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Magnetostructural transition in $NdCu_2$

Evidence for an axis conversion like behaviour

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Abstract. The magnetic (H, T)-phase diagram of the orthorhombic compound $NdCu_2$ was investigated for external magnetic fields up to 15 T parallel to the crystallographic c-direction. Magnetization and magnetostriction measurements reveal an anomalous change of the magnetic properties as well as giant magnetostriction (GMS) and large hysteretic effects. This behaviour is similar to that observed in some other RCu_2 compounds where it has been interpreted as a conversion of the magnetic Ising axis. In contrast to these other RCu_2 compounds, however, the easy axis of magnetization in $NdCu_2$ is the b-axis. The macroscopic measurements are compared with neutron diffraction experiments which reveal GMS along the b-axis and a new magnetic phase with propagation vector $\tau = (0.7\ 0\ 0)$ in the converted crystal.

PACS. 71.20.Eh Rare earth metals and alloys – 75.30.Kz Magnetic phase boundaries (including magnetic transitions, metamagnetism, etc.) – 75.80.+q Magnetomechanical and magnetoelastic effects, magnetostriction – 61.12-q Neutron diffraction and scattering

1 Introduction

In the last years a lot of attention has been paid to the orthorhombic RCu_2 compounds where the interplay between the long-range magnetic exchange interaction and the crystalline electric field leads to various interesting magnetic phenomena. $NdCu_2$ is known for a very complex magnetic (H, T)-phase diagram which has been studied extensively in zero external field and for magnetic fields applied parallel to the easy axis of magnetization, the orthorhombic b-direction (see, for example, [1]). Recently, magnetic phase diagrams have also been proposed for magnetic fields applied parallel to the a- and c-axis. These phase diagrams have been obtained by specific heat measurements in fields up to 6 T [2] and magnetization measurements in pulsed fields up to about 50 T [3]. However, the proposed phase diagrams for fields parallel to c differ from each other. In addition, neutron scattering experiments revealed a magnetic phase transition at $T = 2$ K in a static field of $\mu_0 H = 9.5$ T applied in c-direction [4] that is not present in the proposed phase diagrams [2,3]. For that reason, the magnetic $(H_{\parallel c}, T)$ phase diagram has been re-examined in static fields up to 15 T using magnetization and magnetostriction measurements.

Another point of interest in the RCu_2 series is the effect of a so called Ising axis conversion which has first been discovered in $DyCu_2$ by Hashimoto *et al.* [5]. It is thought to be driven by a quadrupolar exchange and has also been found in $CeCu_2$, $PrCu_2$ and $TbCu_2$ [6]. In these compounds the axis conversion occurs when a strong magnetic field is applied along the c-direction: the direction of easy magnetization changes from the a- to the c-axis and the properties with respect to magnetization, specific heat and magnetostriction are exchanged between these two axes. It could be shown for $DyCu_2$ that this magnetic axis conversion is accompanied by a structural phase transition from the orthorhombic to a hexagonal structure due to magnetoelastic coupling [7]. The distortion of the lattice is accompanied by giant magnetostriction (GMS); in $DyCu_2$ the a-axis contracts by about 1.5% while the c-axis expands by the same value. On the b-axis the effect is smaller than 0.1%. Previous explanations were based on a rotation of the quadrupolar moment within the ac-plane [8]. The axis conversion was found to occur only in RCu_2 compounds with magnetic moments lying in the nearly hexagonal ac-plane. In this paper we present experimental results on $NdCu_2$ which show that an axis conversion occurs also in RCu_2 compounds with the b-axis being the easy axis of magnetization in zero field.

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2 Experimental

The experiments have been performed on large $NdCu_2$ single crystals ($5\times7\times5\,mm^3$, for example) which were already used for magnetic structure determinations. The crystal growing and characterization are described elsewhere [9].

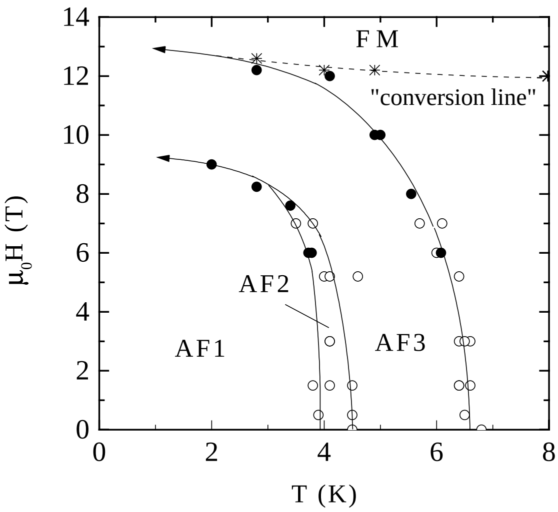

Fig. 1. Phase diagram of $NdCu_2$ for magnetic fields along the c-direction from magnetization and magnetostriction measurements. The open circles refer to scans at constant magnetic field and the closed circles refer to scans at constant temperature. The dotted line represents the metamagnetic transition into the converted state measured by magnetization (stars).

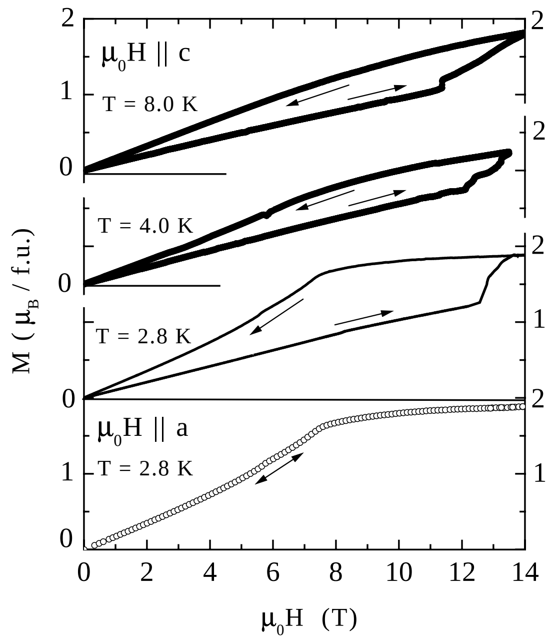

Fig. 2. Magnetization measurements on $NdCu_2$ for magnetic fields along the c-direction at several temperatures. The arrows refer to the direction of the field change. The magnetization curve for magnetic fields parallel to the a-direction is shown for comparison (open symbols).

The magnetization measurements were carried out on a 14 T vibrating sample magnetometer from Oxford Instruments. Thermal expansion and magnetostriction were measured with a capacitance dilatometer described in [10] which was mounted in an Oxford Instruments 15 T magnet cryostat with variable temperature insert. The neutron scattering experiment has been performed on the cold triple-axes spectrometer V2 at the Hahn-Meitner-Institut, Berlin. The instrument was run with bent monochromator and focusing analyzer and an incident-neutron wave vector of $k = 1.5$ Å^{-1}. A Be-filter between sample and analyzer suppressed higher-order contamination. The sample was mounted in the vertical cryo-magnet VM1 (15 T, Oxford Instruments) with the orthorhombic c-axis parallel to the magnetic field. Since the available range of momentum transfer is limited due to the used low neutron energy no Bragg reflection from the lattice could be measured along the a-direction. However, the (0 2 0) reflection is accessible and elastic (0 k 0) scans at this position have been carried out in order to study the influence of strong magnetic fields on the lattice parameter b.

3 Results and discussion

Figure 1 shows the magnetic (H,T)-phase diagram for external magnetic fields parallel to the c-direction as obtained by magnetization and magnetostriction measurements. In zero field, one observes three transitions: at $T_1 = 3.9$ K and $T_2 = 4.5$ K there are two first order phase transitions from the commensurate antiferromagnetic phase AF1 with wave vector $\tau_{\text{AF1}} = (3/5\ 0\ 0)$ into the intermediate incommensurate phase AF2 with $\tau_{\text{AF2}} = (0.603\ 0.003\ 0)$ and from this into the incommensurate antiferromagnetic phase AF3 with $\tau_{\text{AF3}} = (0.615\ 0.044\ 0)$ [1]. At $T_N = 6.5$ K the long range magnetic order disappears and the sample becomes paramagnetic.

Nearly vertical transition lines have been found for fields up to 6 T. Increasing the magnetic field further leads to a flattening of the lines yielding phase transitions at $\mu_0 H \approx 9.5$ T and 13 T at low temperatures confirming the phase transition at 9.5 T observed previously [4].

Figure 2 shows magnetization curves at several temperatures. At $T = 2.8$ K a sudden jump in the magnetization indicates a phase transition at $\mu_0 H_{\text{crit}} \approx 12.5$ T. This transition is accompanied by a large hysteretic effect which has never been observed for magnetic fields applied parallel to the a- or b-direction or for fields lower than 12 T. For decreasing fields one observes two anomalies in the magnetization curve, one at about 7.5 T and one around 5.5 T. Exactly the same changes of the slope are found in the magnetization curve for fields applied parallel to the a-direction in the virgin crystal (compare the lower part of Fig. 2). Repeated field runs show no hysteresis anymore and the magnetization follows the line measured for decreasing field during the first run. This indicates that in zero field the sample has not recovered its initial state. All these features are similar to those found in the RCu_2 compounds where this behaviour is due to a conversion of the Ising axis [5]. The dotted line in Figure 1 appearing at high magnetic fields reflects the temperature dependence of the critical field of the axis conversion defined by the start point of the magnetization jump. Similarly to $CeCu_2$ [6], $PrCu_2$ [11], $DyCu_2$ [12] and $TbCu_2$ [13] the axis conversion occurs also when starting from the paramagnetic state. The corresponding magnetization measurement at $T = 8$ K is shown in the upper part of Figure 2.

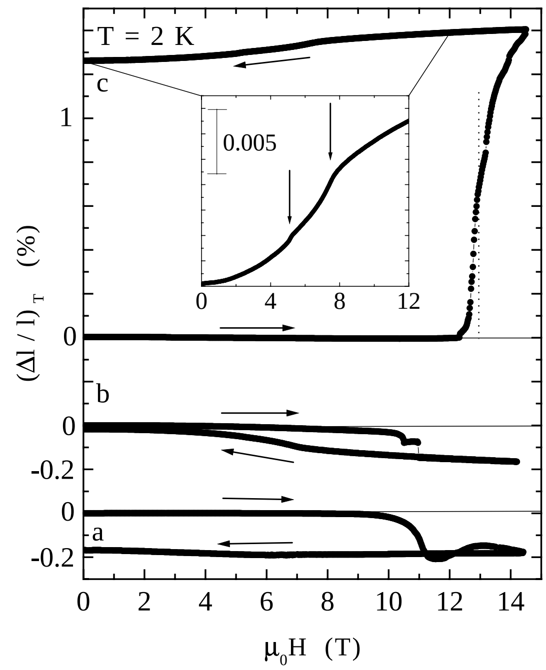

Fig. 3. Magnetostriction of $NdCu_2$ along *a*, *b* and *c*-direction for magnetic fields applied parallel to the *c*-direction. The inset shows the magnetostriction after the conversion process on an enlarged scale.

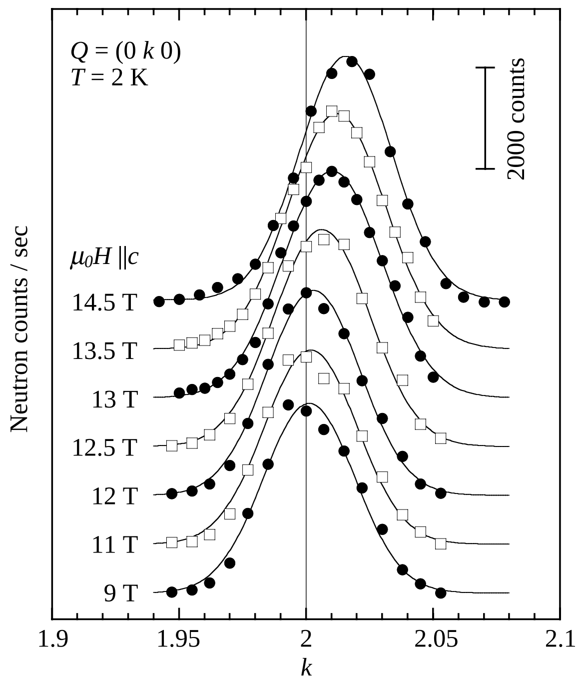

Fig. 4. The (0 2 0) reflection at several magnetic fields applied parallel to the *c*-direction. The scans have been taken for increasing fields. The *k* values refer to the reciprocal lattice units in zero field.

In order to see if structural changes accompany the conversion process we measured the magnetostriction for magnetic fields applied parallel to the *c*-direction. The results are illustrated in Figure 3 for $T = 2$ K. The *a*-axis exhibits a contraction by -0.2% beginning at the magnetic phase transition at $\mu_0 H = 9.5$ T. The contraction is followed by a small expansion that is connected to the axis conversion at 12.5 T. In *c*-direction the application of a magnetic field along the *c*-axis leads to negligible magnetostriction up to $\mu_0 H = 12$ T. Above this field a GMS effect is observed which is associated with the axis conversion process. The crystal expands by $+1.4\%$ in *c*-direction. The magnetostriction in *b*-direction will be discussed later in comparison with the neutron scattering results.

In decreasing field two distinct transitions are observed at $\mu_0 H = 7.7$ T and 5.3 T as indicated by the arrows in the inset of Figure 3. These transitions are also found in the virgin *a*-axis behaviour with field in *a*-direction (not shown here). The contraction of the lattice in *a*- and the expansion in *c*-direction do not relax in decreasing field and are still present in zero field.

To get some insight into the conversion process on an atomic scale we performed neutron diffraction experiments. This method allows to measure the lattice parameters directly. Figure 4 shows (0 *k* 0) scans at several magnetic fields between 9 and 14.5 T (at $T = 2$ K). The position of the (0 2 0) reflection remains unchanged for fields up to 11 T. Beginning at $\mu_0 H = 12$ T, a deviation from this position is observed. At $\mu_0 H = 14.5$ T, the shift corresponds to a contraction of the lattice in *b*-direction of about -0.8%.

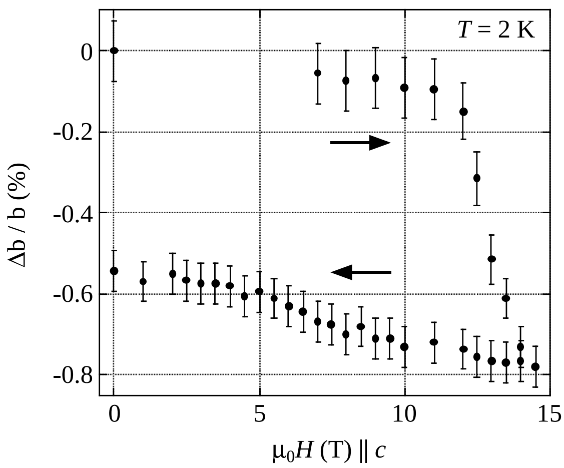

Fig. 5. The field dependence of the lattice parameter *b* determined by the position of the (0 2 0) reflection. The arrows indicate the direction of the field change.

Decreasing the magnetic field does not result in a relaxation of the contraction in *b*-direction as can be seen in Figure 5. The position of the (0 2 0) reflection changes only to slightly smaller *k* values that correspond to an expansion of only about $+0.25\%$ between $\mu_0 H = 14.5$ T and zero field.

These results differ distinctively from the *b*-axis behaviour observed by the magnetostriction measurement shown in Figure 3. There, the contraction is about -0.2% and it relaxes nearly completely in decreasing field. In addition, the contraction occurs already at lower fields and exhibits a step-like behaviour. These differences are thought to be due to the repetition of the conversion cycle. On the one hand, it is known from $DyCu_2$ that repeated conversion may result in the destruction of the sample or at least in microcracks in the single crystal. In macroscopic measurements like magnetostriction such microcracks influence the results considerably whereas, in microscopic measurements like neutron diffraction they are of minor importance. On the other hand, heating the sample up to room temperature for some minutes might not be sufficient for a complete re-conversion of the crystal back into the initial state. This leads to the formation of domains that might explain the step-like behaviour that has neither been observed for the magnetostriction in c-direction (first cycle) nor in the neutron diffraction experiment (first cycle on another single crystal). Therefore, the data obtained by neutron diffraction are more reliable and the magnetostriction measurements in a- and b-direction should only be considered qualitatively.

However, comparing the b-axis behaviour in decreasing field one finds a rather good agreement between the data obtained by the two different techniques. Regarding the expansion of the b-axis as function of the decreasing field, there are two changes of the slope, one around 8 T and one at about 5.5 T (Fig. 5). These indications of phase transitions have also been found in the magnetization and magnetostriction measurements (see Fig. 2 and inset of Fig. 3).

The changes of the lattice parameters are thought to be due to a rearrangement of the atoms in the nearly hexagonal ac-plane. The magnetostriction measurements show that the behaviour of the a- and c-axis in $NdCu_2$ is quite similar to that in $DyCu_2$ and $TbCu_2$, *i.e.* one finds a strong expansion in c-direction and a contraction in a-direction. This indicates that also in $NdCu_2$ a change of lattice symmetry from orthorhombic to hexagonal may occur as found in $DyCu_2$ [7]. The magnetostrictive effect on the a-axis beginning at the phase transition at $\mu_0 H = 9.5$ T might indicate an effect which is prerequisite for the axis conversion at $\mu_0 H = 12.5$ T. The development of a component of the magnetic moment in a-direction could be such an effect. It approaches the situation of the RCu_2 compounds where the a-axis is the easy axis of magnetization. Further neutron diffraction experiments have to be performed in order to prove such a scenario. For a definitive proof of hexagonal symmetry it would be necessary to investigate reflections which are forbidden by hexagonal selection rules – however, no such reflections were accessible in our neutron scattering experiments.

Figure 6 shows a longitudinal scan along the a-direction at zero magnetic field after a conversion cycle. One observes the magnetic reflections τ_{AF1}, $3\tau_{AF1}$ and $5\tau_{AF1}$ which correspond to the well-known phase AF1 with the magnetic ordering vector $\tau_{AF1} = (0.6\ 0\ 0)$ (compare [1]). These reflections have only about 20% of their initial intensity in the virgin state and they appear at exactly the same $\mathbf{Q}$ positions as in the non-converted crystal. The same reduction of intensity has been found for the reflections of the incommensurate magnetic phase between

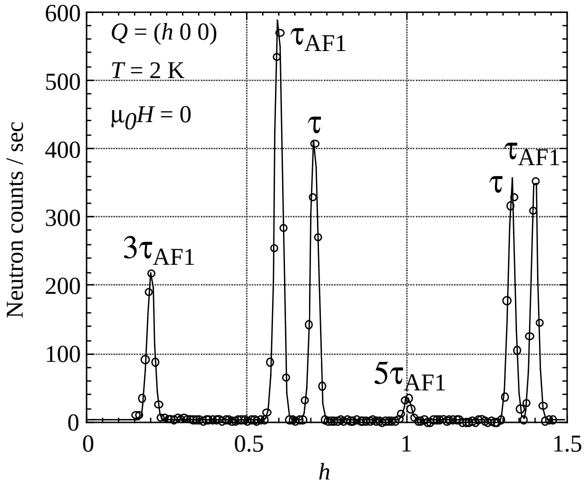

Fig. 6. Longitudinal scan along the a-direction at zero magnetic field after the conversion. One observes magnetic reflections belonging to two different crystallographic lattices. The wave vector Q, *i.e.* h, refers to the virgin lattice.

13.5 and 9.5 T. From that we conclude that about 20% of the crystal forms a domain which had not been converted.

In addition to the reflections of AF1, Figure 6 shows two reflections which are described by another wave vector τ. It is obvious that these two reflections also belong to another crystallographic lattice. Calculating the lattice parameter a from their positions one finds a contraction of the a-axis by -2% with respect to the initial value. This is an order of magnitude more than observed in the macroscopic magnetostriction measurement in Figure 3. Microcracks in the single crystal are again a possible explanation for this difference. Since the additional magnetic reflections in the neutron scattering pattern appear only after the conversion there is no doubt that they belong to a new magnetic phase that is present in the converted part of the crystal only. This phase is stable between zero field and 5.5 T and a magnetic ordering vector of $\tau = (0.7\ 0\ 0)$ (with respect to the converted lattice) can be assigned to it. However, due to the restriction to the ab-scattering plane our experiment cannot exclude that the τ-reflections are slightly split in c-direction. Such a splitting could be the reason why no 5τ-reflection is observed around (0.5 0 0).

The converted state is thought to be present in about 80% of the crystal. However, the intensity of the reflections of the new magnetic phase is the same or even less than the intensity of the reflections described by τ_{AF1} that represent 20% of the crystal. This contradiction can be removed by comparing our results to those obtained for $DyCu_2$ [7]. In $DyCu_2$ three equivalent magnetic domains have been found that are characterized by a propagation turned by 60° in accordance with the hexagonal symmetry of the converted system. Applying this scenario to $NdCu_2$ would mean that the observed τ reflections represent only a third of the converted part of the crystal, *i.e.* about 25% of the whole crystal. This is in reasonable agreement with the observed intensities and it is a further argument that the axis conversion in $NdCu_2$ changes the crystal symmetry from orthorhombic to hexagonal.

4 Conclusion

$NdCu_2$ is found to exhibit a field induced magnetostructural transition similar to the axis conversion found in other RCu_2 compounds. However, $NdCu_2$ is the first compound with the *b*-axis being the easy-axis of magnetization that shows this axis conversion. The magnetostructural transition occurs from an orthorhombic to a probably hexagonal symmetry and is responsible for a GMS effect. After the conversion process a new magnetic phase is observed which unambiguously belongs to the converted crystal. This underlines the fundamental changes of the magnetic and structural properties of $NdCu_2$ during the axis conversion.

Theoretical models for the effect of an axis conversion are based on magnetoelastic coupling leading to strong quadrupolar exchange [8]. However, further models have to be developed to explain the symmetry change of the lattice and the special case of $NdCu_2$.

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