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Magnetic and thermal properties of NdT_2Al_{20} (T: Ti, V, Cr) single crystals

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Abstract. We have succeeded in growing the caged cubic compounds NdT_2Al_{20} (T: Ti, V, Cr) by the Al-self flux method, and measured magnetic susceptibility χ, magnetization M, and specific heat divided by temperature C/T down to 0.5K. From the measurements, the antiferromagnetic phase transition at T_N = 1.45 K was observed for $NdTi_2Al_{20}$, and ferromagnetic phase transition at T_C = 1.8 K and 1.7 K were observed for NdV_2Al_{20} and $NdCr_2Al_{20}$, respectively.

1. Introduction

Recently, cage-structured compounds such as filled skutterudites have attracted much attention due to the novel properties such as the heavy fermion behavior, unconventional superconductivity and multiple ordering in the Pr- and Sm-based compounds[1, 2, 3, 4, 5]. Among them, the compounds RT_2X_{20} (R: rare earths, T: transition metals, X: Al, Zn, and Cd) also have the caged and cubic structure, also show the novel properties. Pr-based compounds PrT_2Zn_{20} (T = Ir, Rh)[6, 7, 8] and PrT_2Al_{20} (T = Ti, V, Cr, Nb)[9, 10, 11, 12, 13, 14] show the superconductivity, multipole ordering, and the unconventional Kondo effect. These novel properties are coming from a non-Kramers Γ_3 doublet CEF ground state which has nonmagnetic quadrupole moments[9, 7, 15, 16]. For instance, $PrTi_2Al_{20}$ is a heavy fermion superconductor based on strong hybridization between conduction electrons and nonmagnetic quadrupole moments of the Γ_3 ground state[17]. Except the PrT_2X_{20} and SmT_2X_{20}, only a few compounds are studied[12, 18, 19, 20, 21]. To clarify these novel properties, systematic studies are important. We were succeeded in making new NdT_2Al_{20} (T: Ti, V, Cr) polycrystalline samples, and studied magnetic and electronic properties[20]. Recently, we have succeeded in growing high quality NdT_2Al_{20} (T: Ti, V, Cr) single crystals, and studied magnetic and thermal properties.

2. Experimental details

Single crystals of NdT_2Al_{20} (T: Ti, V, Cr) were grown by the Al-self flux method. The quality of the samples was checked using a powder x-ray diffractometer with Cu-Kα radiation. All obtained peaks were indexed with the cubic $CeCr_2Al_{20}$ (Fd$\bar{3}$m) type patterns, suggesting the high quality of samples. This is consistent with the fact that the residual resistivity ratio reaches to 20 for $NdTi_2Al_{20}$ (not shown), which is $\sim$ 4 times higher than the previous report[20]. The evaluated lattice parameters a were 14.77 Å, 14.55 Å, and 14.51 Å, which is consistent with the

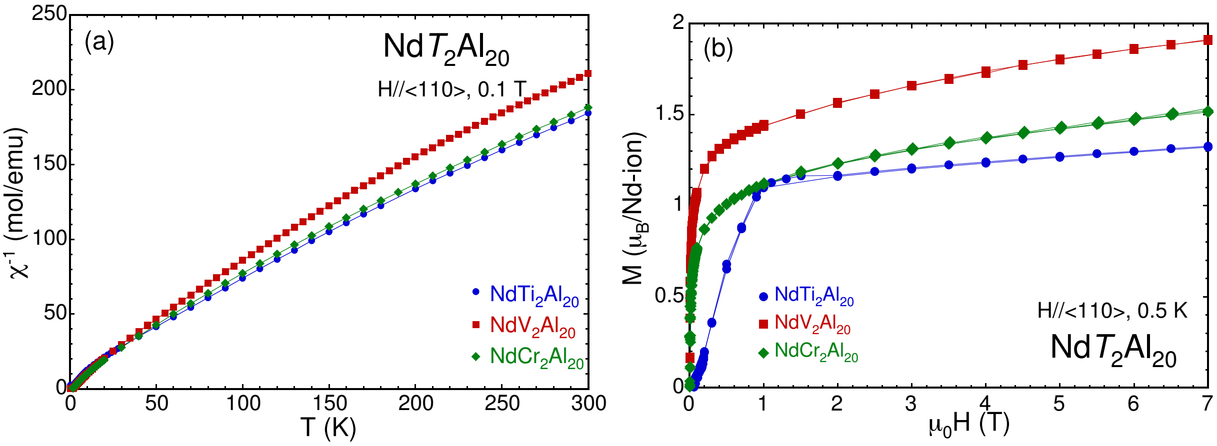

Figure 1. (a) The temperature dependence of the inverse susceptibility ($1/\chi$) for $NdTi_2Al_{20}$ (circle), NdV_2Al_{20} (square), and $NdCr_2Al_{20}$ (diamond) at 0.1 T for $H//\langle 110\rangle$ direction. (b) The field dependence of the Magnetization (M) for $NdTi_2Al_{20}$ (circle), NdV_2Al_{20} (square), and $NdCr_2Al_{20}$ (diamond) at 0.5 K for $H//\langle 110\rangle$ direction.

previous report[19]. The crystal axes of the single crystals were determined from Laue pictures. DC magnetization measurements were carried out in a Quantum Design (QD) MPMS down to 1.9 K, and QD MPMS iHelium3 down to 0.5 K and up to 7 T. Specific heat (C) measurements were performed using a quasi-adiabatic method with a QD PPMS down to 0.5 K and up to 9 T.

3. Experimental results and discussions

Figure 1 (a) shows the temperature dependencies of the inverse susceptibility $(\chi)^{-1}$ for NdT_2Al_{20} (T = Ti, V, and Cr) at 0.1 T and $H//\langle 110\rangle$ direction. At high temperatures, the temperature dependence of the susceptibility (χ) for NdT_2Al_{20} shows Curie-Weiss like temperature dependencies above 200 K, and evaluated the Curie-Weiss temperature Θ_{CW} = -65 K, -79 K, -69 K and the effective magnetic moments of μ_{eff} = 3.98 μ_B, 3.78 μ_B, and 3.95 μ_B,for $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$, respectively. μ_{eff} values are consistent with the theoretical value expected for the full multiplet of Nd^{3+} ($3.64\mu_B$) for these compounds. For $NdCr_2Al_{20}$, the μ_{eff} value becomes smaller than that obtained from the polycrystalline sample[20], and consistent with the no contribution of excess Cr. At low temperatures, χ monotonically increases down to 2 K for NdT_2Al_{20}. Below 2 K, χ^{-1} for $NdTi_2Al_{20}$ shows clear kink due to the antiferromagnetic phase transition at 1.4 K, on the other hand, χ^{-1} for NdV_2Al_{20} and $NdCr_2Al_{20}$ quickly decreases $\sim$ 1.8 K and $\sim$ 1.7 K, indicating the ferromagnetic phase transition. Figure 1 (b) shows the field dependences of the magnetization (M) for $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$ at 0.5 K. The M values at 7 T are below 2 μ_B and similar to the $NdRu_2Zn_{20}$ which has doublet Γ_6 ground state[22]. For $NdTi_2Al_{20}$, M linearly increases at low fields and shows a saturation tendency above 1 T, indicating the antiferromagnetic ground state and consistent with χ^{-1} data. For NdV_2Al_{20} and $NdCr_2Al_{20}$, M increases at very low fields and then shows saturation tendency. The field dependence of M of these two compounds

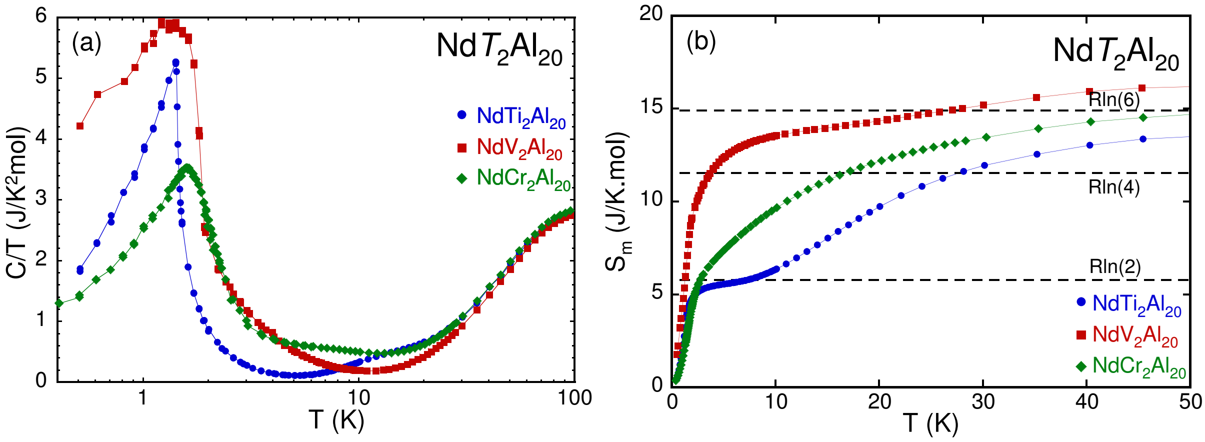

Figure 2. (a) The temperature dependence of the specific heat divided by the temperature (C/T) for $NdTi_2Al_{20}$ (circle), NdV_2Al_{20} (square), and $NdCr_2Al_{20}$ (diamond) at 0 T. (b) the temperature dependences of the magnetic part of the entropy ($S_{\rm m}$) for $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$.

shows hysteresis at very low fields (below 0.05 T). These behaviors indicate that NdV_2Al_{20} and $NdCr_2Al_{20}$ have the ferromagnetic ground state and consistent with the χ^{-1} data.

Figure 2 (a) shows the temperature dependences of the specific heat divided by temperature (C/T) for $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$. At high temperatures, the temperature dependences of C/T for these three compounds shows the almost same values of LaT_2Al$_{20}$, indicating that the $4f$ electrons are well localized. Reflecting the high quality of the sample, no anomaly is observed around 10 K in NdV_2Al_{20} in contrast to the polycrystalline sample[20] which may come from the impurity. At low temperatures, C/T for $NdTi_2Al_{20}$ and $NdCr_2Al_{20}$ shows a broad peak or plateau, which become peak considering only the magnetic part of C/T ($C_{\rm m}/T$) obtained from subtracting the phonon part of C/T, at $\sim$ 20 K and $\sim$ 10 K, respectively. For NdV_2Al_{20}, $C_{\rm m}/T$ shows a peak at higher temperature, $\sim$ 40 K (not shown). These peaks are may come from the Schottky peak due to the CEF. Below 4 K, C/T increases with decreasing temperature, and shows clear anomaly at 1.45 K, 1.8 K, and 1.7 K for $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$, indicating the phase transition. Figure 2 (b) shows the temperature dependencies of the magnetic part of the entropy $S_{\rm m}$ for $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$. At low temperatures, $S_{\rm m}$ quickly increases with increasing temperature and reaches $\sim R\ln 2$ at transition temperatures for these compounds, indicating the CEF doublet Γ_6 ground state for $NdTi_2Al_{20}$ and $NdCr_2Al_{20}$. However, because $S_{\rm m}$ exceeds $\ln 4$ near $T_{\rm C}$, it is difficult to decide the CEF ground state.

4. Summary

We have succeeded in growing single crystals, and studied magnetic and thermal properties of $NdTi_2Al_{20}$, NdV_2Al_{20}, and $NdCr_2Al_{20}$. From magnetization and specific heat measurements, we found that NdV_2Al_{20} and $NdCr_2Al_{20}$ shows the ferromagnetic phase transitions at 1.8 K and 1.7 K, and $NdTi_2Al_{20}$ shows antiferromagnetic phase transition at 1.45 K. The CEF ground state should be doublet Γ_6 ground state for $NdTi_2Al_{20}$ and $NdCr_2Al_{20}$.

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