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Ferromagnetic transition in a caged compound $NdOs_2Zn_{20}$

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Abstract
Magnetic properties in a caged compound $NdOs_2Zn_{20}$ have been studied by the measurements of magnetization M, magnetic susceptibility M/B, and specific heat C. The measurements indicate that $NdOs_2Zn_{20}$ shows a structural transition at $T_s = 62$ K and a ferromagnetic transition at $T_C = 0.6$ K. The structural transition is also observed in the La counter-part $LaOs_2Zn_{20}$ at $T_s = 151$ K. The results of $M(T)/B$, $M(B)$, and $C(T)$ of $NdOs_2Zn_{20}$ are reproduced by a crystalline electrical field scheme with a Kramers doublet ground state.

Keywords: RT_2Zn_{20}, Caged compound, Crystalline electric field

1 Introduction

Caged compounds RT_2Zn_{20} (R: Rare earth, T: Transition metals) have attracted much attention because they show a variety of interesting phenomena arising from $4f$ electrons of the rare-earth ions and the characteristic cage-structure. $YbCo_2Zn_{20}$ has the largest electronic specific heat coefficient of 8 J/K^2 mol, which is considered to sit in the vicinity of a quantum critical point [1]. Heavy fermion states were also reported in CeT_2Zn_{20} (T=Ru, Ir) [2, 3]. $PrIr_2Zn_{20}$ with the non-Kramers Γ_3 doublet ground state of $4f^2$ configuration is the first example showing the superconducting transition in the presence of anti-ferro quadrupole order [4]. LaT_2Zn_{20} (T=Ru, Ir) and $PrRu_2Zn_{20}$ show structural transitions at $T_s = 150$, 200, and 138 K, respectively [5]. The structural transition is thought to be induced by large amplitude vibration of the caged Zn atom at the 16c site located in the middle of the two R ions [6, 7].

In the case of R=Nd, Isikawa *et al.* recently reported that $NdRu_2Zn_{20}$ is a ferromagnet with $T_C = 1.9$ K, whose magnetic properties are well explained by the crystalline electric field (CEF) model [8]. In this paper, we report the magnetic properties of an isoelectronic compound $NdOs_2Zn_{20}$ studied by specific heat and magnetization measurements.

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2 Experiment

Polycrystalline samples of ROs_2Zn_{20} (R=La, Nd) were prepared by the Zn-self flux method. The samples were characterized by powder x-ray diffraction (XRD) measurements with RIGAKU Ultima IV and electron probe microanalysis (EPMA) with a JEOL JXA-8200 analyzer. These characterizations of $NdOs_2Zn_{20}$ revealed that the main phase is of the $CeCr_2Al_{20}$-type [9] and an impurity phase is $NdZn_{11}$. Magnetization was measured between 1.8 and 300 K by a commercial SQUID magnetometer (Quantum Design MPMS). Specific heat was measured from 0.4 to 300 K by a relaxation method (Quantum Design PPMS).

3 Results and discussion

Figure 1 shows the temperature dependence of the specific heat C of ROs_2Zn_{20} (R=La, Nd). Both data show peaks at T_s = 151 K and 62 K, respectively. These peaks are attributed to structural transitions, because a similar peak due to the structural transition was observed at 150 K in the isostructural compound $LaRu_2Zn_{20}$[5]. The anomaly in the specific heat of $NdOs_2Zn_{20}$ at the putative structural transition at 62 K is tiny. It probably results from distribution of the transition temperature due to inhomogeneity in the sample. To confirm the bulk nature of the transition, transmission electron microscopic measurements are in progress.

The specific heat divided by the temperature, C/T, at $T \leq 5$ K is shown in the inset of Fig. 1. On cooling below 3 K, C/T gradually increases and exhibits a peak at $T_C = 0.6$ K. We measured the specific heat in magnetic fields up to B=1 T to identify the phase transition. The peak shifts to higher temperatures and becomes broader with increasing the magnetic field, suggesting a ferromagnetic transition. In the isostructural ferromagnet $NdRu_2Zn_{20}$, similar behavior was reported[8]. To corroborate the ferromagnetic state, magnetization measurements at T <1.8 K are desired.

The magnetic part of the specific heat C_m and the magnetic entropy S_m of $NdOs_2Zn_{20}$ are shown in Fig. 2. We estimated C_m by subtracting C of $LaOs_2Zn_{20}$ from that of $NdOs_2Zn_{20}$,

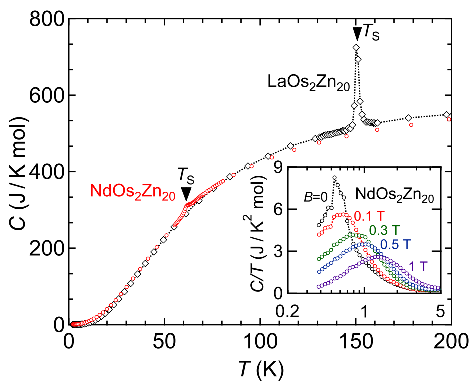

Figure 1: Temperature dependence of the specific heat C of ROs_2Zn_{20} (R=La and Nd). The inset shows C/T of $NdOs_2Zn_{20}$ in magnetic fields B = 0, 0.1, 0.3, 0.5 and 1 T.

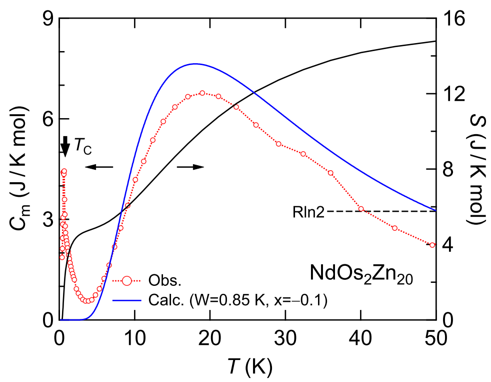

Figure 2: Temperature dependence of the magnetic specific heat C_m and the magnetic entropy S_m. The (blue) solid line shows the Schottky-type specific heat calculated for a CEF scheme described in the text.

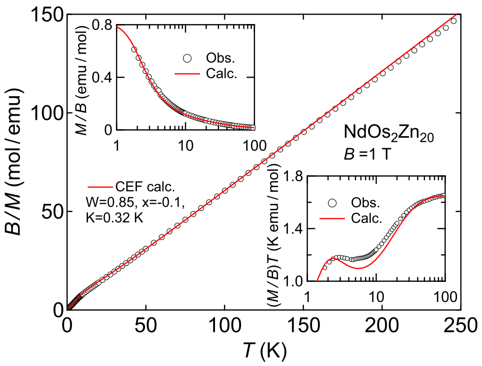

Figure 3: Temperature dependence of the magnetic field divided by magnetization B/M of $NdOs_2Zn_{20}$ measured at $B = 1$ T. The upper and lower insets show the temperature dependences of M/B and $(M/B)T$, respectively. The (red) solid lines are calculations using the CEF parameters of $W = 0.85$ K, $x = -0.1$ and the inter-site ferromagnetic interaction coefficient with $K = +0.32$ K.

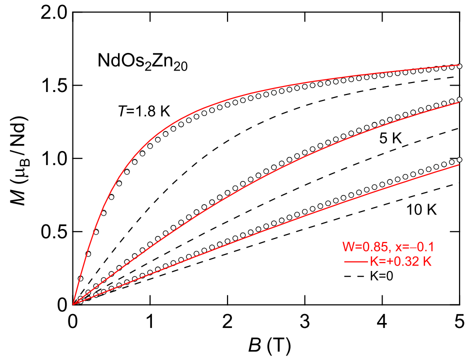

Figure 4: Magnetization curves of $NdOs_2Zn_{20}$ up to 5 T at 1.8, 5 and 10 K. The solid lines are calculated magnetization curves using the CEF parameters of $W = 0.85$ K, $x = -0.1$ and inter-site interaction coefficient $K = +0.32$ K. The dashed lines are the calculation with no inter-site magnetic interaction.

$C_m = C(NdOs_2Zn_{20}) - C(LaOs_2Zn_{20})$. The value of S_m approaches $0.8R\ln 2$ at 2 K which is followed by a plateau between 2 and 5 K, indicating that the CEF ground state is a Kramers doublet. The broad peak of C_m at 20 K is attributed to a Schottky anomaly due to the CEF splitting as will be discussed later.

Figure 3 shows the temperature dependence of the magnetic field divided by magnetization B/M of $NdOs_2Zn_{20}$ measured at B=1 T. Above 50 K, B/M obeys the Curie-Weiss law with the effective magnetic moment of 3.68 μ_B/Nd, which agrees with 3.62 μ_B/Nd expected for a free trivalent Nd ion. The magnetizations of $NdOs_2Zn_{20}$ at 1.8, 5 and 10 K are shown in Fig. 4. At 1.8 K, M reaches the value of 1.6 μ_B/Nd at 5 T. To determine CEF levels and the inter-site magnetic interaction strength, we performed mean-field calculations based on the following Hamiltonian.

$$\mathcal{H} = \mathcal{H}_{CEF} - g_J \mu_B \boldsymbol{J}\boldsymbol{H} - K\langle \boldsymbol{J} \rangle \boldsymbol{J}, \tag{1}$$

where $\mathcal{H}_{CEF}$ is the CEF Hamiltonian for the subspace of $J = 9/2$ multiplet, $g_J = 8/11$ is the Landé g-factor and μ_B is the Bohr magneton, K is a coefficient of the magnetic inter-site interaction between the Nd ions. $\mathcal{H}_{CEF}$ is described by

$$\mathcal{H}_{CEF} = W\left[\frac{x}{60}(O_4^0 + 5O_4^4) + \frac{1-|x|}{1260}(O_6^0 - 21O_6^4)\right], \tag{2}$$

where the notation by Lea *et al.* is used [10]. We employ parameters of $W = 0.85$ K, $x = -0.1$ and $K = 0.32$ K to reproduce the magnetization curves. We assumed the cubic symmetry of Nd site to simplify the calculation, although $NdOs_2Zn_{20}$ exhibits the structural transition at $T_s = 62$ K to an undetermined structure.

The calculated magnetization curves in the magnetic field along the [100] direction are shown by the (red) solid line in Fig. 4. We note that the calculated curves in the field along the [110] and [111] direction are almost the same as that for along the [100] direction. The calculations agree with the experimental data, suggesting the validity of the CEF parameters used for the calculation. In order to deduce the inter-site interaction, the calculation with no inter-site interaction coefficient K is shown by the dashed line. With increasing magnetic field, M rises more rapidly than the calculation, supporting the ferromagnetic inter-site interaction. We confirmed that M/B, $(M/B)T$, and B/M are reproduced by the calculation as shown by the (red) solid line in Fig. 3.

The set of the parameters for $NdOs_2Zn_{20}$ yields the CEF level scheme; Γ_6 doublet (0) -$\Gamma_8^{(1)}$ quartet (39 K) - $\Gamma_8^{(2)}$ quartet (92 K). The doublet ground state in $NdOs_2Zn_{20}$ is consistent with the expectation from the magnetic entropy. The calculated Schottky-type specific heat is shown by the (blue) solid line in Fig. 2. The peak position is roughly reproduced, whereas the peak height is a little suppressed compared with the calculation. One possible reason for this discrepancy is that the lattice contribution of the specific heat is overestimated. Another is a symmetry lowering of the Nd site due to the structural transition. The lowering symmetry of the Nd site splits the excited quartet states into two doublets. The splitting possibly broadens the Schottky peak.

Finally, let us discuss why the ferromagnetic transition temperature T_C = 0.6 K for NdT_2Zn_{20} with T = Os is lower than T_C=1.9 K for T=Ru. In GdT_2Zn_{20} (T = Ru, Os), T_C = 20 K for T=Ru is approximately five times higher than T_C = 4.2 K for T = Os [11]. It was pointed out that a magnetic correlation between the Gd^{3+} and Ru atom favors the higher T_C for $GdRu_2Zn_{20}$. On the analogy, a magnetic correlation between the Nd^{3+} and Ru atom would play a role to stabilize the ferromagnetic order in $NdRu_2Zn_{20}$.

4 Conclusion

We have synthesized caged compounds ROs_2Zn_{20} (R = La, Nd) and measured the magnetization and specific heat. Both compounds show structural transitions at T_s = 151 and 62 K, respectively. A ferromagnetic order was observed in $NdOs_2Zn_{20}$ at T_C = 0.6 K. The magnetic entropy estimated from the specific heat approaches $0.8R\ln 2$ at 2 K. The CEF calculation revealed that the ground state is a Kramers doublet.

Acknowledgments

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