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Low Temperature Magnetic Ordering in $NdAgAl_3$

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We present the detailed study of magnetic, thermodynamic, and transport properties of polycrystalline $NdAgAl_3$. The compound crystallizes in $BaNiSn_3$-type tetragonal structure with the space group $I4mm$. Magnetic, heat capacity and transport measurements indicate the possible antiferromagnetic nature of the ordering below 2 K. The compound shows the Schottky anomaly in heat capacity data. Magnetoresistance is negative at low temperature.

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1. Introduction

Several RTX_3 compounds have been reported in the last few decades (where R = rare earth, T = $3d/4d/5d$ — transition metal and X = p-block element), which exhibit very interesting ground state properties. For example, the $PrRhSn_3$, with well-ordered crystal structure, displays spin glass behaviour [1]. The Kondo lattice and ferromagnetism are indicated in $CeNi_xGa_{4-x}$ [2]. Strong magnetocrystalline anisotropy is observed in $RCuGa_3$ [3]. $CeRhSi_3$ [4] reveals the quantum critical point as well as heavy-fermion behaviour and it crystallizes in the tetragonal $BaNiSn_3$-type structure. Based on these intriguing physical properties, we are concentrating on RTX_3-type intermetallic compounds. Our recent reports of $RCuGa_3$ series (R = La, Pr, Nd and Gd) show prominent magnetocrystalline anisotropy behaviour [3], $RCoSi_3$ (R = Pr, Nd and Sm) exhibits CEF effects as well as interesting magnetoresistance [5] reveals the ferromagnetic order at low temperature. In Ref. [6] the neutron scattering and study of physical properties of $NdCuAl_3$ revealed that it crystallizes in non-centrosymmetric tetragonal $BaNiSn_3$ type structure and it has antiferromagnetic ordering at 2.35 K. Therefore, in continuation of our work, we report herewith the systematic investigations of structure, magnetic properties, specific heat and electrical resistivity of polycrystalline $NdAgAl_3$.

2. Experimental

The polycrystalline sample of $NdAgAl_3$ was synthesized by arc-melting under the purified argon atmosphere. The elements of high purity Nd (99.6%), Ag (99.9999%), Al (99.9999%) were taken in the stoichiometric 1:1:3 ratios. The resulting alloy button was turned over and remelted several times to ensure the homogeneity. The sample was closed in an evacuated quartz ampoule and it was annealed for 20 days at 700 °C. Powder X-ray diffraction data were taken using Cu K_α to confirm the phase purity and energy dispersive X-ray spectroscopy analysis was carried out to ensure the stoichiometry of annealed sample.

Magnetization was measured as a function of temperature as well as the function of magnetic field using the Quantum Design Physical Property Measurement System (PPMS) from 2 K to 300 K up to 5 T. Also, the heat capacity was measured using PPMS and relaxation technique down to 1.8 K in the applied magnetic fields up to 9 T. Four probes resistivity measurements were carried out using PPMS from 2–300 K up to 9 T.

3. Results and discussion

Figure 1 shows powder X-ray diffraction pattern of $NdAgAl_3$ recorded at room temperature along with structural Rietveld refinement profile using GSAS software. The refinement data reveal that the $NdAgAl_3$ compound adopted tetragonal $BaNiSn_3$-type structure with the space group $I4mm$, which is distorted from $BaAl_4$ tetragonal structure. The corresponding lattice parameters and atomic positions are given in Table I, which are in good agreement with the reported isotypic $RCoSi_3$ (R = Pr, Nd and Sm), $RCuGa_3$ (R = La, Pr, Nd and Gd) [3, 5] and $NdCuAl_3$ [6] possessing tetragonal $BaNiSn_3$-type structure.

TABLE I

Refinement results for $NdAgAl_3$.

a [Å]	c [Å]	V_{cell} [Å^3]	χ^2	R_p [%]	R_{wp} [%]
$NdAgAl_3$					
4.2740	11.1219	203.15	5	36	24
$NdAgAl_3$ ($BaNiSn_3$-type structure)					
atom	site	x	y	z	
Nd	2a	0.0000	0.0000	0.0000	
Ag	2a	0.0000	0.0000	0.6409	
Al1	2a	0.0000	0.0000	0.4218	
Al2	4b	0.0000	0.5000	0.2415	

The inverse magnetic susceptibility (χ^{-1}) of $NdAgAl_3$

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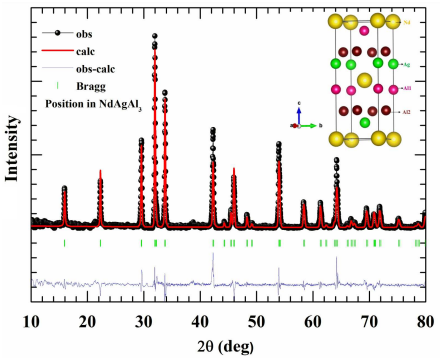

Fig. 1. Powder X-ray diffraction pattern of $NdAgAl_3$ along with Rietveld refinement using GSAS program. Inset shows the unit cell of $NdAgAl_3$ with the atoms represented by different colours and ball size.

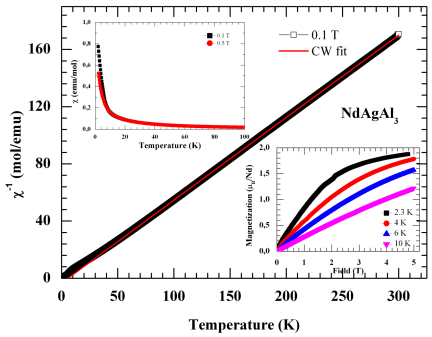

Fig. 2. Inverse magnetic susceptibility of $NdAgAl_3$. The upper inset shows susceptibility $\chi(T)$ in 0.1 and 0.5 T below 100 K. The lower inset shows magnetization versus applied field at selected temperatures.

measured as a function of temperature in the range 2–300 K in an applied field of 0.1 T, is shown in Fig. 2. The complete magnetic saturation has not been attained down to 2 K. It is obvious that measurements below 2 K are needed to state the precise type of magnetic ordering. The inverse magnetic susceptibility was fitted with modified Curie–Weiss law in the paramagnetic region (50–300 K), which can be defined as $\chi = \chi_0 + (C/(T - \Theta_P))$, where χ_0 is the temperature-independent term, attributed to the Pauli susceptibility and Van Vleck paramagnetism. Θ_P is the paramagnetic Curie temperature and C is the Curie constant which can be expressed in terms of the effective moment as $C = \mu_{\rm eff}^2/8$. The fit yields the values of $\chi_0 = 4.1\times10^{-5}$ emu/mole, $\Theta_P = 6$ K and $\mu_{\rm eff} = 3.7\ \mu_{\rm B}$/Nd. The value of effective magnetic moment is close to the theoretical value of Nd^{3+} (3.62 $\mu_{\rm B}$) free ion. Lower inset of Fig. 2 shows the isothermal magnetization $M(H)$ measured with respect to the field at different temperatures such as 2.3, 4, 6, and 10 K. The magnetic isotherm at 2 K is nonlinear at low fields and it lacks saturation at 5 T. Observed value of 1.9 $\mu_{\rm B}$/Nd is much lower than the expected value for Nd^{+3} ions (3.2 $\mu_{\rm B}$) but it is similar as for $NdCuAl_3$ [6]. This behaviour yields to the fact that the compound orders magnetically at temperatures less than 2 K. Comparing our results with that of isostructural $NdCuAl_3$ [6] one could see also the similarity of the observed values and behaviours of magnetic properties. Therefore, we could suppose the existence antiferromagnetic ordering below 2 K.

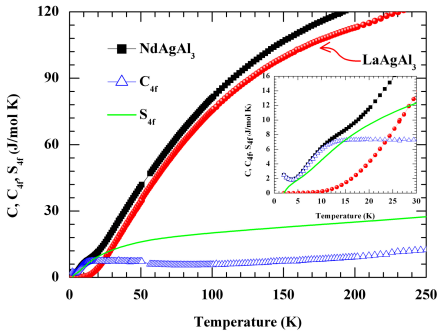

Fig. 3. Temperature dependent heat capacity of $NdAgAl_3$ between 2 and 250 K. C_{4f} and S_{4f} (see text) are also plotted. Inset shows the low-temperature range behaviour.

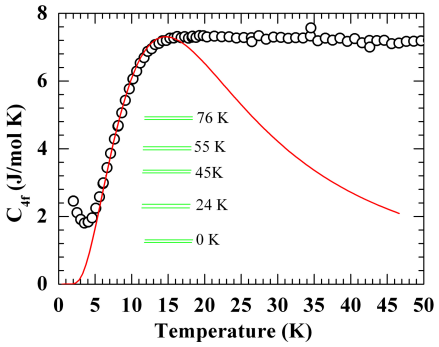

Fig. 4. The plot of C_{4f}/T versus T for $NdAgAl_3$ in various applied magnetic fields.

The temperature dependent heat capacity of $NdAgAl_3$ along with isostructural nonmagnetic reference system $LaAgAl_3$ (our sample [7]) is shown in Fig. 3. The zero field heat capacity data of $NdAgAl_3$ show the increasing dependence when approaching to 2 K, which is mainly due to the magnetic contribution to the heat capacity near to the magnetic ordering temperature and thereby confirms the expectation of magnetic ordering at less than 2 K. The magnetic $4f$ contribution to the heat capacity of the compound was deduced by subtracting the polycrystalline nonmagnetic analogue $LaAgAl_3$. The C_{4f} data shows a broad maximum centered well above ordering temperature (around 20 K — Fig. 5) indicating the significant Schottky anomaly contribution owing to CEF effect.

The magnetic entropy (S_{4f}) of $NdAgAl_3$ is estimated by integrating the C_{4f}/T vs. T dependence and it is shown in Fig. 3. The magnetic entropy (S_{4f}) is increasing linearly with temperature. Figure 4 shows the C_{4f}/T versus T dependences in different applied magnetic fields up to 9 T for $NdAgAl_3$. For higher magnetic fields, at 6 T, the Schottky type broad maximum appears at 4.5 K and then for 9 T it shifts to 7 K.

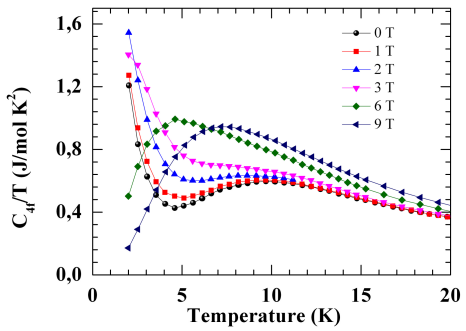

Fig. 5. Low temperature detail of dependence of the $4f$-derived heat capacity (C_{4f}). Solid line represents the fit to the expression (1) for the Schottky heat capacity.

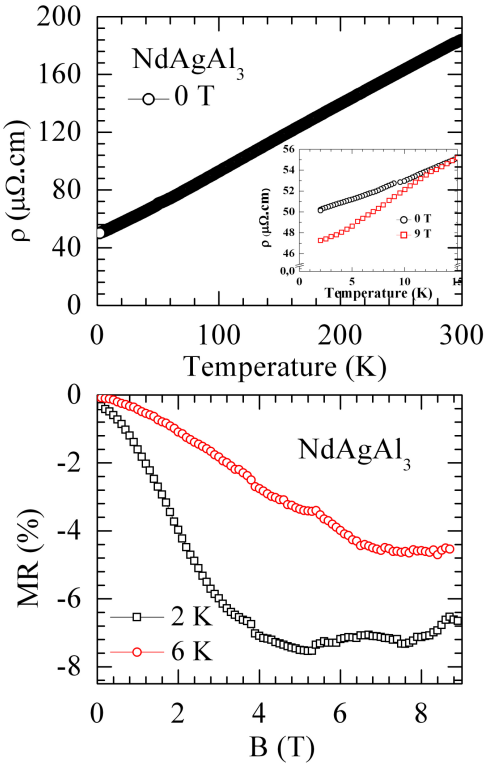

Fig. 6. (a) Electrical resistivity $\rho(T)$ of $NdAgAl_3$ measured in zero magnetic field. Insets shows the low temperature $\rho(T)$ versus T also for 9 T. (b) Magnetoresistance of $NdAgAl_3$ at 2 K and 6 K as function of magnetic fields up to 9 T.

This behaviour is probably due to the complex magnetic ordering. In Fig. 5 the low temperature detail of the Schottky heat capacity fit by well-known formula (e.g. [5]) is shown together with determined schematic energy levels diagram. The free ion Nd^{3+} ($J = 9/2$) is expected to yield to the tenfold degenerated ground state, which is split into five doublets. However, the best fit, which is giving $\Gamma_1 = 24$ K, $\Gamma_2 = 45$ K, $\Gamma_3 = 55$ K and $\Gamma_4 = 76$ K, is good only for low temperature part with maximum at 15 K.

Electrical resistivity of $NdAgAl_3$ compound was measured as a function of temperature from 2 K to 300 K in an applied field of 0 T and 9 T, as it is shown in Fig. 6. The compound exhibits normal metallic behaviour. The resistivity ratio (RR) determined from the relation $\rho(300\text{ K})/\rho(2\text{ K})$ yields to RR= 3.7, specifying the rather fair quality of the ternary sample. The resistivity of the compound is reduced below 10 K while applying high magnetic field due to the short-range correlation. Figure 6 shows the MR as a function of applied magnetic fields at two different temperatures such as 2 K and 6 K. The magnetoresistance (MR) can be defined as $\mathrm{MR}(T,H) = (\rho(T,H) - \rho(T,0))/\rho(T,0) \times 100\%$. $NdAgAl_3$ exhibits negative MR behaviour of about 7.5% at 2 K. This negative MR corresponds with behaviour of $C_{4f}(T)$ in the vicinity of 2 K, when magnetic field more than 4 T suppresses magnetic correlations.

4. Conclusion

Taking into account the observed magnetic, thermodynamic, and transport properties of $NdAgAl_3$ we suppose that below 2 K it will undergo to the magnetic ordering. We expect the antiferromagnetic ordering to be favored. Therefore, the low temperature measurements below 2 K are necessary to confirm the detailed magnetic behaviour. CEF was calculated from the Schottky heat capacity data.

Acknowledgments

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