Metamagnetic behaviour of Nd$_3$Pt$_{23}$Si$_{11}$

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ABSTRACT

In the course of studying the R$_3$Pt$_{23}$Si$_{11}$ series (R=rare earth), we have recently synthesized Nd$_3$Pt$_{23}$Si$_{11}$. It crystallizes in a face-centered cubic structure (*Fm-3m* space group) with a lattice parameter of 16.8493 (4) Å at room temperature. Heat capacity and magnetic measurements show the existence of a magnetic transition at 1.6 K. Measurements of the susceptibility and magnetization down to 95 mK reveal metamagnetic behavior confirming antiferromagnetic ordering. Powder neutron diffraction experiments show that the magnetic propagation vector is k=[000] and that the 6 neodymium moments in the unit cell are antiferromagnetically coupled in pairs.

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1. Introduction

Investigation of the R–Pt–Si (R=rare earth element) ternary phase diagram was mainly driven by the discovery of novel ternary compounds with peculiar physical behavior. The Ce–Pt–Si phase diagram has triggered a lot of interest [1,2] because some cerium-based compounds present interesting physical properties such as unconventional heavy fermion superconductivity in CePt$_3$Si [3], heavy-fermion properties in CePtSi [4], Kondo lattice behavior in CePtSi$_2$ and CePt$_2$Si$_2$ [5,6] and most recently, the observation of unusual ferromagnetic ordering in Ce$_3$Pt$_{23}$Si$_{11}$ [7–9]. The compound Yb$_3$Pt$_{23}$Si$_{11}$ has been recently synthesized and was found to be a moderately enhanced paramagnetic compound with a fairly unstable 4f electronic shell [10]. With the aim of finding novel compounds with remarkable magnetic properties, we have successfully synthesized new compounds in this series. In this paper, we report investigations on the magnetic behavior of the compound Nd$_3$Pt$_{23}$Si$_{11}$ at very low temperatures.

2. Experimental procedure

High quality polycrystalline samples of Nd$_3$Pt$_{23}$Si$_{11}$ have been prepared in an induction furnace. Stoichiometric proportions of the different constituents; Nd (99.9%, Johnson Matthey), Pt (99.95%, Alfa Aesar) and Si (99.9999%, Alfa Aesar), were melted in a cold copper crucible under a highly-purified argon atmosphere. The samples were melted several times to improve the homogeneity. Mass losses during this first step were less than 0.5% and no further heat treatment was applied.

Sample structure and quality were checked by conventional X-ray powder diffraction (Cu-Kα radiation on a Philips PW1730 diffractometer), whilst structural refinements were performed using the FullProf program [11]. Nd$_3$Pt$_{23}$Si$_{11}$ crystallizes in the same fcc structure (*Fm-3m* space group) as Ce$_3$Pt$_{23}$Si$_{11}$ [12]. At room temperature, the value of the refined lattice parameter is 16.8493(4) Å. More details on the crystallographic analysis can be found in Ref. [9].

Magnetization measurements were performed on an almost spherical sample of diameter 2.5 mm using different magnetometers based on the extraction method. In the 1.8–300 K temperature range, we used a commercial Quantum Design MPMS magnetometer, and a second experimental setup, equipped with resistive detection coils and a superconducting magnet, which can supply magnetic fields of ± 10.5 T. For temperatures down to 65 mK, we used a SQUID magnetometer equipped with a miniature dilution refrigerator developed at the Institut Néel.

In the 0.38–300 K temperature range, the heat capacity was measured using the relaxation method with a Quantum Design PPMS.

Powder neutron diffraction experiments were carried out at the Institut Laue-Langevin (ILL) in Grenoble on the CRG (CNRS Grenoble/CSIC Spain)—two-axis diffractometer D1B using a wavelength of 2.52 Å, selected by a pyrolytic graphite (002). The powdered sample was installed inside a ^{3}He cryostat capable of reaching a base temperature of 400 mK.

3. Magnetic and thermodynamic properties

The inverse of the magnetic susceptibility is deduced from the isothermal magnetization curves in the temperature range 1.8–300 K. From room temperature down to 25 K, the inverse susceptibility varies linearly with temperature, in agreement with the Curie–Weiss law $\chi = C/(T-\theta_p)$, where $C = \mu_{eff}^2/3k_B$ is the theoretical Curie constant and $\mu_{eff} = g_J\mu_B[J(J+1)]^{1/2}$, the effective magnetic

moment of the rare earth ion. Setting the theoretical value of the effective moment of the Nd^{3+} ion to be $\mu_{eff} = 3.62 \mu_B$, the experimental thermal variation of the inverse susceptibility is very reproducible, adjusting the value of the paramagnetic Curie temperature θ_p to 15 ± 0.5 K, as shown by the straight line in Fig. 1. A negative value for θ_p suggests antiferromagnetic correlations between the rare earth ions.

Below 25 K, $1/\chi$ deviates from the Curie–Weiss law and is characteristic of crystal electric field effects (CEF). CEF interactions partially lift the degeneracy of the fundamental multiplet $J = 9/2$, leading to a reduction of the theoretical magnetic moment, $\mu = g_J \mu_B J = 3.27 \mu_B$.

Magnetization curves, in the temperature range 1.8–3 K are shown in Fig. 2. The Arrott plots of the curves at 1.8 and 2 K (inset in Fig. 2) intercept the x-axis at a non-zero value, demonstrating that $Nd_3Pt_{23}Si_{11}$ remains paramagnetic down to 1.8 K. Under high fields, the magnetization tends to a maximum value of $1.5 \mu_B$. This value is a factor of two smaller than the theoretical saturated moment $3.27 \mu_B$, strongly suggesting the reduction of the magnetic moment by the CEF interactions.

Specific heat measurements, displayed in Fig. 3, show that a transition occurs at 1.6 K. The steep initial slope of the magnetization at 1.8 K points to ferromagnetic-type behavior, in an apparent
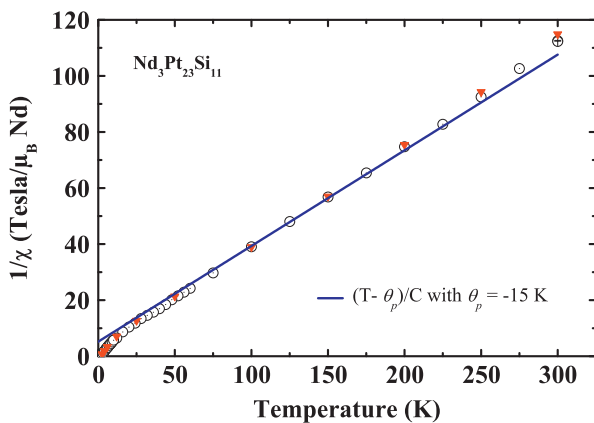
Fig. 1. Thermal variation of the inverse susceptibility of $Nd_3Pt_{23}Si_{11}$. The open dots represent the susceptibility deduced from measurements in magnetic fields up to 8 T using the in-house magnetometer with resistive detection coils. The full triangles represent the susceptibility deduced from the measurements using the commercial Quantum Design MPMS magnetometer. The straight line corresponds to the Curie–Weiss law calculated with the theoretical values of the effective moment of the Nd^{3+} ion.
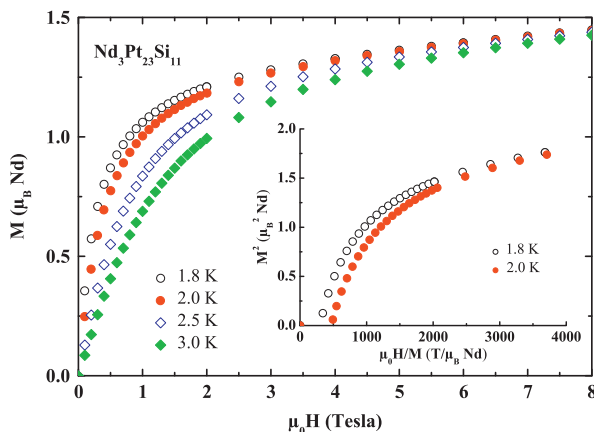
Fig. 2. The field dependence of the magnetization between 1.8 and 3 K. The inset shows Arott's plots of the magnetization at 1.8 and 2 K.

contradiction with the negative value found for θ_p. The entropy associated with the lambda anomaly of the specific heat is close to R $ln(2)$, thus confirming a doublet CEF ground state for the Nd^{3+} ions. In order to gain further insight as to the nature of this transition, magnetic susceptibility measurements were performed down to 95 mK. As shown in Fig. 4(a), one observes a sharp peak at $T^* = 1.6$ K in the susceptibility, which is characteristic of a magnetic phase transition. The value of T^* is in very good agreement with the transition temperature observed in the heat capacity measurements. At first glance, this transition looks ferromagnetic because of the steep increase of χ when T^* tends to 1.6 K in the paramagnetic phase, and the positive intercept of $1/\chi$ with the temperature axis $\theta_p^{LT} = +1.5$ K (see Fig. 4(b)). However, the maximum value of $\chi = 0.035$ emu/cm^3 is one order of magnitude less than the expected value at T_C for a ferromagnet, $\chi_{max} = 1/N$, where N is the demagnetizing factor. For a spherical sample, which is almost the case here, $N = 3$, leading to $\chi_{max} = 1/3$ emu/cm^3.

The magnetization curves measured at 95 mK and 1.8 K using the setup with the ^{3}He dilution in fields up to 8 T are shown in Fig. 5 (a) and compared with the one measured at 1.8 K using the setup with the ^{4}He cryostat. It can be seen that the magnetization curves at 1.8 K are in full agreement. Above 4 T, the curves at 95 mK and 1.8 K superpose and reach the same value of $1.45 \mu_B$ at 8 T. The Arrott plot at 95 mK (inset Fig. 5(a)) is consistent with antiferromagnetic behavior. Fig. 5(b) shows the magnetization loop measured in the low field region at 95 mK. At 0.1 T, a magnetization jump is observed. In the loop at 1.3 K (not shown here) a similar jump is observed at 0.08 T, highlighting the metamagnetic behavior in the ordered phase. A non-negligible hysteresis is associated to the metamagnetic transition. The fact that this transition occurs under very low fields suggests a weak magnetic anisotropy. We also observe a very small remanent magnetization of $\pm 0.02 \mu_B$, which could be reasonably ascribed to a small amount of ferromagnetic impurities. All of this leads us to the conclusion that the transition at T^* is antiferromagnetic.

4. Powder neutron diffraction studies

In order to check the magnetic arrangement in the ordered phase, powder neutron diffraction studies have been performed down to 400 mK. To ensure good thermal coupling at these very low temperatures, it is necessary to use a sealed copper sample holder with a low He pressure. Therefore, in the collected diffraction patterns, two very intense peaks due to the copper diffraction are present at $2\theta = 74.49°$ and $88.69°$. Due to the Q-dependence of the magnetic form factor of the rare earth ions, the stronger magnetic signal is observed in the low 2θ region. Also, since the CEF effects reduce the magnetic moment of the Nd ions, one expects a small magnetic contribution to the powder diffraction pattern. This makes the powder data analysis rather difficult. In Fig. 6(a), we compare the diffraction patterns collected at 400 mK and 4 K. In both patterns, some peaks cannot be indexed in the crystallographic structure and we attribute them to impurity phases. At 400 mK, we observe no extra peaks at the positions forbidden by the crystallographic structure. Thus, as in $Ce_3Pt_{23}Si_{11}$ [8], the fcc lattice translations are also translations for the magnetic structure in $Nd_3Pt_{23}Si_{11}$ and the magnetic propagation vector is then $k = [000]$. Only the intensity of the (311) and (331) reflections increases significantly in the ordered phase, as illustrated in Fig. 6(a). The pattern difference (Fig. 6(b)), reveals a tiny intensity for the peaks with even Miller indices, however the intensity is less than the experimental uncertainty. Fig. 7 clearly shows that the intensity at the (311) position increases while the temperature is lowered, confirming a magnetic contribution. However, the thermal evolution of the (222) reflection is less obvious.

The Nd ions occupy the 24d sites of the $Fm-3m$ space group. They are located at the positions: Nd_1 (0.25,0.25,0), Nd_2 (0.25,−0.25,0), Nd_3