Magnetic Phase Transitions in SmPd$_2$Al$_3$

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Abstract. We present results of specific heat, magnetization and AC susceptibility
measurements of SmPd$_2$Al$_3$ single crystal. The compound exhibits strong uniaxial
magnetocrystalline anisotropy with the easy-magnetization direction along the hexagonal $c$-
axis. A transition to a ferromagnetic ordering has been observed at $T_C = 12.4$ K followed by
several order-order magnetic phase transitions below 5 K. The magnetization curves measured
below 4 K exhibit a series of metamagnetic transitions in fields $< 1$ T, which indicates that the
AF ground state is gradually destroyed with applying a magnetic field along the $c$-axis.

1. Introduction

The localized 4f-electron magnetism of lanthanide ions is by rule characterized by stable magnetic
moments reflecting population of the energy levels of the ground state multiplet determined by the
total angular momentum $J$ and split by the crystal field (CF) interaction. Sm represents an exception of
the rule. The Sm magnetism is not governed by the ground state multiplet $J = 5/2$ only, since the first
(second) excited multiplet $J = 7/2$ ($J = 9/2$) is also populated at elevated temperatures because of
having a low energy of 0.2072 eV (0.4456 eV).

Since the Sm magnetic moment at low temperatures is small ($gJ = 0.71 \mu_B$ for the Sm$^{3+}$ free ion) the
exchange interactions can be very peculiar and several magnetic phases may appear in the same
material within different temperature and magnetic-field intervals, respectively. Here we study the
magnetic phase transitions in an SmPd$_2$Al$_3$ single crystal. We measured the magnetization ($M$), AC
susceptibility ($\chi_{AC}$) and specific heat ($C_p$) as a functions of temperature ($T$) and the external magnetic
field ($B$) applied along the principal crystallographic directions [100] and [001].

SmPd$_2$Al$_3$ as well as the REPd$_2$Al$_3$ (RE = La, Ce, Nd, Gd) counterparts adopt the same hexagonal
PrNi$_2$Al$_3$–type crystal structure [1]. The so far published results on SmPd$_2$Al$_3$ behavior have been
obtained by measuring only polycrystals and point to several magnetic phase transitions below 12 K
[1]. The specific-heat measurements of SmPd$_2$Al$_3$ have been reported revealing anomalies at 3, 6 and
12 K, respectively, indicating magnetic phase transitions [2]. Three magnetic phase transitions at 4,
4.3 and 12 K [1], respectively, have been proposed from susceptibility data. The $M(B)$ curves
measured at 2.5 K indicated magnetic phase transitions at $\sim$ 0.3 and 2 T, respectively [3].

2. Experimental details

The single crystal of SmPd$_2$Al$_3$ has been pulled in a triarc furnace by Czochralski method from a
stoichiometric melt. The single crystal was a 20-mm long cylinder of a diameter of 3 - 4 mm. The
crystal quality was checked by Laue technique. A small part of the crystal was pulverized and X-ray
powder diffraction (XRPD) data were recorded on a Seifert powder diffractometer equipped with a monochromator providing the Cu Kα radiation. The XRPD data were analyzed by means of the Rietveld profile procedure using the program FullProf. The crystal composition was checked by EDX analysis on a FE-SEM Tescan. Laue technique was also used for crystal orientation. The sample for the magnetization measurements had the form of a small beam (1x1x1.5 mm) with rectangular planes; two of them were oriented perpendicular to the crystallographic a- and c-axis, respectively. The specific-heat samples were small plates (1.5x1.5x0.2 mm) with the main planes perpendicular to the a- and c-axis, respectively.

The $C_p$, $M$ and $\chi_{AC}$ were measured using Quantum Design PPMS and the MPMS apparatuses, respectively; $C_p$ at temperatures from 1.8 to 300 K ($M$ in the temperature range from 1.8 to 400 K) in magnetic fields up to 9 T, $M$. The AC-susceptibility measurements were performed from 1.8 to 30 K in an AC magnetic field 0.03 mT of a frequency of 497 Hz.

3. Results and discussion

Structure and composition

We have successfully prepared a high-quality SmPd$_2$Al$_3$ single crystal as confirmed by Laue patterns. The XRPD data contained only the reflections corresponding to the hexagonal structure of the PrNi$_2$Al$_3$ type (space group P6/mmm) with the lattice parameters $a = 5.293$ Å, $c = 4.064$ Å, which are in good agreement with previously published data. The proper 1:2:3 stoichiometry of the grown crystal has been confirmed by EDX analysis within the accuracy of the method. No spurious phase has been located and no concentration gradients have been detected.

Specific heat

In zero magnetic field a sharp $\lambda$-shape anomaly in the $C_p(T)$ dependence peaking at 12.4 K and three much smaller peak-like anomalies located at 4.4, 3.9, 3.4 K, respectively (see Figure 1), have been observed. We denote these temperatures as the transition temperatures $T_C$, $T_1$, $T_2$, $T_3$, respectively. The 12.4-K anomaly is gradually enhanced with increasing magnetic field applied along c ($B//c$) but rests at nearly the same temperature. Although such evolution is rather unusual, the magnetization and resistivity data presented lower confirm that this specific heat anomaly is apparently associated with the onset of a ferromagnetic ordering in SmPd$_2$Al$_3$ at $T_C$ 12.4 K. The low-temperature anomalies, which are smeared out in fields $B//c$ higher than 1 T, are presumably associated to the order-order magnetic phase transitions. The 12.4-K peak remains nearly intact by the magnetic field up to 9 T applied along the a-axis (not shown in the Figure) that indicates strong magnetocrystalline anisotropy.

Figure 1. Temperature dependence of the specific heat measured in various magnetic fields applied parallel to c-axis. The inset shows details of the low-temperature anomalies.

Figure 2. Temperature dependence of the AC susceptibility measured with the excitation field applied along the a- and c-axis. The inset shows the low-temperature detail and critical temperatures are pointed by the arrows.
The low-temperature anomalies become continuously smeared out with increasing the field $B//a$ (not shown in the Figure). This indicates that the low temperature phases ($T < T_1$) have nonzero $AF$ components in the basal plane of the hexagonal structure. For comparing of our results with existing literature data on SmPd$_2$Al$_3$ we have collected together the so far existing information about critical temperatures of the anomalies of the $C_p$, $M$ and $\chi_{AC}$ in Table 1.

Table 1. List and comparison of published critical temperatures of the anomalies in the specific heat, $AC$ susceptibility, low-field magnetization and electrical resistivity data with our experimental results.

| Property | Source       | Anomaly location (K) |
|----------|--------------|----------------------|
| $C_p$    | [this work]  | $T_C$ $T_1$ $T_2$ $T_3$ |
| $C_p$    | [1]          | 12.5 4.3 4 --- |
| $C_p$    | [4]          | 12.5 4.7 --- 3.7 |
| $\chi_{AC}$ | [this work] | 12.5 4.5 --- 3.7/3.4 |
| $M$      | [this work]  | 12.3 4.4 --- 3.4 |
| $M$      | [3]          | 12.5 --- 4 3.3 |
| $M$      | [1]          | 12.5 4.3 4 --- |

$AC$ susceptibility and magnetization

The $\chi_{AC}$ vs. $T$ plots for the $AC$ magnetic field applied along the $a$- and $c$-axis, respectively, are shown in Figure 2. The $a$-axis data are characterized by a very low nearly temperature independent signal free of any considerable anomaly exceeding signal noise, whereas the curve measured in the $AC$ field applied along the $c$-axis exhibits three anomalies presumably indicating magnetic phase transitions. When cooling, the $c$-axis data exhibit a sudden upturn commencing at $T_C = 12.4$ K, a shoulder at 4.5 K and a sharp peak at 3.7 K. The latter two features may be tentatively associated with the magnetic phase transitions at $T_1$ and $T_2$, respectively, that implies from comparing data in Table 1. Theory [5], however, says that $T_N$ is the temperature where is the maximum of the specific heat and the maximum in $\partial(\chi T)/\partial T$ vs. $T$, which frequently does not coincide with the maximum in $\chi$ vs. $T$ in real systems. That is also in our case where the maximum in $\partial(\chi T)/\partial T$ vs. $T$ is at 3.4 K, which matches the $T_3$ value determined from specific-heat data.

Figure 3. Hysteresis loops measured at temperature 1.8 K with applied magnetic field parallel and perpendicular on the basal plane.

Figure 4. Figure shows virgin curves of SmPd$_2$Al$_3$ measured along the easy axis magnetization at various temperatures.

The dramatic difference between the magnetization curves measured at 1.8 K in the magnetic field applied along the $a$ and $c$-axis, respectively, seen in Figure 3 corroborates the conclusion about the
uniaxial anisotropy. Whereas the magnetic moment in the field $B_{/\parallel c}$, which saturates above 2 T, is dominant a very weak paramagnetic-like linear (in higher fields) response is observed for $B_{/\parallel a}$. The slight non-linearity and hysteresis observed in the latter case in fields $< 2$ T is considered rather as a result of imperfect geometry of experiment than the intrinsic hard-axis behavior. The $c$-axis magnetic moment of 0.19 $\mu_B$/f.u. at 1.8 K observed in the field of 7 T is nearly 5 times lower than the expected ordered moment for the ground-state multiplet of the free Sm$^{3+}$ ion ($gJ\mu_B = 0.71 \mu_B$), which is presumably responsible for the entire magnetic behavior of SmPd$_2$Al$_3$. The strange shape of the $c$-axis curve in fields weaker than 2 T is fully reproducible and is apparently connected with the specific features of samarium magnetism in SmPd$_2$Al$_3$.

The detailed view of the magnetization loops for $B_{/\parallel c}$ is presented in Figure 4. A series of four possible metamagnetic-like transitions can be identified on the 1.8-K virgin magnetization curve. The onsets of transitions denoted by arrows can be found at 0.03, 0.35, 0.5 and 0.75 T, respectively. The transitions at lower fields can be traced still on the curve measured at 2.5 K, but they are gone above 3.4 K. The magnetization loops measured at 1.8 and 2.5 K exhibit not only some metamagnetic-like features but also pronounced hysteresis. All these features vanish at temperatures above 3.4 K. This implies that the ground-state phase, which forms below $T_3$ is antiferromagnetic and apparently rather complicated. The magnetization curves measured at $T_3 < T < T_C$, however, still show strong tendency to saturate as usually found in ferromagnets.

4. Conclusions

We have prepared a single crystal of SmPd$_2$Al$_3$ compound and studied magnetic phase transitions by measuring of $C_p$, $M$ and $\chi_{AC}$ in magnetic field applied along the principal crystallographic axes. The experiments reveal the strong uniaxial anisotropy and four magnetic phase transitions on the $C_p(T)$ at $T_3 = 3.4$ K, $T_2 = 3.9$ K, $T_1 = 4.4$ K and $T_C = 12.4$ K, respectively, and detected in part also on the $M(T)$ and $\chi_{AC}(T)$ curves, respectively. Our results point to a complex magnetic phase diagram. Although this material becomes ferromagnetic below $T_C = 12.4$ K an antiferromagnetic ground state seems to establish at low temperatures. The series of four metamagnetic transitions detected at 0.03, 0.35, 0.5 and 0.75 T, respectively, underlines the complexity of the Sm magnetism, which is characterized by a small Sm magnetic moment and interplay of the crystal field and exchange interactions.

To prove our scenario on the multiphase magnetic diagram as well as on the magnetic phase transitions suitable microscopic experiments are strongly desired; namely neutron diffraction and $\mu$SR spectroscopy. The high neutron absorption by Sm for the standard thermal neutron wavelength implies hot neutrons inevitable for a successful neutron diffraction experiment.

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