID magnetometer (Quantum Design) from 2 to 300 K in a magnetic field $H = 5$ kOe. The hydrostatic pressure up to 1 GPa was generated by a non-magnetic piston (zirconia) and cylinder (BeCu) clamp-type cell [14]. Daphne 7373 oil (Daphne, Idemistu) was used as a pressure-transmitting medium [15]. The pressure was determined from values of the superconducting transition temperature for tin [16].

3. Results and discussion

Figure 1 shows the magnetic susceptibility $\chi$ measured at ambient pressure for YbAl$_3$ and reference material of LuAl$_3$ single crystals. A small low-temperature Curie-tail indicates that these samples are of high-purity. For YbAl$_3$ two main characteristic features were clearly observed; a broad maximum of $\chi_{max}$ centered at $T_{max} \sim 116$ K and another small maximum at $T_{ml} \sim 17$ K. Below 5 K, the susceptibility reached a constant value of $\chi_0$. These features are in good agreement with the previous reports [8-11]. Reference compound LuAl$_3$ was a Pauli-paramagnet. Since the magnitude of susceptibility for LuAl$_3$ was less than 3% of that for YbAl$_3$, the temperature dependence of susceptibility of YbAl$_3$ are governed mainly by the 4$f$-contribution from Yb ions. We also confirmed that the pressure effects on the susceptibility of LuAl$_3$ were negligibly small.

![Figure 1. Temperature dependence of magnetic susceptibility at ambient pressure for YbAl$_3$ and LuAl$_3$.](image1)

![Figure 2. Temperature dependence of magnetic susceptibility for YbAl$_3$ at ambient pressure and 1.07 GPa.](image2)
The temperature dependences of the susceptibility for YbAl₃ with and without pressure are shown in figure 2. In this figure, both the Curie-tail and background signal of the pressure cell are subtracted from the raw data. With increasing pressure, the susceptibility increased in all temperature regions. Both $T_{\text{max}}$ and $T_{\text{ml}}$ decreased with increasing pressure. The fact that $T_{\text{max}}$ is proportional to Kondo temperature $T_K$ indicates that $T_K$ decreases with increasing pressure.

If the susceptibility of $\chi$ obeys a scaling assumption $\chi(T,P) = \chi(T/T_0(P))(T/T_0(P))$, where $f$ is a universal function of $T/T_0$, then the effects of pressure is interpreted to decrease (or increase) characteristic temperature $T_0$ uniformly in all temperature regions. To obtain some insight into the energy scaling law for YbAl₃, in figure 3, we plot $\chi T/C_f$ versus $T/T_{1/2}$ for various pressures, where $C_f$ is the $J=7/2$ Curie constant for Yb³⁺-free ions and $T_{1/2}$ is defined as the temperature where $\chi T/C_f = 0.5$. $\chi T/C_f$ can be looked on as the square of the relative magnetic moment $\mu(T)/\mu(\infty)^2$, where $\mu(\infty)$ is Yb³⁺ moment at high temperature. $T_{1/2}$ is also proportional to $T_K$ [17]. At ambient pressure, $T_{1/2}$ was 300 K and decreases with increasing pressure for YbAl₃. The pressure-dependent susceptibility of YbAl₃ is scaled onto a universal curve, as clearly seen in figure 3. However since the $\chi T/C_f$ plot suppresses the low temperature anomaly, another scaling plot is required to see the features in the low temperature region.

Thus, in figure 4 we plotted $\chi/\chi_{\text{max}}$ versus $T/T_{\text{max}}$ in various pressures, where $\chi_{\text{max}}$ is a maximum value of susceptibility at $T_{\text{max}}$. Above Kondo coherent temperature $T_{\text{coh}}$, the pressure dependence of susceptibility obeys a universal curve, as expected from the results in figure 3. Below $T_{\text{coh}}$, the deviation from the universal curve occurs at high pressures. This indicates that the susceptibility below $T_{\text{coh}}$ is not scaled by $T_K$. Therefore, it can be concluded that the overall temperature dependence of susceptibility of YbAl₃ cannot be explained by a single energy scale.

Using an experimental bulk modulus of 65.2 GPa [18], we calculate the Grüneisen parameters $\Gamma(T_i) = -\partial \ln T_i/\partial \ln V = BT_i^{-1} \partial T_i/\partial P$, where $T_i$ is a characteristic temperature in the susceptibility, $V$ the sample volume and $B$ the bulk modulus. The $\Gamma(\chi_{\text{max}}^{-1})$ is also calculated since $\chi_{\text{max}}^{-1}$ will be proportional to $T_{\text{coh}}$ [13]. The obtained Grüneisen parameters of pressure-dependent susceptibility for YbAl₃ are listed in table 1. Our previous result $\Gamma(A^{1/2})$ is also listed in Table 1, where $A$ is a coefficient of the $T^2$ term in the temperature dependence of electrical resistivity below $T_{\text{coh}}$ [7]. The Grüneisen parameters for $T_K$ $\Gamma(T_K)$ are $-2.2$, determined from the pressure dependence of $T_{\text{max}}$ and $T_{1/2}$. This small Grüneisen
parameter of $T_K$ reflects the high Kondo temperature in YbAl$_3$, since the magnitude of $\Gamma(T_K)$ is roughly inversely proportional to $T_K$ [13]. We determined the Grüneisen parameters for $T_{coh}$ $\Gamma(T_{coh})$ from the pressure dependence of $T_{int}$, $\chi_0^{-1}$ and $A^{1/2}$ observed below $T_{coh}$. $\Gamma(T_{coh})$ are two to five times larger than $\Gamma(T_K)$, which indicates that the ratio of $T_{coh}/T_K$ is decreased by pressure in YbAl$_3$. This suggests immediately that the two energy scales $T_{coh}$ and $T_K$ are not uniquely related mutually in YbAl$_3$.

| $\Gamma(T_K)$ (GPa$^{-1}$) | $\Gamma(T_{coh})$ (GPa$^{-1}$) |
|--------------------------|-----------------------------|
| $\Gamma(T_{max})$        | $\Gamma(T_{1/2})$           |
| $-2.2$                   | $-2.2$                      |
| $\Gamma(T_{int})$       | $\Gamma(\chi_0^{-1})$      |
| $-12$                    | $-8$                        |
| $\Gamma(A^{1/2})$       |                             |
| $-4.6$                   |                             |

4. Summary
The magnetic susceptibility of the intermediate-valence compound YbAl$_3$ has been measured over a wide temperature range under hydrostatic pressure. We found that the temperature and pressure dependences of magnetic susceptibility for YbAl$_3$ do not obey the single energy scaling law below $T_{coh}$ while $T_{coh}/T_K$ is decreased by applied pressure in YbAl$_3$.

Acknowledgements
The authors are grateful for the financial support of the Nitto Foundation.

References
[1] Havinga E E, Buschow K H J and van Daal H J 1973 Solid State Commun. 13 621
[2] Buschow K H, Goebel U and Dormann E 1979 phys. stat. sol. (b) 93 607
[3] Klaasse J C P, de Boer F R and de Chatel P F 1981 Physica 106B 178
[4] van Daal H J, van Aken P B and Bouchow K H J 1974 Physics letters 49A(3) 246
[5] Palenzena A 1972 Journal of the Less Common Metals 29(3) 289
[6] Suga S, Sekiyama A, Imada S, Shigemoto A, Yamasaki M, Dallera C, Braicovich L, Lee T L, Sakai O, Ebihara T and Ōnuki Y 2005 J. Phys. Soc. Jpn. 74 2880
[7] Ohara S, Chen G F, Sakamoto I 2001 J. Alloys and Compounds 323-324 632
[8] Cornelius A L, Lawrence J M, Ebihara T, Riseborough P S, Booth C H, Hundley M F, Pagliuso P G, Sarrao J L, Thompson J D, Jung M H, Lacerda A H and Kwei G H 2002 Phys. Rev. Lett. 88 117201
[9] Hiess A, Boucherle J X, Givord F, Schweizer J, Lelièvre-Berna E, Tasset F, Gillon B and Canfield P C; 2000 J. Phys.: Condens. Matter 12 829
[10] Ebihara T, Bauer E D, Cornelius A L, Lawrence J M, Harrison N, Thompson J D, Sarrao J L, Hundley M F and Uji S 2003 Phys. Rev. Lett. 90 166404
[11] Bauer E D, Booth C H, Lawrence J M, Hundley M F, Sarrao J L, Thompson J D, Riseborough P S and Ebihara T 2004 Phys. Rev. B 69 125102
[12] Burdin S and Zlatić 2009 Phys. Rev. B 79 115139
[13] Thompson J D and Lawrence J M 1994 Handbook on the physics and Chemistry of Rare Earths vol. 19 ed by K. A. Gshneidner Jr., L. Eyring, G. H. Lander ans G. R. Choppin (Elsevier)
[14] Uwatoko Y, Hotta T, Matsuoka E, Mori H, Ohki T, Sarrao J L, Thompson J D, Mori N, Oomi G 1998 Rev. High Pressure Sci. Technol. 7 1508
[15] Murata K, Yoshino H, Yadav H O, Honda Y, Shirakawa N 1997 Rev. Sci. Instrum. 68 249
[16] Smith T F and Chu C W 1967 Phys. Rev. 159 353
[17] Yoshimori A and Kasai H 1983 J. Magn. Magn. Mater. 31-34 475
[18] Kumar R S, Svane A, Vaitheeswaran G, Kanchana V, Bauer E D, Hu M, Nicol M F and Cornelius A L 2008 Phys. Rev. B 78 075117