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The magnetic structures of $NdCu_2$ determined by single crystal neutron diffraction

M. Loewenhaupt [1], Th. Reif [1], R.R. Arons [1], E. Gratz [2], M. Rotter [2], B. Lebech [3]

[1] Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, Postfach 1913, D-52425 Jülich, Germany (Fax: +49 2461 61 26 10, Tel.: +49 2461 61 31 39)
[2] Institut für Experimentalphysik, TU Wien, Wiedner Hauptstrasse 8–10, A-1040 Wien, Austria (Fax: +43 1 586 31 91, Tel.: +43 1 588 01 55 41)
[3] Risø National Laboratory, Postbox 49, DK-4000 Roskilde, Denmark (Fax: +45 42 37 01 15, Tel.: +45 46 77 47 05)

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Abstract. We investigated the magnetic structure of $NdCu_2$ by means of neutron diffraction as a function of temperature between 1.5 K and 8 K in zero external field. The diffraction data were obtained on two single crystals with different orientations using the triple-axis-spectrometer TAS6 at the DR-3 reactor at Risø. Two magnetic phases were observed between 1.5 K and $T_N = 6.5$ K. From 1.5 K to 4.1 K the magnetic reflections can be described by the commensurate wave vector $\tau = (3/5\ 0\ 0)$ and its higher harmonics 3τ and 5τ. Below 2.5 K the structure is completely squared-up. For 4.1 K $\leq T \leq$ 6.5 K the magnetic structure is incommensurate with the chemical lattice and can be described by the wave vector $\tau^* = (0.62\ 0.044\ 0)$. In both phases the Nd-moments are oriented along the easy **b**-direction.

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1. Introduction

A large amount of material has been published up till now concerning the rare earth transition metal $(R-T)$ compounds. The AB_2 stoichiometry with the cubic Laves-phase structure has particulary profited in this respect partly due to the relative simplicity of this crystal structure and also because of its wide range of formation, being the only structure to exist for all the $3d$-elements up to Ni to the right of Cr in the periodic table. Considerably less attention has been paid, however, to the compounds with Cu which crystallize in the orthorhombic $CeCu_2$ structure. At first Sherwood et al. [1] reported metamagnetism in the RCu_2 compounds. They concluded that there exists a relatively weak antiferromagnetic exchange interaction in this series with the highest Néel temperature found for $TbCu_2$ at 54 K. A comprehensive study of the magnetization of the RCu_2 intermetallics has been published by Hashimoto in 1979 [2]. The first neutron diffraction experiments by Brun et al. [3] on $TbCu_2$ where interpreted in terms of a collinear antiferromagnetism of the R-spins directed along the a-direction at 4.2 K. In the meantime a number of investigations of the specific heat and the thermal expansion [4] as well as neutron diffraction [5] performed on RCu_2 revealed that in most of these orthorhombic intermetallics one or more first- or second-order magnetic transitions exist below T_N. It is interesting to note that among the cubic 1:2 compounds with other $3d$-metals spin reorientations in the ordered state are much less frequently observed although there are few known in RCo_2 ($HoCo_2$ and $NdCo_2$) and in some compounds within the RMn_2 series (see e.g. [6]). We may assume that in the low-symmetric $CeCu_2$-type structure, a subtle interplay of the exchange interaction on the one hand and the crystal field on the other hand exists, which is the origin for the magnetic instabilities observed in these 1:2 Cu based R-intermetallics. It is obvious that the crystal field plays an important role for the stability of the magnetic structure, since no such magnetic phase transitions in zero field have been observed in $GdCu_2$ [7]. In $NdCu_2$ with $T_N = 6.5$ K a change of the spin arrangement at 4.1 K has been found in our recent investigation [8]. There we proposed a preliminary magnetic phase diagram based on magnetization, specific heat and magnetoresistance measurements on powdered samples. A recently performed neutron diffraction measurement [9] revealed below 4.1 K a commensurate spin arrangement with a wave vector given by $\tau = (3/5\,0\,0)$. From the observation of the third harmonic 3τ below 4 K down to 1.4 K a progressive squaring-up has been deduced. The magnetic phase above 4.1 K can be described by a sinusoidal oscillating component with an incommensurate, two-dimensional wave vector $\tau^* = (0.62\,0.044\,0)$. The aim of the present investigation was to study the magnetic phase diagram (with and without an external magnetic field) in detail using a single crystal of $NdCu_2$ of high quality. In this publication we mainly report the results of the zero field experiments.

2. Experimental

Two single crystals of $NdCu_2$ have been grown by the Czochralski method. From these crystals two samples were spark cut in the form of parallelepipeds with edges along the main symmetry directions *a*, *b* and *c*. A larger crystal with dimensions $(5 \times 7.5 \times 5.2)\ mm^3$ was used for measurements in the (*ac*)-plane. Data in the (*ab*)-plane were taken on a smaller crystal with dimensions $(4 \times 4 \times 5)\ mm^3$. For both crystals the mosaic spread was less than 0.5°. As structural data we determined lattice and positional parameters in agreement with those given in [9] (orthorhombic $CeCu_2$-type structure with space group Imma (D_{2h}^{28}), number 74, and $a = 0.437$ nm, $b = 0.698$ nm, $c = 0.737$ nm, $y(Cu) = 0.0506$, $z(Cu) = 0.167$ and $z(Nd) = 0.5404$).

Neutron diffraction experiments were carried out at the cold neutron source TAS 6-spectrometer at the DR 3-reactor at Risø National Laboratory. The triple-axis-spectrometer was operated in a two-axis mode with a pyrolytic graphite ((002) reflection) monochromator set at 13.6 meV. The small crystal was mounted in the sample environment of a ^4He cryostat which could be operated down to 1.5 K. The large crystal was mounted in the sample chamber of a 5 *T* split coil cryo-magnet (run in zero field mode). The temperature readings were slightly different in the two sample environments, but we fixed the temperature scale by setting T_N exactly equal to 6.5 K. The applied corrections were of the order of a few tenths of K.

Strong nuclear reflections like (200) suffered severely from extinction, leaving only few of the weak nuclear reflections for intensity calibration of the magnetic reflections. No extinction or absorption corrections, however, were applied to the data. The Lorentz factor for the peak intensity was taken as $1/\sin 2\Theta$.

3. Results and discussion

3.1. Low temperature magnetic phase at $T \leqq 4.1$ K

The magnetic reflections as investigated by neutron powder diffraction on $NdCu_2$ below 4.1 K [9] could be interpreted as belonging to a fundamental wave vector $\tau = (3/5\,0\,0)$ and its third harmonic 3τ. These findings are fully confirmed by our single crystal data taken in the (*ab*)- and (*ac*)-plane. However, in addition to τ and 3τ reflections we could also identify reflections belonging to the *fifth harmonic* 5τ. These were found at positions with integer Miller indices like e.g. (001), (003) or (100), (201). In the (*bco*) chemical cell these points belong to the zone boundary. Thus the appearance of reflections of this type can also be interpreted as a simple antiferromagnetic Fourier component with propagation vector along *a*-direction and period *a*. The observation of the fifth harmonic enables us to propose a magnetic structure at the lowest temperatures ($T \leqq 2.5$ K) where all Nd magnetic moments have the same magnitude. *We observe complete squaring-up.* Figure 1a and 1b show the low temperature patterns (reciprocal space) of magnetic reflections of type τ (circles), 3τ (squares) and 5τ (triangles) and of allowed nuclear reflections (full circles) in the (*ab*)- and (*ac*)-plane, respectively. No magnetic reflections are observed on the *b*-axis and all magnetic reflections with large Miller index *k* are very weak. From this we can deduce that all moments are oriented along the *b*-direc-

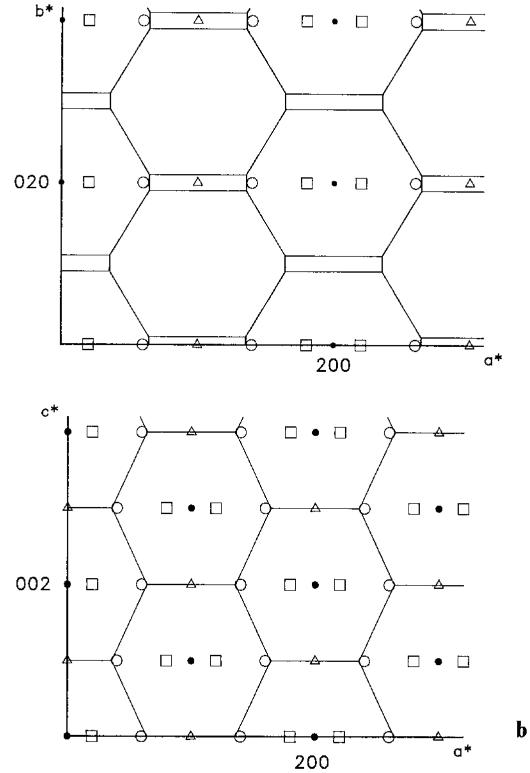


Fig. 1. Positions of magnetic (open symbols) and nuclear (full circles) reflections in the reciprocal (*ab*)-plane **a** and (*ac*)-plane **b** of $NdCu_2$ in the low temperature phase. The different open symbols denote the different harmonics of the wave vector: ○ for τ, □ for 3τ and △ for 5τ. The line pattern refers to the Brillouin zone boundaries of the chemical unit cell

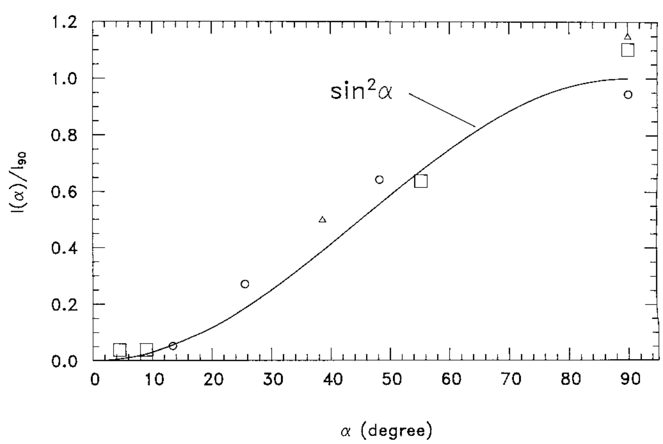
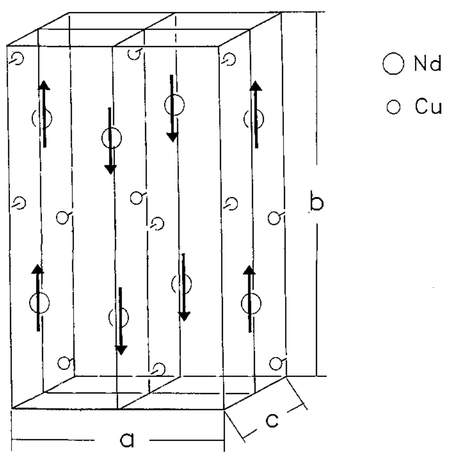


Fig. 2. Angular dependence of the magnetic intensities at $T = 1.5$ K in the (*ab*)-plane. The angle α is between the moment direction and **Q**. The experimental data are best reproduced if the moment direction is along the *b*-direction. The different symbols denote the data for the different harmonics of the wave vector: ○, for τ; □, for 3τ and △, for 5τ

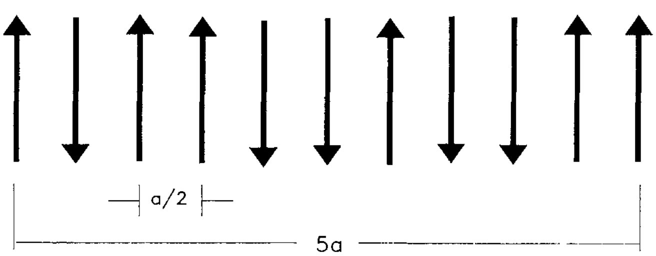
tion yielding a collinear magnetic structure for $NdCu_2$. The dependence of the magnetic intensities of some reflections on the orientation factor $\sin^2\alpha$ (see (2)) is shown in Fig. 2.

In the a-directions the magnetic reflections are found on lines shifted by integer (odd or even) values of $\frac{2\pi}{c}$ in c-direction. In b-direction the magnetic reflections occur only on lines shifted by even values of $\frac{2\pi}{b}$. This yields ferromagnetic (bc)-planes stacked in a complicated antiferromagnetic order along the a-direction comprising five unit cells with altogether ten (bc)-planes. This reduces the structure determination to a one-dimensional problem. The stacking in the a-direction can then be determined from the Fourier coefficients of τ, 3τ and 5τ reflections and an appropriate choice of phases from the following equation:

$$\mu_i(R_i)=\mu_\tau \sin\left(\frac{2\pi}{a} R_i\cdot\tau+\phi_1\right) + \mu_{3\tau} \sin\left(\frac{2\pi}{a} R_i\cdot 3\tau+\phi_3\right) + \mu_{5\tau} \sin\left(\frac{2\pi}{a} R_i\cdot 5\tau+\phi_5\right). \quad (1)$$



a



b

Fig. 3. a Chemical unit cell of $NdCu_2$. Also shown is the spin configuration of the low temperature phase for the first three (bc)-planes. The full magnetic unit cell comprises five cells or ten (bc)-planes in a a-direction. **b** Stacking sequence of the magnetic moments of Nd in the a-direction in the low temperature phase. Each arrow represents the spin direction of a ferromagnetic (bc)-plane. The first three arrows on the left correspond to the sequence shown in **a**

Here R_i are the positions of the ferromagnetic (bc)-planes at 0, $a/2$, $a, \ldots 5a$. From the powder data [9] the first and third Fourier coefficient at $T \leqq 2.5$ K has been determined to be $\mu_\tau=(2.32\pm0.1)\,\mu_B$ and $\mu_{3\tau}=(0.90\pm0.1)\,\mu_B$, respectively. The single crystal data are consistent with these values for μ_τ and $\mu_{3\tau}$; for the coefficient of the fifth harmonic we obtain $\mu_{5\tau}=(0.36\pm0.1)\,\mu_B$. With $\phi_1=\phi_5=\pi/2$ and $\mu_3=-\pi/2$ we obtain the unique moment of $\mu=1.78\,\mu_B$ for all Nd ions. The chemical unit cell is presented in Fig. 3a to visualize the ferromagnetic (bc)-planes. In Fig. 3b we show the stacking sequence in the a-direction. Here each arrow represents a spin in a (bc)-plane. In Tables 1 and 2 we give the observed and calculated intensities of magnetic and nuclear reflections at $T\leqq 2$ K obtained for the (ab)- and (ac)-planes employing two different single crystals and experimental set-ups. The magnetic intensities are calculated for τ and 3τ using

$$|F_M|^2=(1/4)\,(e^2\gamma/2mc^2)^2\,\mu^2\,\sin^2\alpha \quad (2)$$

for the $(000)^\pm$ structure factor with $(e^2\gamma/2mc^2)=0.2695\cdot10^{-12}$ cm. α is the angle between the scattering vector **Q** and the direction of the magnetic moment and μ is the amplitude of the sinusoidal oscillation in μ_B. For 5τ we have to drop the factor (1/4) in (2) as in this case each reflection consists of two satellites. The summation of the two amplitudes yields a factor of 4 for the intensities which are proportional to the square of the structure factor. Alternatively we have calculated the magnetic and nuclear reflections by evaluating the structure factor for the five-fold extended unit cell and the magnetic structure as shown in Fig. 3 using the ICPW program of W. Schäfer. For perfect squaring-up both methods give, of course, the same results. The magnetic form factor, $f(Q)$, is taken as isotropic and is approximated by $f^2(Q)=\exp[-(Q[\text{Å}^{-1}]/6.6)^2]$.

For increasing temperatures the intensities of the higher harmonic reflections decrease much faster than

Table 1. Magnetic and nuclear reflections of $NdCu_2$ in the low temperature phase measured on the small crystal in the (ab)-plane at $T=1.8$ K

(hkl)	2Θ	I_{calc}	I_{obs}
$(000)^{1+}$	19.34	41765	18800
$(020)^{1+}$	45.77	3345	5298
$(040)^{1+}$	92.33	558	646
$(200)^{1-}$	46.16	17781	14850
$(220)^{1-}$	63.47	7391	7282
$(\bar{2}20)^{3+}$	41.61	73	81
$(\bar{2}40)^{3+}$	89.45	10	1036
$(000)^{3+}$	60.53	2086	2158
$(020)^{3+}$	75.77	1171	610
$(220)^{3-}$	41.61	73	71
$(420)^{3-}$	90.29	1735	133
$(400)^{5-}$	32.52	2403	1760
(020)N	41.06	47240	23980
(040)N	89.07	199176	50840
(220)N	82.71	30322	18190
(200)N	68.10	804256	100000

Table 2. Magnetic and nuclear reflections of $NdCu_2$ in the low temperature phase measured on the large crystal in the (*ac*)-plane at $T = 2.2$ K

(hkl)	2Θ	I_{calc}	I_{obs}
$(000)^{1+}$	19.34	66825	41420
$(101)^{1+}$	57.08	21934	9958
$(002)^{1+}$	43.71	23072	23160
$(200)^{1-}$	93.43	28449	30130
$(202)^{1-}$	61.83	16766	16980
$(103)^{1-}$	61.43	11718	14550
$(000)^{3+}$	60.52	3338	3651
$(\bar{1}01)^{3+}$	32.38	5665	5675
$(\bar{2}02)^{3+}$	39.38	3839	3624
$(\bar{1}03)^{3+}$	66.23	1654	2024
$(\bar{2}04)^{3+}$	83.64	759	843
$(400)^{3-}$	76.04	2775	2629
$(301)^{3-}$	79.28	4203	4273
$(202)^{3-}$	39.38	3839	3730
$(303)^{3-}$	73.88	1517	1539
$(304)^{3-}$	96.24	759	888
$(400)^{5-}$	32.52	3851	4522
$(301)^{5-}$	19.12	6110	4303
$(501)^{5-}$	71.47	1746	893
$(303)^{5-}$	59.78	1123	1823
$(101)N$	38.00	242	1953
$(002)N$	38.81	3578	3838
$(004)N$	83.27	27504	32050
$(202)N$	81.25	2199	1037
$(200)N$	68.10	1286809	158260
$(103)N$	69.72	1054399	141300

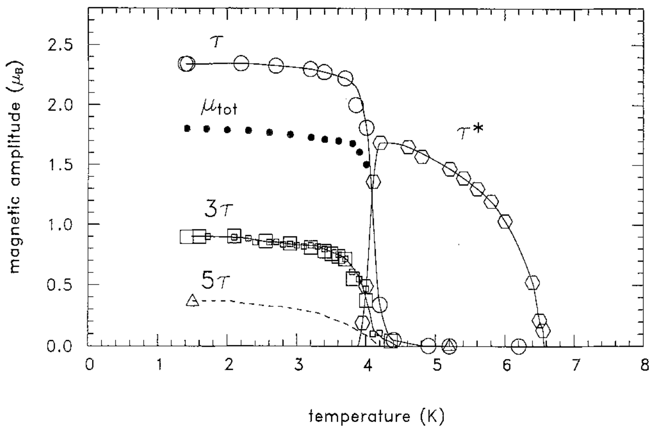


Fig. 4. Temperature dependence of the magnetic intensities in the two phases of $NdCu_2$. ○, for τ measured at (1.4 0 0); large and small □, for 3τ measured at (1.8 0 0) in the (*ab*)- and (*ac*)-plane, respectively; △, for 5τ measured at (1 0 0). The open hexagons: sum of the intensities of τ^* measured at (1.38 ± 0.044 0). The dashed curve is a guess

those connected with the fundamental wave vector τ. Figure 4 shows the variation of the intensities of τ and 3τ with temperature. At the phase transition temperature (4.1 K) the intensity of τ drops within a few tenth K from a finite value to zero while the intensities of the higher harmonic 3τ and presumably 5τ disappear continuously according to power laws. Towards the

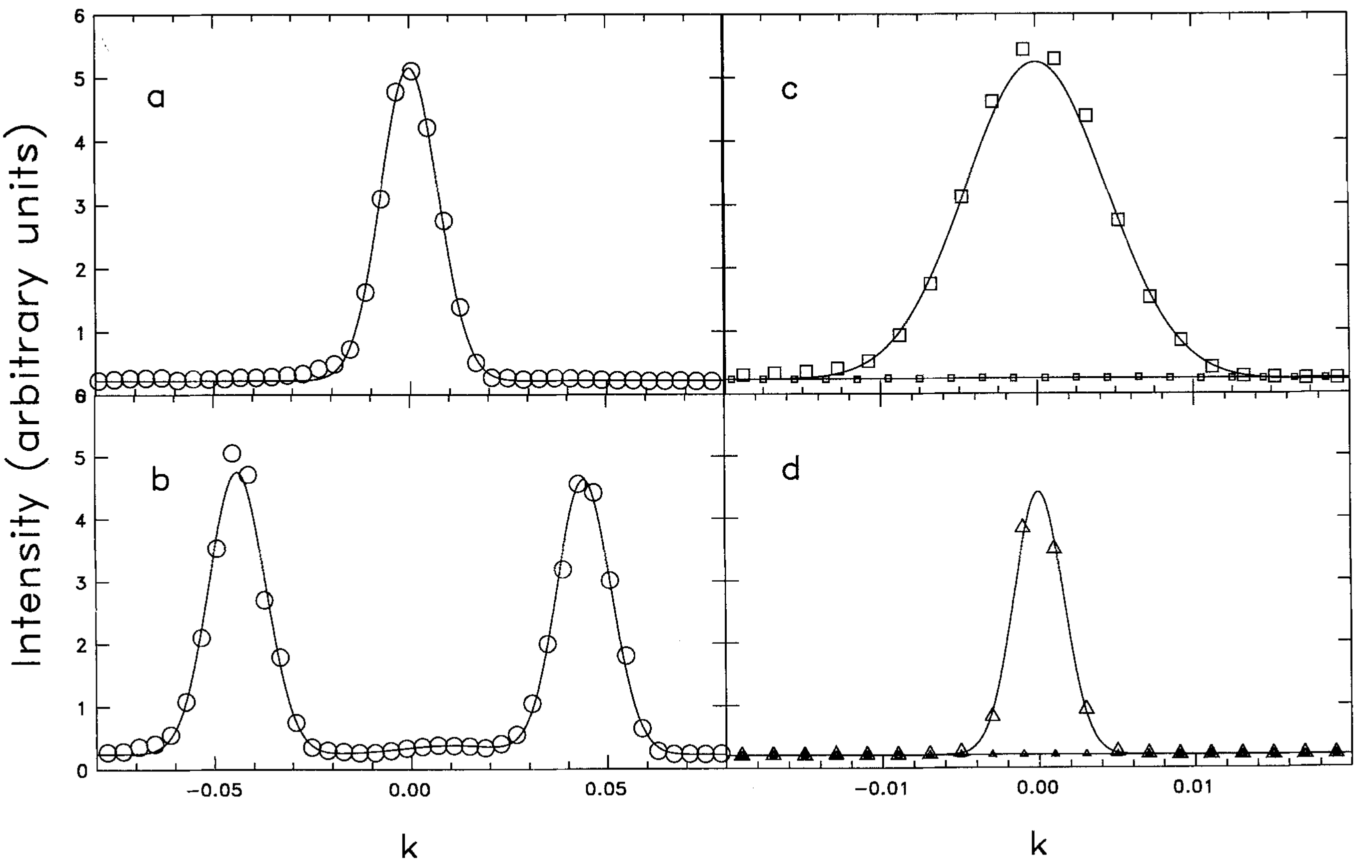


Fig. 5a–d. Selected scans of magnetic reflections of the low and high temperature phases of $NdCu_2$: **a** (1.4*k*0) at $T = 4$ K for τ, **b** (1.38*k*0) at $T = 4.9$ K for τ^*, **c** (1.8*k*0) at $T = 2$ K and $T = 4.5$ K for 3τ, marked by large and small symbols, respectively, and **d** (1*k*0) at $T = 2$ K and $T = 5$ K for 5τ, marked by large and small symbols, respectively

phase transition the squaring-up of the long-range ordered components of the Nd magnetic moments gradually diminishes indicating that at the phase transition only the Fourier component of the fundamental wave vector τ is left. The character of the phase transition at 4.1 K is of first order. The magnetic structure of the second phase in zero field at $4.1\ \mathrm{K} \leqq T \leqq 6.5$ K is characterized by the existence of magnetic reflections belonging to just one, incommensurate, two-dimensional propagation vector $\tau^* = (0.62\ 0.044\ 0)$, which is only slightly displaced from the fundamental wave vector τ of the low temperature phase. In Fig. 5a we show the fundamental reflection τ at (1.4 0 0) at $T = 4$ K in the low temperature phase and in Fig. 5b the corresponding reflection τ^* at $(1.38 \pm 0.44\ 0)$ at $T = 4.9$ K in the high temperature phase. At $T = 4.1$ K, the intensities of τ and of the sum of the intensities of the two reflections of τ^* with $a \pm b$-component are equal within the experimental error. The higher harmonics 3τ and 5τ are only present in the low temperature phase as seen in Fig. 5c, d. The high temperature phase will be discussed in more detail below.

3.2. High temperature magnetic phase at 4.1 K ≦ T ≦ 6.5 K

As already deduced from neutron powder diffraction the positions of the magnetic reflections of the high temperature phase can be described by one incommensurate, two-dimensional wave vector $\tau^* = (0.62\ 0.042\ 0)$ [9]. This is confirmed by our single crystal data which give only a slightly larger value of 0.044 for the b-component of the wave vector. Figure 6a, b show the patterns of magnetic (open circles) and nuclear (full circles) reflections in the (ab)- and (ac)-plane, respectively. Actually, there are no magnetic reflections lying exactly in the (ac)-plane because of the small b-component of the wave vector. However, the coarse resolution perpendicular to the scattering plane makes it possible to observe the sum of the two reflections which are lying just above and below the (ac)-plane. We have indicated this situation by using two concentric circles to mark the projections of the magnetic reflections onto the (ac)-plane in Fig. 6b. All magnetic reflections in the high temperature phase occur practically at the same lines in reciprocal space as the reflections due to the fundamental wave vector τ in the low-temperature phase. Hence, there are again ferromagnetic (bc)-planes. However, because of the small b-component of τ^* the (bc)-planes are not exactly ferromagnetic. The period in b-direction of the wave vector corresponds to approximately 23 unit cells. The intensities of the magnetic reflections follow the curve shown in Fig. 2 for the low temperature phase. Therefore, the sinusoidally modulated moments are also oriented along the b-direction in the high temperature phase.

As mentioned previously, the intensity of a magnetic reflection connected with τ in the low temperature phase agrees approximately with the sum of the intensities of the two corresponding reflections of τ^* in the high temperature phase close to 4.1 K (see Fig. 4). Thus, the amplitude of the sinusoidal modulation is continuous at the phase transition. The value of the amplitude of τ^*

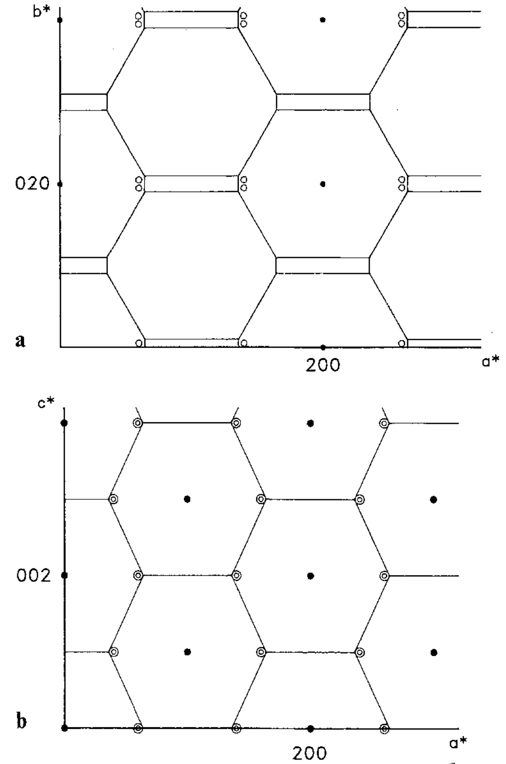


Fig. 6. Positions of magnetic (open symbols) and nuclear (full circles) reflections in the reciprocal (ab)-plane **a** and (ac)-plane **b** of $NdCu_2$ in the high temperature phase. The symbols ⊙ for the magnetic reflections in the (ac)-plane indicate that the reflections are lying slightly above and below the plane due to the small b-component of the wave vector τ^*. The line pattern refers to the Brillouin zone boundaries of the chemical unit cell

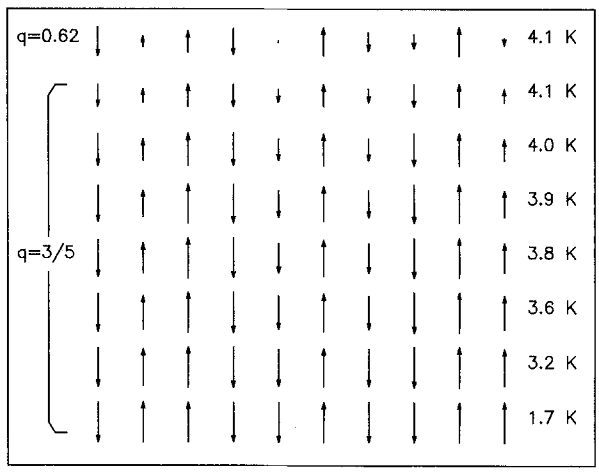


Fig. 7. Stacking sequence of the magnetic moments in the a-direction at different temperatures. The sequence is a square wave at 1.5 K with 1st, 3rd and 5th harmonics ($\tau = 3/5$). For increasing temperatures the sequence becomes more sinusoidal with only the 1st harmonic present at 4.1 K, where the sequence becomes incommensurate ($\tau^* = 0.62$). The amplitudes of the harmonics were deduced from Fig. 4

is about $(1.75 \pm 0.10)\,\mu_B$ close to the phase transition. From the powder data we found an amplitude of $(1.74 \pm 0.10)\,\mu_B$ for τ^* at $T = 5.2$ K [9]. Hence, the phase transition occurs when the *amplitude of the sinusoidal modulation* approaches the value of the (squared-up) *magnetic* *moment* at low temperature (see dotted curve in Fig. 4). The transition of the stacking sequence of the magnetic moments in the a-direction from the squared-up structure at $T = 1.5$ K to the sinusoidal modulated, commensurate ($\tau = 3/5$) and incommensurate ($\tau^* = 0.62$) structures at $T = 4.1$ K is illustrated in Fig. 7. For increasing temperatures above 4.1 K the intensities of the τ^* reflections decrease continuously and disappear at $T_N = 6.5$ K according to a power law. The phase transition into the paramagnetic state is of second order.

4. Conclusions

In conclusion we want to emphasize that the use of a single crystal of $NdCu_2$ for the neutron diffraction experiments widens considerably our understanding of the magnetic structures in zero field below T_N, if compared to our previous powder diffraction experiment:

● The observation of the fifth harmonic 5τ in the commensurate state for $T \leqq 4.1$ K, only observable in the single crystal experiment, unambiguously confirms the complete squaring-up of the Nd moments towards zero Kelvin.

● The phase transition at 4.1 K is of first order type. The analysis of the present single crystal data revealed that the phase transition occurs if the amplitude of the sinusoidal modulation ($1.75\,\mu_B$) is approximately equal to the value of the squared-up magnetic moment at low temperature ($1.78\,\mu_B$).

The results of both studies, neutron powder diffraction and single crystal experiment, are consistent. We can describe the magnetic structures of $NdCu_2$ in zero field by a commensurate wave vector $\tau = (3/5\ 0\ 0)$ and its third and fifths harmonics in the low temperature phase ($\leqq 4.1$ K) and by an incommensurate, two dimensional wave vector $\tau^* = (0.62\ 0.044\ 0)$ in the high temperature phase ($4.1\ \mathrm{K} \leqq T \leqq 6.5$ K). The value of the magnetic moment at 1.5 K is $(1.78 \pm 0.1)\,\mu_B$ per Nd atom which is in excellent agreement with the single crystal magnetization measurements.

To determine the spin structure in the whole magnetic phase diagramm (H vs. T) single crystal experiments in an external field are in progress.

Note added in proof. In preliminary measurements in finite fields we observed between the commensurate, low-temperature phase and the incommensurate, high-temperature phase at $T = 3$ K and $H = 0.75$ T another phase. Its main feature is an additional, very small splitting (± 0.014) in a-direction of the τ, 3τ and 5τ reflections. Recent specific heat data in zero field (Y. Onuki, private communication) showed that the transition at 4.1 K is actually a double transition at 3.97 K and 4.22 K. Thus the intermediate phase exists also in zero field. Specific heat measurements in finite fields and neutron diffraction experiments on the large $NdCu_2$ single crystal with better temperature resolution to investigate the details of the transition from the commensurate to the incommensurate phase are currently underway.

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