Transport and magnetic properties of new heavy-fermion antiferromagnet YbNi$_3$Al$_9$

S Ohara, T Yamashita, Y Mori and I Sakamoto

Department of Engineering Physics, Electronics and Mechanics, Graduate School of Engineering, Nagoya Institute of Technology, Nagoya 466-8555, Japan
E-mail: ohara.shigeo@nitech.ac.jp

Abstract. We have synthesized a new Yb-based Kondo-lattice compound YbNi$_3$Al$_9$. This compound crystallizes in a trigonal ErNi$_3$Al$_9$-type structure (space group R32), in which the Yb-ion is arranged in a two-dimensional honey-comb lattice perpendicular to the c-axis. We report the first measurements of electrical resistivity and magnetization for single-crystalline samples of YbNi$_3$Al$_9$. The electrical resistivity of YbNi$_3$Al$_9$ is characteristic of the typical properties of heavy-fermion antiferromagnets with a Neel temperature of $T_N = 3.4\text{K}$. The transport and magnetic properties exhibit large anisotropy in the low-temperature region owing to an interplay among the crystalline-electric-field effect, the Ruderman-Kittel-Kasuya-Yoshida interaction, and the Kondo effect. Below $T_N$, the metamagnetic transition is observed at a very low magnetic field of around 1 kOe with the field applied along the $a$-axis. The magnetic structure of YbNi$_3$Al$_9$ is highly sensitive to the applied magnetic field.

1. Introduction

Many Ce- and Yb-based intermetallic compounds have attracted considerable interest owing to their various physical properties such as unusual superconductivity and heavy-fermion behavior and the phenomena they exhibit, such as valence fluctuations and anomalous magnetic transitions[1]. These properties and phenomena arise from the interplay among the crystalline-electric-field (CEF) effect, the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction, and the Kondo effect. Numerous studies have been conducted on heavy-fermion Ce-based compounds. However, relatively few studies have been conducted on heavy-fermion Yb-based compounds. This is mainly due to difficulties in the sample preparation of Yb-based compounds, which is due to the high vapor pressure of Yb[2] and also because most of the known Yb-based intermetallic compounds are found to be usual mixed-valent or non-magnetic metals.

Recently, we have succeeded in synthesizing single crystals of YbNi$_3$Al$_9$ through an investigation of the Al-rich parts of the phase diagrams of Yb-T-Al systems, where T is a transition metal. The crystal structures of ternary aluminides RNi$_3$Al$_9$ ($R = \text{Y, Gd, Dy, Er}$) were reported by Gladyshevskii[3]. However, to the best of our knowledge, the physical properties of YbNi$_3$Al$_9$ have not yet been investigated in detail.

We have found that YbNi$_3$Al$_9$ crystallizes in the trigonal ErNi$_3$Al$_9$-type structure (space group R32)[3]. In this crystal structure, three types of layers are observed to pile up along the c-axis: Al$_3$-layers with a triangular mesh, Ni$_3$-layers with a triangular mesh, and Yb$_2$Al$_3$-layers. In the Yb$_2$Al$_3$-layer, the Yb ion is arranged in a two-dimensional honey-comb lattice site and at the center of the Al$_3$ triangle. The Yb ion occupies a site equivalent to 6c. The stacking sequence
of these layers is repeated three times along the $c$-axis: $\text{Al}_3(B)\text{-Ni}_3(C)\text{-Al}_3(A)\text{-Al}_3(C)\text{-Al}_3(B)\text{-Yb}_2\text{Al}_3(A)$ [3]. Here, we use the notation of the site $A$, $B$ and $C$ that is introduced for the stacking of the closed-packed layers. The unit cell contains 78 atoms. The shortest Yb-Yb distance of $a/\sqrt{3} = 0.42$ nm in the Yb$_2$Al$_3$ layer is less than half of the Yb-Yb distance of $c/3 = 0.91$ nm along the $c$-axis. We expect that the unique two-dimensional magnetic honeycomb lattice structure will give rise to unusual magnetic properties. Here, we report the first results of electrical resistivity and magnetization measurements on single crystalline YbNi$_3$Al$_9$.

2. Experimental

Single crystals of YbNi$_3$Al$_9$ are synthesized in Al flux [4]. The excess Al surrounding the grown crystals is removed using centrifugal force and then corroded away using HCl solution. Crystallographic characterization is performed by means of X-ray powder diffraction. Since the X-ray powder diffraction pattern of YbNi$_3$Al$_9$ is in good agreement with that of ErNi$_3$Al$_9$, we conclude that the crystal structure of YbNi$_3$Al$_9$ is trigonal ErNi$_3$Al$_9$-type [3]. Additional reflections of impurities are not observed. The obtained lattice parameters are $a = 0.7270(5)$ nm and $c = 2.736(4)$ nm. The single crystalline nature is confirmed using the Laue method. The magnetization $M$ and electrical resistivity $\rho$ are measured in the temperature range of 1.8-300 K. Magnetization measurements are carried out using a SQUID magnetometer (Quantum Design) in magnetic fields $H$ of up to 50 kOe. The electrical resistivity measurements are performed using the usual dc four-probe method.

3. Results and Discussion

Temperature dependence of electrical resistivity for YbNi$_3$Al$_9$ and LuNi$_3$Al$_9$ with the current $J||a$-axis ($\rho_a$) and $J||c$-axis ($\rho_c$) is shown in Fig. 1. The resistivity of LuNi$_3$Al$_9$ shows typical metallic behavior. Slight anisotropy is observed between $\rho_a$ and $\rho_c$. In both cases, the residual resistivity is less than 1 $\mu\Omega$cm and the residual resistivity ratio is approximately 30. These values indicate that the crystals of LuNi$_3$Al$_9$ are of good quality. The electrical resistivity of YbNi$_3$Al$_9$ corresponds to the typical properties of heavy-fermion antiferromagnets with a magnetic ordering temperature of 3.4 K. The low value of resistivity at 1.5 K (approximately 5 $\mu\Omega$cm) indicates minimal impurity scattering.

The magnetic contribution of YbNi$_3$Al$_9$ to resistivity, $\rho_m$, is obtained by subtracting the resistivity of LuNi$_3$Al$_9$ from that of YbNi$_3$Al$_9$; this magnetic contribution is plotted against $\log T$ in Fig. 2. $\rho_m$ along the $a$-axis is proportional to $-\log T$ between 100 and 30 K. The $-\log T$
dependence is characteristic of the Kondo scattering. Along the c-axis, the \(-\log T\) dependence is relatively weak. This may be due to a characteristic two-dimensional array of magnetic Yb ions perpendicular to the c-axis in YbNi\(_3\)Al\(_9\). The broad hump around 30 K is attributed to the competition between the Kondo effect and the CEF effect which is expected to divide the \(J = \frac{7}{2}\) multiplet in four Kramer’s doublets. The strong reduction of the magnetic scattering at \(T = 3.4\) K is attributed to the magnetic ordering.

Figure 3 shows the temperature dependence of reciprocal magnetic susceptibilities \(\chi^{-1} = (M/H)^{-1}\) of YbNi\(_3\)Al\(_9\) measured in a magnetic field of \(H = 1\) kOe. In both magnetic field directions, the susceptibilities of YbNi\(_3\)Al\(_9\) show Curie-Weiss behavior above 80 K. Below 30 K, a large deviation from the Curie-Weiss law are observed. This behavior may be ascribed to the CEF effect. The effective magnetic moment \(\mu_{\text{eff}}\) and the paramagnetic Curie temperatures \(\Theta_p\) are estimated to be 4.55\(\mu_B/\text{Yb-ion}\) and \(-12\) K for the a-axis and 4.65\(\mu_B/\text{Yb-ion}\) and +12K for the c-axis, respectively. These values of \(\mu_{\text{eff}}\) agree with those expected in case of free Yb\(^{3+}\) ions (i.e., \(\mu_{\text{eff}} = 4.54\mu_B\)), accounting for the experimental accuracy. Since LuNi\(_3\)Al\(_9\), in which the rare-earth element Lu is not expected to carry magnetic moment, shows diamagnetism, the Ni ion in the compounds has no magnetic moment. The room-temperature susceptibility value of LuNi\(_3\)Al\(_9\) is \(-3\times10^{-4}\) emu/mol.

The magnetization curves for both \(H||a\)- and \(H||c\)-axis in a low magnetic field region at 2K are shown in Fig. 4. With increase in the magnetic field, the magnetization along the a-axis shows a rapid increase up to \(H_1\), followed by a linear increase, and then a metamagnetic transition at around \(H_2 = 1\) kOe. It should be noted that these field-induced anomalies occur at very low fields. Since these anomalies disappear at the magnetic transition temperature (Fig. 6), they must be an intrinsic property of YbNi\(_3\)Al\(_9\). Above \(H_2\), the magnetization for a-axis is gradually increased and reaches 1.60\(\mu_B\) at 50 kOe, as shown in the inset of the figure. In contrast, the magnetization along the c-axis increases smoothly and reaches 1.75\(\mu_B\) at 50 kOe. These values are consistent with the magnitude of the magnetic moment of the ground state Kramer’s doublet induced by CEF. Residual magnetization and hysteresis are not observed along both directions, within experimental accuracy.

For obtaining the temperature dependence of magnetic susceptibility in case of YbNi\(_3\)Al\(_9\) at around the magnetic ordering temperature of 3.4 K, we measured the magnetic field dependence of magnetization along the a-axis in the temperature range from 2 to 4 K in steps of 0.2 K. As shown in Fig. 4, we determine \(\chi_a\) and \(\chi_c\) as the slopes of \(M-H\) curves in the low field region.
The temperature dependence of $\chi_a$ and $\chi_c$ is shown in Fig. 5. The different susceptibility scales between $\chi_a$ and $\chi_c$ are noteworthy. There is a large anisotropy in the magnitude of susceptibility along two crystallographic directions. $\chi_a$ shows a peak at 3.2 K, while $\chi_c$ is almost constant below 3.4 K. These characteristic features of susceptibility can be ascribed to the antiferromagnetic ordering with the magnetic easy plane perpendicular to the $c$-axis. This result is consistent with the observation of metamagnetic transition along the $H$/$a$-axis.

We have estimated the field-induced spontaneous magnetization $M_{s,a}$ using the intercept of linear extrapolation of the $M$-$H$ curve, as shown in Fig. 4. The value of $M_{s,a}=0.18\mu_B$ at 2K amounts to approximately 10% of that expected for the Kramer’s doublet ground state of the Yb$^{3+}$ ion. The field-induced spontaneous magnetization may be considered to arise from a canted antiferromagnetic structure.

Finally, we show the temporary magnetic field-temperature phase diagram of YbNi$_3$Al$_9$ for the magnetic field along the $a$-axis in Fig. 6. The magnetic structure of YbNi$_3$Al$_9$ is highly sensitive to the magnetic field. A more detailed study is required to understand the magnetic structure of YbNi$_3$Al$_9$.

4. Summary
We have synthesized single crystals of YbNi$_3$Al$_9$. YbNi$_3$Al$_9$ is a heavy-fermion antiferromagnet with $T_N = 3.4$K. The magnetic structure of YbNi$_3$Al$_9$ is highly sensitive to the magnetic field. To obtain more detailed information about the magnetic phase diagram and the magnetic structure of YbNi$_3$Al$_9$, specific heat and neutron diffraction experiments under magnetic fields are being conducted. A more detailed analysis of the magnetic properties of YbNi$_3$Al$_9$ will be reported elsewhere.

Acknowledgments
The authors acknowledge Dr. Y. Uwatoko, Dr. Y. Aoki, and Dr. K. Mibu for their helpful discussions.

References
[1] See e.g., Doniach 1977 Valence Instabilities and Relation Narrow Band Phenomena (New York: Plenum)
[2] Fisk Z., Maple M. B. 1992 J. Alloys Compd. 183 303
[3] Gladyshevskii R. E., Cenzual K., Flack H. D., and Parthe E. 1993 Acta Cryst. 49 468
[4] Ohara S., Chen G. F., Sakamoto I. 2001 J. Alloys Compd. 323-324 632