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Authors
Matsumoto, Yuji
Matsuda, Tatsuma D
Tateiwa, Naoyuki
et al.

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Single Crystal Growth and Physical Properties of UT_2Al_{20} (T=Transition Metal)

Yuji Matsumoto,* Tatsuma D. Matsuda, Naoyuki Tateiwa, Etsuji Yamamoto and Yoshinori Haga
Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan

Zachary Fisk
Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan and University of California, Irvine, CA 92697, U.S.A.

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We have studied the single crystal growth and the physical properties of the UT_2Al_{20} system. We have successfully grown UT_2Al_{20} (T = Cr, Ti, W and Mo) using Al self flux and have characterized the crystals using electron probe microanalyzer (EPAM) and X-ray diffractometer. We have also measured their magnetic and electric properties. The ground states of UT_2Al_{20} (T = Cr, Mo and W) are Pauli paramagnetic as is likely also in UTi_2Al_{20}. The effective masses of UCr_2Al_{20} and UTi_2Al_{20} are only weakly enhanced.

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I. INTRODUCTION

Many anomalous physical properties have been observed in RT_2X_{20} (R=Rare earth metal, T=transition metal, X=Al, Zn) system. For example, quadrupolar order occurs in PrT_2X_{20} systems [1–3] arising from a Γ_3 ground state, an extreme heavy fermion state was observed in $YbCo_2Zn_{20}$ with specific coefficient reaching 8 J/mol K^2 [4], and the effect of geometric frustration was observed in YMn_2Zn_{20} [5]. These properties derive from the interesting crystal structure. This crystal structure is a cubic $CeCr_2Al_{20}$-type structure with space group ($Fd\bar{3}m$). Strong hybridization between f and conduction electrons is expected with the R ions coordinated by sixteen Al ions. Further, the R and T atoms occupy the 8a and 16d sites forming diamond and pyrochlore lattices, respectively. The UT_2X_{20} compounds are worth investigating because the extended 5f wave function in uranium could lead to interesting features due to strong hybridization, and if UT_2X_{20} has f^2 configuration, quadrupolar order is possible. Previous studies found UT_2Zn_{20} systems to have heavy fermion behavior [6,7]. There are few studies on UT_2Al_{20} systems [7–11], prompting the studies of the single crystal growth and physical properties of UT_2Al_{20} systems reported here.

*E-mail: matsumoto.yuji@nitech.ac.jp

II. EXPERIMENTS

Single crystals of UT_2Al_{20} (T=Ti, Cr, Mo, W) were grown in Al flux by using 3N8 (99.98) U, 4N Ti, 3N Cr, 3N Mo, 3N W and 5N Al. These mixtures were placed in an alumina crucible and sealed in an evacuated quartz tube. The sealed tubes were heated to 1150 °C, soaked for 12 h, then cooled down to 700 °C in 96 h. The excess Al was spun off in a centrifuge. We also attempted growths of UT_2Al_{20} (T = V, Ta, Nb) but only obtained small single crystals.

The single crystals of UT_2Al_{20} were characterized by an electron probe microanalyzer (EPAM), a X-ray powder diffractometer and a single crystal X-ray diffractometer. X-ray powder diffraction patterns of UT_2Al_{20} are shown in Fig. 1. Almost all the Bragg peaks of UT_2Al_{20} can be indexed as the cubic $CeCr_2Al_{20}$ type structure. The content and the homogeneity of the crystals were analyzed by using EPMA. Figure 2 shows scanning electron microscope images of UTi_2Al_{20}. The crystal structure of UTi_2Al_{20} was verified via X-ray single crystal diffraction measurements. Table 1 shows atomic site parameters and isotropic atomic displacement parameter B_{eq} of UTi_2Al_{20}.

DC magnetic measurements were performed using a superconducting quantum interference device magnetometer (Quantum Design MPMS) at temperatures from 300 to 2 K. The shape of UT_2Al_{20} single crystals are octahedral and its side direction is the [110] direc-

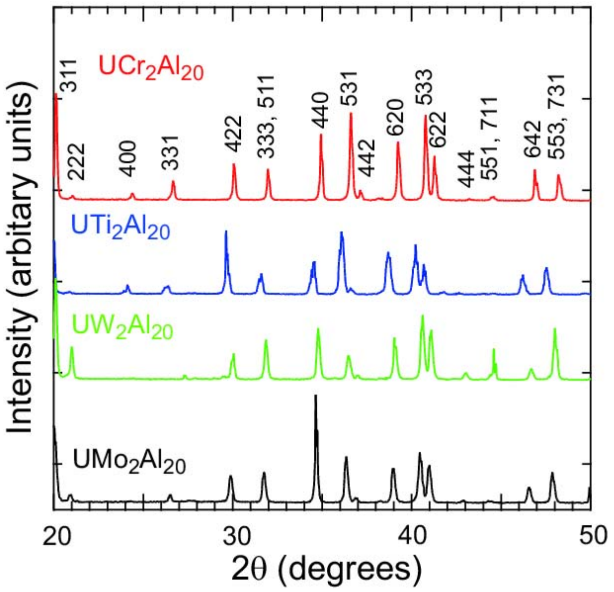

Fig. 1. (Color online) X-ray powder diffraction patterns of UT_2Al_{20} (T = Cr, Ti, W and Mo) for angle ranging from 20 to 50 degrees. The indexes are for the cubic $CeCr_2Al_{20}$ structure.

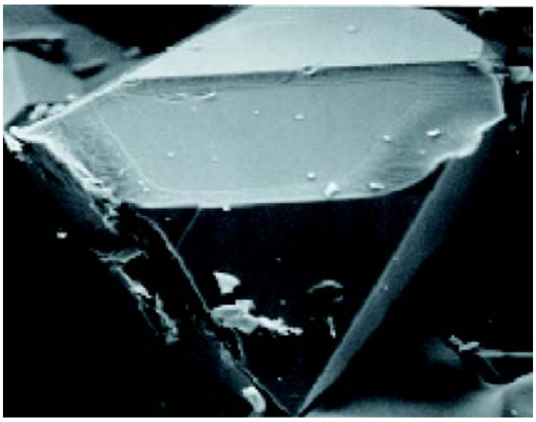

Fig. 2. Scanning electron microscopic images of UTi_2Al_{20}. The sample length is about 0.2 mm.

tion. We used several single crystals for UTi_2Al_{20} and UW_2Al_{20} measurements because we could not obtain a large enough single crystal to measure magnetic properties. The transport measurements were performed using a conventional DC four terminal technique in a ^{3}He cryostat.

III. RESULTS AND DISCUSSION

We measured the magnetic properties of UT_2Al_{20} (T=Ti, Cr, Mo, W). Figure 3 shows the magnetic susceptibility of UT_2Al_{20} (T=Ti, Cr, Mo, W) as a function of temperature with a magnetic field parallel to the [110] direction at 0.1 T. In the susceptibilities of UT_2Al_{20}, we

Table 1. Atomic site parameters and isotropic atomic displacement parameters B_{eq} of UTi_2Al_{20}.

Atom	Wyckoff-Symbol	x	y	z	B_{eq}
U	8(a)	0.125	0.125	0.125	0.45795
Ti	16(d)	1/2	0	0	0.26924
Al1	16(c)	0	0	0	0.97906
Al2	96(g)	0.05989	0.19011	–0.07351	0.67902
Al3	48(f)	0.125	0.125	–0.23733	0.51585

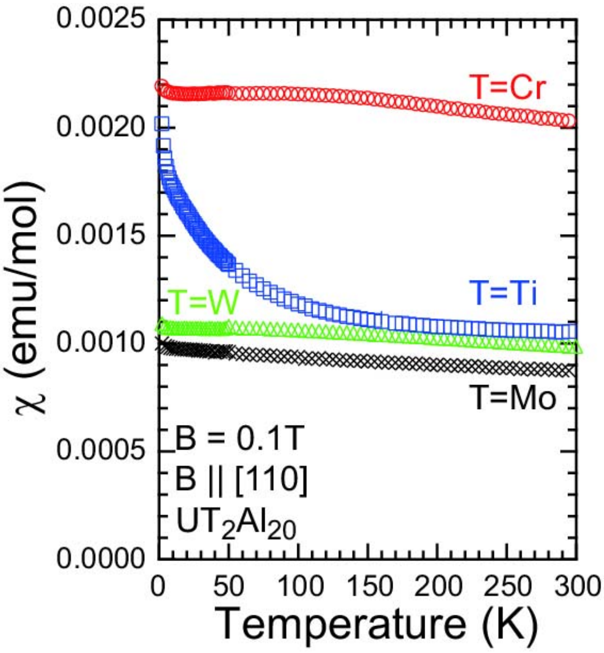

Fig. 3. (Color online)Temperature dependence of the magnetic susceptibility of UT_2Al_{20} at 0.1 T with fields parallel to the [110] direction. The circles, squares, triangles and crosses denote UCr_2Al_{20}, UTi_2Al_{20}, UW_2Al_{20} and UMo_2Al_{20}, respectively.

see no evidence for an obvious phase transition. The susceptibilities of UCr_2Al_{20}, UW_2Al_{20} and UMo_2Al_{20} are almost temperature independent, indicating that the ground states of UCr_2Al_{20},UW_2Al_{20} and UMo_2Al_{20} are Pauli paramagnetic. If the ground states of UCr_2Al_{20}, UW_2Al_{20} and UMo_2Al_{20} are Pauli paramagnetic with $\chi(T = 0\ K)$ of UCr_2Al_{20}, UW_2Al_{20} and UMo_2Al_{20} being 0.002, 0.0011 and 0.0007 emu/mol respectively, we estimate that the electronic specific coefficients γ are about 70, 30 and 20 mJ/mol K^2 , respectively, based on a Wilson ratio $R_w = 2$. According to previous specific heat measurements, the γ of UCr_2Al_{20} is about 80 mJ/mol K^2 [9,10]. Therefore, the $\chi(T = 0\ K)$ seem reasonable values. In the Okuda *et al.*, study [9], UCr_2Al_{20} shows weak ferromagnetic order around 200 K. On the other hand, UCr_2Al_{20} does not show ferromagnetic behavior in ref. [10]. Our measurements are similar to those of Swatek *et al.* It is possible that the weak ferromagnetic

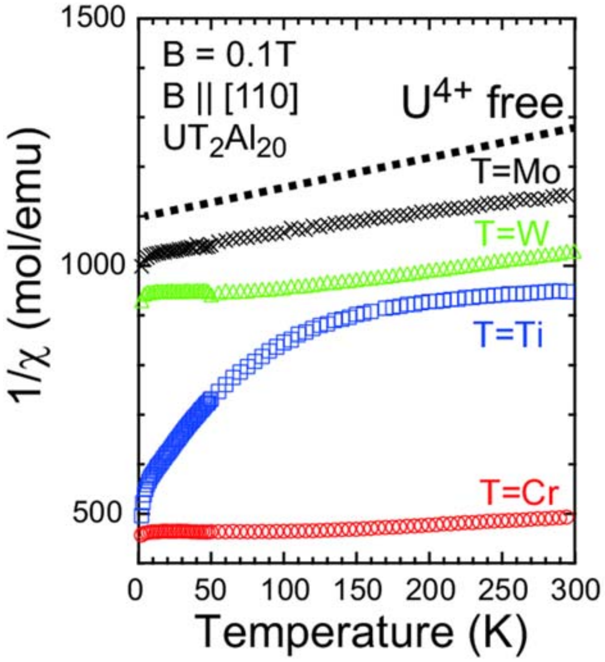

Fig. 4. (Color online) Temperature dependence of inverse magnetic susceptibility of UT_2Al_{20} at 0.1 T with fields parallel to the [110] direction. The slope of broken line is calculated by using U^{4+}. The circles, squares, triangles and crosses denote UCr_2Al_{20}, UTi_2Al_{20}, UW_2Al_{20} and UMo_2Al_{20}, respectively.

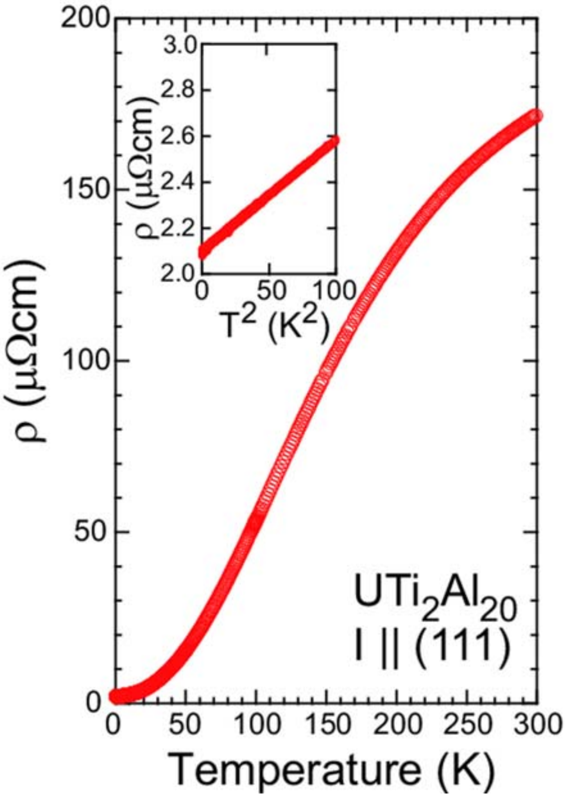

Fig. 5. (Color online) Temperature dependence of the resistivity of UTi_2Al_{20}. The inset is the resistivity vs. T^2 curve for temperature ranging from 0 to 10 K. The current is applied in the (111) plane.

order is due to the impurity phase described by Okuda *et al.*

On the other hand, the susceptibility of UTi_2Al_{20} differs from that of the other UT_2Al_{20} systems in that the susceptibility of UTi_2Al_{20} is temperature dependent and does not saturate at our lowest temperature, 2 K. The susceptibility of UTi_2Al_{20} is almost temperature independent at temperatures between 300 to 100 K, then gradually increases with decreasing temperature below 100 K, and finally rapidly increasing below 5 K. This increase of susceptibility below 5 K is likely due to an impurity phase because the increase was rapidly suppressed by a magnetic field. The increase below 100 K was not suppressed at 7 T, so it seems possible that the increase in the susceptibility below 100 K is not due to an impurity phase but rather an intrinsic property.

Figure 4 shows the inverse magnetic susceptibility of UT_2Al_{20} (T=Ti, Cr, Mo, W) as a function of temperature. The slope of the broken line is calculated using U^{4+}. The inverse magnetic susceptibility of UT_2Al_{20} at high temperature deviates from that expected for the U^{4+} configuration, implying that the valence of the U ion in UT_2Al_{20} is not U^{4+} but that the f electrons are itinerant.

Because the susceptibility of UTi_2Al_{20} differs from that of other UT_2Al_{20} systems, we measured its resistivity. Figure 5 shows the resistivity of UTi_2Al_{20} as a function of temperature. The inset in Fig. 5 shows the resistivity as a function of T^2. Using the expression $\rho = \rho_0 + AT^2$, we estimate the value of the A coefficient to be 0.005 $\mu\Omega$cm/K^2 and the residual resistivity ρ_0 of 2.1 $\mu\Omega$cm/K^2 (residual resistance ratio (RRR) is about 80) . A Fermi liquid T^2 dependence is observed below 10 K, and the ground state of UTi_2Al_{20} is likely to be paramagnetic. The value of the A coefficient of UTi_2Al_{20} is similar to that of paramagnetic CeNi whose electronic specific heat coefficient γ is about 65 mJ/mol K^2 [12]. This suggests that the effective mass of UTi_2Al_{20} is similar to that of CeNi and a little enhanced. The value of residual resistivity indicates that the UTi_2Al_{20} crystal is of high quality, and that possibly the increase of susceptibility below 100 K is an intrinsic property. However, as we described in Section II, we used several single crystals to measure the magnetic properties, and impurity phases were observed, although the value of ρ_0 is small. Therefore it is possible that the increase of susceptibility below 100 K is also from an impurity phase. Further detailed investigation is required in UTi_2Al_{20} to resolve this question.

We summarize our results. We have successfully grown single crystals of UT_2Al_{20} (T = Cr, Ti, W and Mo) by using Al flux. The samples were characterized using EPAM and X-ray diffraction method. We have measured the magnetic and electric properties of UT_2Al_{20} (T = Cr, Ti, W and Mo). These results indicate that the ground sates of UT_2Al_{20} (T = Cr, Mo and W) are paramagnetic, the ground state of UTi_2Al_{20} is likely to be paramagnetic, with the effective masses of UCr_2Al_{20} and UTi_2Al_{20} little enhanced.

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