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High field magnetization of a NdCu₂ single crystal

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Abstract

Studies of the high-field magnetization on a single crystal of NdCu₂ with the field along the three principal crystallographic directions are presented. A crossing of the magnetization curves along the *a*- and *b*-axes is observed. The order of magnetization along each axis in high magnetic fields is then ($\mu_c < \mu_b < \mu_a$). This behaviour is discussed in terms of a microscopic hamiltonian including crystal-field and Zeeman interactions.

The intermetallic compound NdCu₂ crystallizes in the orthorhombic structure of the CeCu₂ type with space group D_{2h}⁸ (Imma) and exhibits antiferromagnetic order below $T_N = 6.3$ K (e.g. Refs. [1, 2]). Inelastic neutron spectroscopy has revealed that the orthorhombic crystal field splits the ground state multiplet ⁴I_{9/2} of Nd³⁺ into five Kramers doublets. The energy of the first excited doublet is $\Delta_1 = 33$ K [3]. We have shown that at sufficiently low temperatures and in low magnetic fields, where only the ground-state doublet is populated, the magnetocrystalline anisotropy in NdCu₂ is determined by the wavefunction of this doublet [2]. In high magnetic fields, however, the full crystal-field hamiltonian should be used for the description of the magnetization.

We studied two single crystals of NdCu₂. The first one has been prepared by remelting and slow cooling of a polycrystalline sample in a resistance furnace at Ural

State University. A single-crystalline grain of dimensions $2 \times 2 \times 1.5$ mm³ has then been extracted from the ingot. The second crystal has been grown by the Czochralski method at the University of Tsukuba. The resulting high-quality single crystal displayed the resistance ratio $\rho_{300\text{ K}}/\rho_{1.5\text{ K}} = 85$.

The temperature dependencies of the magnetic susceptibility along the principal crystallographic directions (Fig. 1) show that the anisotropy of the magnetization in the range around room temperature ($\mu_b < \mu_c < \mu_a$) differs from the low-temperature range ($\mu_c < \mu_a < \mu_b$). This behaviour can be well described using the full set of crystal-field parameters given in Ref. [3], obtained from the analysis of inelastic neutron scattering. Agreement between experiment and calculations of the temperatures of crossing of the susceptibility curves can be taken as a sensitive validity test of the set of crystal-field parameters in our microscopic hamiltonian

$$\mathcal{H} = \mathcal{H}_{\text{CF}} - g_J \mu_B \mathbf{H} \cdot \mathbf{J}, \quad (1)$$

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Fig. 1. Magnetic susceptibility of NdCu₂ along the principal crystallographic directions. Full lines represent the calculated susceptibility according to the crystal-field parameters given in the text.

Fig. 2. Magnetization of a NdCu₂ single crystal measured in quasi-static magnetic fields up to 28 T together with the calculated magnetization curves (full lines).

where $\mathcal{H}_{CF}$ is the crystal-field hamiltonian of the required orthorhombic symmetry [3], g_J is the Landé factor and J is the total angular momentum. Simultaneously, the calculations based on Eq. (1) suggest crossing of the magnetic isotherms in high magnetic fields.

Here, we present studies of the high-field magnetization of a single crystal of NdCu₂ for the three principal

Fig. 3. Magnetization curves of NdCu₂ in static magnetic fields up to 18 T. The full lines are the curves calculated by using Eq. (2) (see text).

crystallographic directions in comparison with the calculated magnetization. The magnetization isotherms were measured in quasi-static magnetic fields up to 28 T at 1.4 and 4.2 K (Fig. 2) in the High Field Installation of the University of Amsterdam and in static magnetic field up to 18 T (Fig. 3) in the National High Magnetic Field Laboratory in Los Alamos. While the magnetization curve along the *b*-axis on the first crystal showed only two metamagnetic transitions below 4 T at 2 K, three sharp metamagnetic transitions are visible for the new crystal, confirming its high quality [4]. In fields above the third transition, the magnetic moment along the *b*-axis is higher than that along the *a*- and *c*-axes. With increasing further field the $\mu_b(B)$ curve gradually saturates, at 1.4 K reaching about $2.3\mu_B/\text{f.u.}$ in 28 T in the case of the first crystal.

No metamagnetic transitions are observed along the *a*- and *c*-axes. The *a*-axis magnetization curve has a weak S-shape, saturates much slower and crosses the *b*-axis curve. The *c*-axis magnetization has the slowest saturation tendency in high fields. Thus the order of magnetization along each axis in magnetic fields up to 28 T becomes $\mu_c < \mu_b < \mu_a$.

This behaviour can be described using the full set of crystal field parameters given in Ref. [3]: $B_2^0 = 1.35$ K, $B_2^2 = 1.56$ K, $B_4^0 = 2.23 \times 10^{-2}$ K, $B_4^2 = 0.01 \times 10^{-2}$ K, $B_4^4 = 1.96 \times 10^{-2}$ K, $B_6^0 = 5.52 \times 10^{-4}$ K, $B_6^2 = 1.35 \times 10^{-4}$ K, $B_6^4 = 4.89 \times 10^{-4}$ K and $B_6^6 = 4.25 \times 10^{-3}$ K. The magnetization of NdCu₂ along principal crystallo-

graphic axes can then be calculated using the formula

$$\mu = g_J \mu_B \sum_i \langle \eta_i | J | \eta_i \rangle \frac{\exp(-\varepsilon_i/k_B T)}{Z}, \quad (2)$$

where η_i and ε_i are the i th eigenvector and eigenvalue of the total hamiltonian (1), respectively. For the calculation, we have chosen the notation $a = y$, $b = z$ and $c = x$. We may assume that the exchange interaction influences mainly the low field ($B < 7$ T) magnetization data. Moreover, for the precise calculation detailed knowledge of the magnetic structure is necessary. For these reasons, we have neglected the influence of the exchange interaction in present analysis and restricted our calculation only to the fields above 7 T.

The calculated magnetization also shows the crossing of the a - and b -axes magnetization and the shape of calculated curves follows the shape of experimental ones. The fields of crossing of the calculated and experimental curves are in a good agreement. The position of crossing presents another sensitive test of validity of crystal-field parameters. The discrepancy in the value of magnetization can be ascribed to the neglected influence of the

exchange interaction. At temperatures well above the Néel temperature, the effect of the exchange interaction should be much smaller. Therefore, we suggest high-field magnetization experiments to be performed also in paramagnetic range.

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