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High-field magnetization studies of $NdNi_5$

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Abstract

Magnetization studies on single-crystalline $NdNi_5$ in external fields up to 35 T are reported. $NdNi_5$ is a ferromagnet with a T_C of 7.2 K and the easy magnetic direction is along the a direction. The spontaneous magnetic moment is $2.1\mu_B$/f.u. Along the c-axis, the magnetization amounts to $1.65\mu_B$/f.u. at 35 T. The magnetization process is analyzed within the crystalline-electric-field approach in combination with exchange interactions, taking into account the contribution of the Ni sublattice. The studies confirm that the charge-formed ground state of the f^3 (Nd^{3+}) subsystem is the state Γ_8 with a dominant $|\pm 5/2\rangle$ contribution. The full magnetization curves, up to 300 T, have been calculated. The magnetization curve along the c-axis proceeds to saturation with a metamagnetic-like transition at 150 T.

Keywords: Anisotropy; Intermetallics; Magnetization

The hexagonal compound $NdNi_5$ orders ferromagnetically below 7.2 K. The electronic and magnetic properties are well described within the crystalline-electric-field (CEF) approach in combination with exchange interactions, taking into account the contribution from the Ni sublattice [1,2]. In this paper, high-field magnetization studies on single-crystalline $NdNi_5$ samples are reported and discussed.

Magnetization measurements were performed on single-crystalline $NdNi_5$ at 1.5 K along the three principal directions of the hexagonal cell in magnetic fields up to 35 T at the Amsterdam High-Field Installation. The single crystalline sample was grown by the tri-arc Czochralski method in a purified argon atmosphere.

The high-field magnetization curves are shown in Fig. 1. The data at low fields coincide well with those reported in Ref. [1]. The Nd ion moment favours the a-axis within the hexagonal plane. The spontaneous moment at 1.5 K is $2.1\mu_B$/f.u., which amounts to 60% of the full trivalent neodymium moment. This reduction of the Nd moment is due to the crystal field effect [1,2]. The easy-axis magnetization at 35 T amounts to $3.3\mu_B$/f.u., indicating the attainment of the full Nd moment. A large anisotropy of the Nd magnetic moment is observed. Along the hard axis, i.e. the c-axis, the magnetization at 35 T reaches only $1.65\mu_B$/f.u. Moreover, there is a difference between the magnetization along the a and b directions which increases with increasing field ($0.3\mu_B$/f.u. in 35 T). The magnetization process is analyzed using the two-sublattice model by considering the contribution from both Nd and Ni sublattices [3]:

$$M_T = M_R + M_{Ni}. \quad (1)$$

The Ni moment (M_{Ni}) is fully induced by external fields B_o and by the presence of the Nd moment (M_R):

$$M_{Ni} = \chi_{Ni} B_o + \alpha M_R, \quad (2)$$

where $\alpha = \chi_{Ni} n_{RNi}$, with χ_{Ni} the temperature-independent susceptibility of the Ni sublattice. n_{RNi}, n_{RR} and

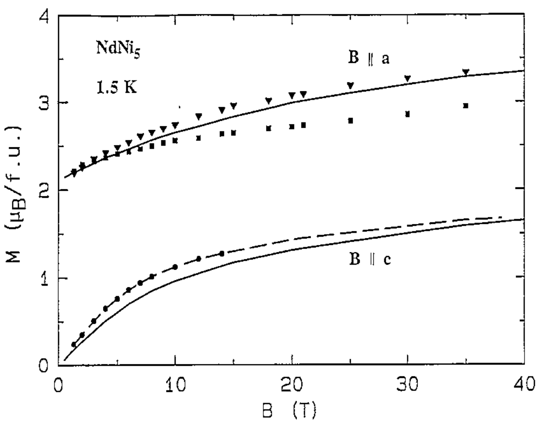

Fig. 1. High-field magnetization curves at 1.5 K for $NdNi_5$ measured along the a-axis (▼), the b-axis (■) and the c-axis (●, dashed line: in a semi-continuous field). The full lines are the calculated curves taking into account the Ni contribution.

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n_{NiNi} denote respectively the inter-sublattice and intra-sublattice molecular field coefficients.

The Nd magnetic moment M_R is calculated by considering the following Hamiltonian of the Nd^{3+} ion:

$$H_R = \sum\sum B_n^m O_n^m + g\mu_B \boldsymbol{J}(-ng\mu_B\langle J\rangle + \boldsymbol{B}_o). \qquad (3)$$

The first term is the CEF Hamiltonian for the lowest multiplet $^4I_{9/2}$ ($J = 9/2$, $S = 3/2$, $L = 6$, the Landé factor $g = 8/11$). It contains four parameters B_n^m for the hexagonal symmetry. The second term includes the Zeeman effect of the external field and the exchange interactions between the Nd ions. n is the molecular-field (MF) coefficient, $n = n_{RR} + \alpha n_{RNi}$.

The full set of CEF parameters, $B_2^0 = +3.35$ K, $B_4^0 = +14.5$ mK, $B_6^0 = -0.35$ mK and $B_6^6 = -13.5$ mK, $\chi_{Ni} = 3.9 \times 10^{-3} \mu_B/T$, $\alpha = 0.05$, $n = 3.5$ T f.u./μ_B, have been used for $NdNi_5$ giving a consistent description of the temperature dependence of the specific heat [1,2], of the magnetization, and of the paramagnetic susceptibility [1]. Our calculated magnetization curves (Fig. 1, full lines) provide overall agreement with the experimental data up to 35 T. The full magnetization curves calculated up to 300 T are shown in Fig. 2. The magnetization increases slightly along the a-axis, whereas along the c-axis it proceeds to saturation with a metamagnetic-like transition at 150 T. In zero field, the Nd moment is $2.1\mu_B$ and lies along the a-axis. With applied field along the c-axis, the Nd moments are slowly rotated to the field direction. At 15 T, the moments still make an angle of 29° with the field. Above 250 T, the moments become fully parallel to the field direction. For the Nd^{3+} ion, the ground state multiplet $^4I_{9/2}$ is split into five doublets. The energy level scheme resulting from the above CEF parameters for $NdNi_5$, shown in Fig. 3, is in good agreement with inelastic-neutron-scattering (INS) observations [4]. The charge-formed ground state Γ_8 with the dominant $|\pm 5/2\rangle$ state characterized by $\langle J_x\rangle = \pm 1.66$ and $\langle J_z\rangle = 1.17$ results from higher-order CEF interactions. The substantial reduction of the local magnetic moment and the large magnetocrystalline anisotropy in this compound are caused by the anisotropic charge distribution in the vicinity of the f-shell electrons. The influence of the magnetic field applied along the c-axis on the energy level scheme is shown in Fig. 3. The metamagnetic transition around 150 T is related to the crossing of energy levels. An applied field of 250 T transfers the ground state from the dominate $|\pm 5/2\rangle$ state to the $|-9/2\rangle$ state with the maximum value of the Nd moment.

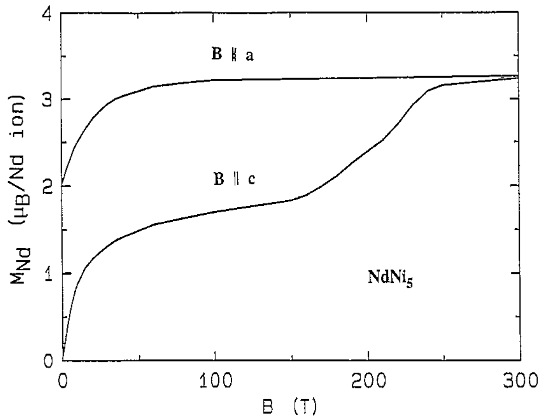

Fig. 2. Calculated field dependences of the Nd moment up to 300 T. A metamagnetic-like transition is found along the c-axis at 150 T.

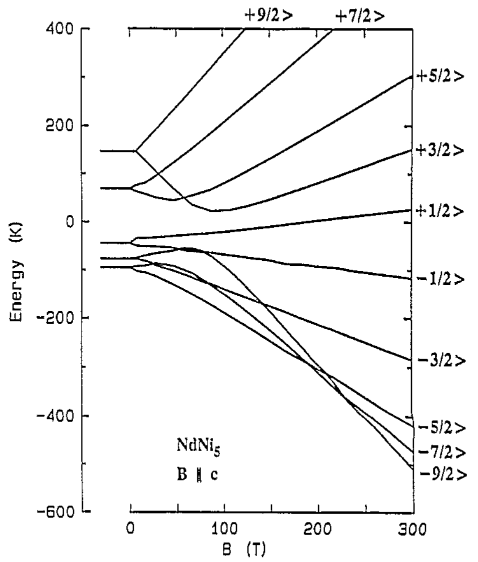

Fig. 3. Field variation of the energy level scheme of $NdNi_5$ for fields along the c-axis. Above 250 T, the $|-9/2\rangle$ state with the maximum value of the Nd moment becomes the ground state.

In summary, the magnetization curves of $NdNi_5$ are well described by the CEF and MF parameters up to 35 T. The easy direction of magnetization is the a-axis. A large magnetic anisotropy is observed between the a and b axes which increases with increasing field. The studies confirm that the charge-formed ground state of the f^3 (Nd^{3+}) subsystem is the state Γ_8 with a dominant $|\pm 5/2\rangle$ contribution, confirming the important role of the higher-order CEF terms for $NdNi_5$.

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