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To cite this version:


HAL Id: jpa-00228352
https://hal.archives-ouvertes.fr/jpa-00228352
Submitted on 1 Jan 1988

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ANTIFERROMAGNETIC PHASES IN NdCo$_2$Si$_2$

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Abstract. - Magnetic characteristics of NdCo$_2$Si$_2$ have been studied by magnetic and neutron diffraction measurements using a single crystal. Three antiferromagnetic phases were found below 32 K. A simple collinear antiferromagnetic phase is stable in the range 0-15 K where a metamagnetic process is observed. Square-wave structures with the propagation vectors $\mathbf{K} = (0, 0, 0.928)$ and $(0, 0, 0.785)$ appear for $15 K < T < 24 K$ and $24 K < T < 32 K$ (= $T_N$), respectively.

Ternary compounds RCo$_2$Si$_2$ (R = rare earth) crystallize in the tetragonal ThCr$_2$Si$_2$ type structure, in which the same atoms lie in alternate layers stacked along the c-axis with a sequence of RSiCoSi... Most of RCo$_2$Si$_2$ compounds have been reported to be antiferromagnetic [1-3]. Their magnetic characteristics are very interesting in connection with the layer structure. Recently, a metamagnetic magnetization process and appearance of successive magnetic transitions in PrCo$_2$Si$_2$ have been found [4]. The magnetic behaviour is similar to that obtained for the Ising system with competing exchange interactions [5]. In the present study, magnetic and neutron diffraction measurements on the single crystal NdCo$_2$Si$_2$ have been carried out.

The temperature dependence of magnetic susceptibilities $\chi_c$ and $\chi_a$ along the c- and a-axes is shown in figure 1. Three anomalies were observed at $T_1 = 15$ K, $T_2 = 24$ K and $T_N = 32$ K in $\chi_c$ vs. T curve. The rapid changes in $\chi_c$ at $T_1$ and $T_2$ suggest that the transitions are of first order. On the other hand, $\chi_a$ is almost temperature independent below $T_N$. The compound exhibits large magnetic anisotropy, and it is believed to be due to the crystalline electric field effects.

Figure 2 shows the magnetization curves along the c-axis at various temperatures. The magnetization in the c-plane is linear against applied field and very small. The value at 4.2 K is 0.1 $\mu_B$/f.u. at 54 kOe (Fig. 2a). The easy direction of magnetization is the c-axis. There is no magnetic anisotropy in the c-plane. At 4.2 K, a steplike magnetization process is seen along the c-axis; the magnetization increases rapidly around $H_{c1} = 40$ kOe and $H_{c2} = 58$ kOe with increasing field. The large hysteresis loops observed indicate that the transitions are of first order. The magnetization at the maximum field in this study reaches to 0.66 $\mu_B$/f.u. Leciejewicz et al. [3] have reported from neutron diffraction studies that only Nd atom is magnetic and has nearly theoretical Nd$^{3+}$ moment, 3.27 $\mu_B$, at 4.2 K. The magnetization obtained at the maximum field is much smaller than the full Nd moment. The spin configuration does not reach a ferromagnetic arrangement. Therefore, additional

![Fig. 1. - Temperature dependence of magnetic susceptibilities along the c- and a-axis of the NdCo$_2$Si$_2$ single crystal.](image)

![Fig. 2. - Magnetization curves along the c-axis of the NdCo$_2$Si$_2$ single crystal at (a) 4.2 K, (b) 16 K, (c) 20 K and (d) 35 K. Magnetization in the c-plane is only shown in (a).](image)
steplike increases in magnetization should appear at higher fields. The magnetizations just after the first and second abrupt increases are nearly equal to one fourteenth and one fifth of the full moment, respectively. Therefore, small fractions of Nd moments are reversed at the fields. The magnetization process along the c-axis changes drastically with temperature. For $T_1 < T < T_2$, the transition at $H_{c1}$ disappears, but the step at $H_{c2}$ remains. The magnetization at 16 K is given in figure 2b as an example. For $T_2 < T < T_N$, there is no more transition within this experimental field up to 60 kOe, but a precursor of metamagnetic transition is seen at high field (Fig. 2c). For $T > T_N$, the magnetization curve becomes linear (Fig. 2d).

Neutron diffraction measurements on the single crystal have been performed at the Research Reactor Institute, Kyoto University. Antiferromagnetic superlattice peaks were observed on lattice rows parallel to the $c^*$-axis, but not on the $c^*$-axis below $T_N$. This fact suggests that the magnetic moments lie along the c-axis and the propagation vector is given as $k = (0, 0, k) \ [2 \pi /c]$ for each phase: magnetic structures consist of (001) ferromagnetic planes with moments modulated along the c-axis. The thermal variation of integrated intensity for the $(1 0 1 - k)$ magnetic superlattice peaks is shown in figure 3. As evidenced from the figure, there are three antiferromagnetic phases. Below $T_1 = 15$ K, a $k = 1$ phase is stable in which the magnetic unit cell is in accordance with the chemical one and the sequence of ferromagnetic planes is $+ - + -$. At $T_1$, the $k = 1$ phase disappears and a $k = 0.928$ phase appears and is stable up to $T_2 = 24$ K. In this range, additional small superlattice peaks which correspond to the third harmonics were observed, indicating that the magnetic structure is a square-wave structure. A phase with $k = 0.785$ develops at $T_2$ and persists up to $T_N = 32$ K. Within the experimental accuracy, the magnitude of the propagation vectors may be considered as fraction numbers as $k = 13/14 \simeq 0.928$ and $k = 11/14 \simeq 0.785$. Therefore, it should particularly be mentioned that the magnetic cell may be fourteen times as large as the chemical unit cell for both phases. The magnetic behaviour of NdCo$_2$Si$_2$ is similar to that obtained for the Ising systems with competing exchange interactions [5].

Measurements of the magnetization at high fields and neutron diffraction under magnetic fields are now in progress for analysis of the magnetization process and for discussion on the possibility of the successive magnetic transitions.

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Fig. 3. – Thermal variation of integral intensity of the $(1 0 1 - k)$ magnetic superlattice peaks of NdCo$_2$Si$_2$.

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