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Commensurate–incommensurate magnetic phase transitions in $PrCu_2Ge_2$ and $NdFe_2Ge_2$

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Abstract

New neutron diffraction and ac magnetic susceptibility data concerning phase transitions and magnetic structures of $PrCu_2Ge_2$ and $NdFe_2Ge_2$ are presented here. In the case of $PrCu_2Ge_2$ at low temperatures collinear antiferromagnetic ordering of the AFI type is observed. In the temperature range between $T_t = 5.5$ K and $T_N = 15.0$ K the collinear and a sine wave modulated structure coexist. The latter phase is a long period modulation along the a-axis described by the propagation vector $\mathbf{k} = (0.0336(5), 0, 0)$. In the case of $NdFe_2Ge_2$ the situation is similar: a collinear AFII-type low temperature phase ($\mathbf{k} = (0, 0, 1/2)$) changes into a non-collinear one ($\mathbf{k} = (0, 0, 0.46(1))$) at about 12 K. In both compounds in all phases magnetic moments localized on rare earth ions are parallel to the c-axes.

1. Introduction

Ternary rare earth RT_2X_2 (R—rare earth, T—3d, 4d or 5d transition metal and X = Si or Ge) intermetallic compounds have been intensively studied for several decades due to the wide spectrum of different physical behaviour types displayed by this group's members [1, 2]. Most of them crystallize in the body centred tetragonal structure of the $ThCr_2Si_2$ type (space group $I4/mmm$) [3]. The R, T and X atoms occupy the 2a, 4d and 4e sites, respectively. The atoms are arranged in planes stacked perpendicularly to the c-axis with the sequence R–X–T–X–R. In these compounds, except for the RMn_2X_2 series, magnetic moments are localized only on the rare earth atoms [1, 2].

Neutron diffraction data indicate that the rare earth magnetic moments form different orderings (see figure 23 in [1]). In the case of the compounds with Pr and Nd, magnetic

orderings change with composition and temperature [4, 5]. In $PrFe_2Si_2$ and $NdFe_2Si_2$, collinear orderings of the AFII type with the propagation vector $\mathbf{k} = (0, 0, 1/2)$ and in $PrCu_2Si_2$ with $\mathbf{k} = (0, 0, 1)$ are stable up to the Néel temperatures. For $PrFe_2Ge_2$, magnetic ordering of the AFII type is stable up to $T_t = 9$ K while above this value a sine wave modulated with $\mathbf{k} = (0, 0, k_z)$ ($k_z = 0.473$) is observed. In $PrCo_2Si_2$ and $NdCo_2Si_2$ at 1.5 K, the magnetic ordering is collinear and described by the propagation vector $\mathbf{k} = (0, 0, 1)$. With increasing temperature a change of the structure into an incommensurate, sine wave modulated with $\mathbf{k} = (0, 0, k_z)$ is observed. In $PrNi_2Si_2$, $NdNi_2Si_2$, $NdNi_2Ge_2$, $PrCo_2Ge_2$ and $NdCo_2Ge_2$, sine wave modulated orderings with $\mathbf{k} = (0, 0, k_z)$ exist in the temperature range between 1.5 K and the Néel temperatures.

In this work we report results of ac magnetic susceptibility and neutron diffraction measurements as a function of temperature for $PrCu_2Ge_2$ and $NdFe_2Ge_2$.

Results of magnetic susceptibility, electrical resistance and heat capacity measurements performed for these compounds indicate two phase transitions at 4.2 and 16 K for $PrCu_2Ge_2$ [6] and at 13.5 and 16.4 K for $NdFe_2Ge_2$ [7]. In previous neutron diffraction experiments the magnetic structures of both compounds were determined. In $PrCu_2Ge_2$ the Pr magnetic moments at 1.6 K [8] and 4.2 K [9] form collinear antiferromagnetic structure of the AFI type, consisting of ferromagnetic layers perpendicular to the *c*-axis, which are coupled antiferromagnetically with the sequence of signs $+ - + -$. In $NdFe_2Ge_2$ the Nd moments form collinear antiferromagnetic structure of the AFII type with the sequence of ferromagnetic layers $+ - - +$ [9]. This order is stable up to 13(1) K.

Since the above-mentioned experiments were performed by means of instruments with rather low resolution, we have collected new data for both $PrCu_2Ge_2$ and $NdFe_2Ge_2$ using the E6 diffractometer at the BERII reactor in the Berlin Neutron Scattering Centre. This instrument offers a better incident neutron intensity as well as a much higher resolution.

2. Experimental details

Experiments were carried out on polycrystalline samples, prepared as was reported in previous papers [8, 9].

Magnetic measurements were performed by means of the versatile Lake Shore instrument, model 7225. Temperature dependences of the complex ac magnetic susceptibility $\chi_{ac} = \chi' - i\chi''$, where χ' is the real and χ'' the imaginary component, were measured at a frequency of the driving field $f = 125$ Hz and an amplitude $H_{ac} = 2$ Oe.

Neutron diffraction patterns were obtained on the E6 diffractometer installed at the BERII reactor (Hahn-Meitner Institut, Berlin). The incident neutron wavelengths were 2.437 Å in the case of $PrCu_2Ge_2$ and 2.446 Å in the case of $NdFe_2Ge_2$ and the measurement temperatures ranged from 1.5 to 25 K. The patterns were analysed with the Rietveld profile method, with the use of the FULLPROF programme [10].

3. Results

3.1. $PrCu_2Ge_2$

The temperature dependence of the magnetic susceptibility χ' of $PrCu_2Ge_2$ (see figure 1) reveals at 15.3 K a maximum having typical features for a phase transition from an antiferromagnetic to a paramagnetic state. The observed Néel temperature is lower than the one observed in [6] ($T_N = 16$ K). Also at low temperatures the susceptibility dependence differs from the dependence reported in [6]. These discrepancies may originate from different sample

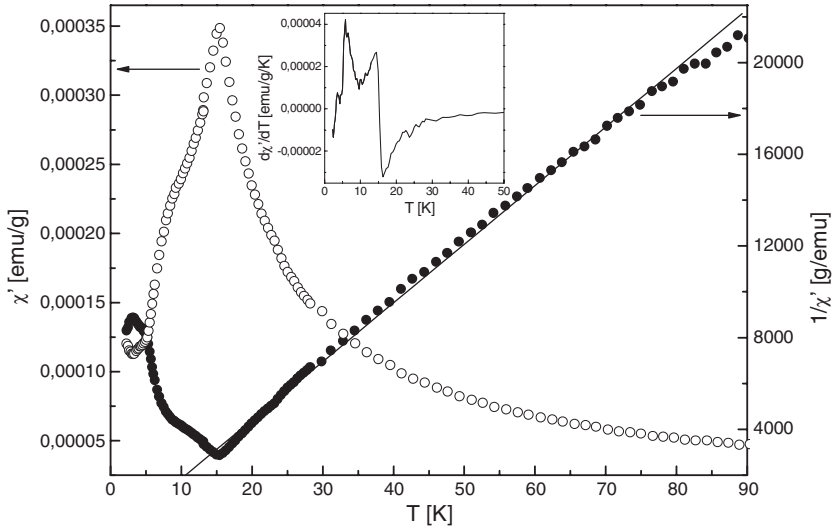

Figure 1. The temperature dependence of the real part of magnetic susceptibility χ' (open symbols) and the reciprocal magnetic susceptibility $1/\chi'$ (filled symbols) of $PrCu_2Ge_2$ measured at 2 Oe and at 125 Hz. The inset shows the derivative $d\chi'/dT$.

preparation procedures: in [6] the sample was melted and annealed at 800 °C and in our case only melted. At low temperatures $\chi'(T)$ has a minimum at 3.3 K and changes slope at 5.3 and 7.7 K. The maximum at 6 K in the temperature dependence of $d\chi'/dT$ is responsible for the transition from an antiferromagnetic collinear ordering to a non-collinear one (both described below). Above 15.3 K the reciprocal magnetic susceptibility $1/\chi'$ satisfies the Curie–Weiss law with a paramagnetic Curie temperature of 2.8 K and an effective Pr magnetic moment equal to 3.61 μ_B, which slightly exceeds the free Pr^{3+} ion value (3.58 μ_B).

Neutron diffraction patterns obtained at 1.5, 5, 9.9, 15.7 and 18.8 K are shown in figure 2. In the three middle sections of this figure the vertical scale has been extended in order to show the magnetic peak details. In the patterns measured at 15.7 and 18.8 K only peaks corresponding to the crystal structure are observed. Nuclear intensities were calculated for the atomic positions characteristic for a structure of the $ThCr_2Si_2$ type:

- the Pr atoms at the 2a sites: (0, 0, 0);
- the Cu atoms at the 4d sites: (0, 1/2, 1/4) and (1/2, 0, 1/4);
- the Ge atoms at the 4e sites: (0, 0, z) and (0, 0, $-z$).

An analysis concerning the atomic order within the 4d and 4e sublattices and general stoichiometry was performed. The results indicated that the stoichiometry was exact in the sample investigated.

The values of the lattice parameters and the z parameter corresponding to the minimum of the reliability factor are listed in table 1.

The 002 and 101 peaks in figure 2 look split. This may be caused by a strain, similar to what was observed in the case of ErCuSi [11].

In the low temperature diffraction patterns (collected at 1.5, 3.3 and 5 K) additional peaks of magnetic origin are indexable on a tetragonal unit cell identical to the chemical one and obey the $h + k + l = 2n + 1$ rule with the 000 reflection absent. This indicates that the Pr magnetic moments situated at the (0, 0, 0) and (1/2, 1/2, 1/2) sites are coupled antiferromagnetically and are pointing along the c-axis. Such an arrangement is usually denoted as AFI type.

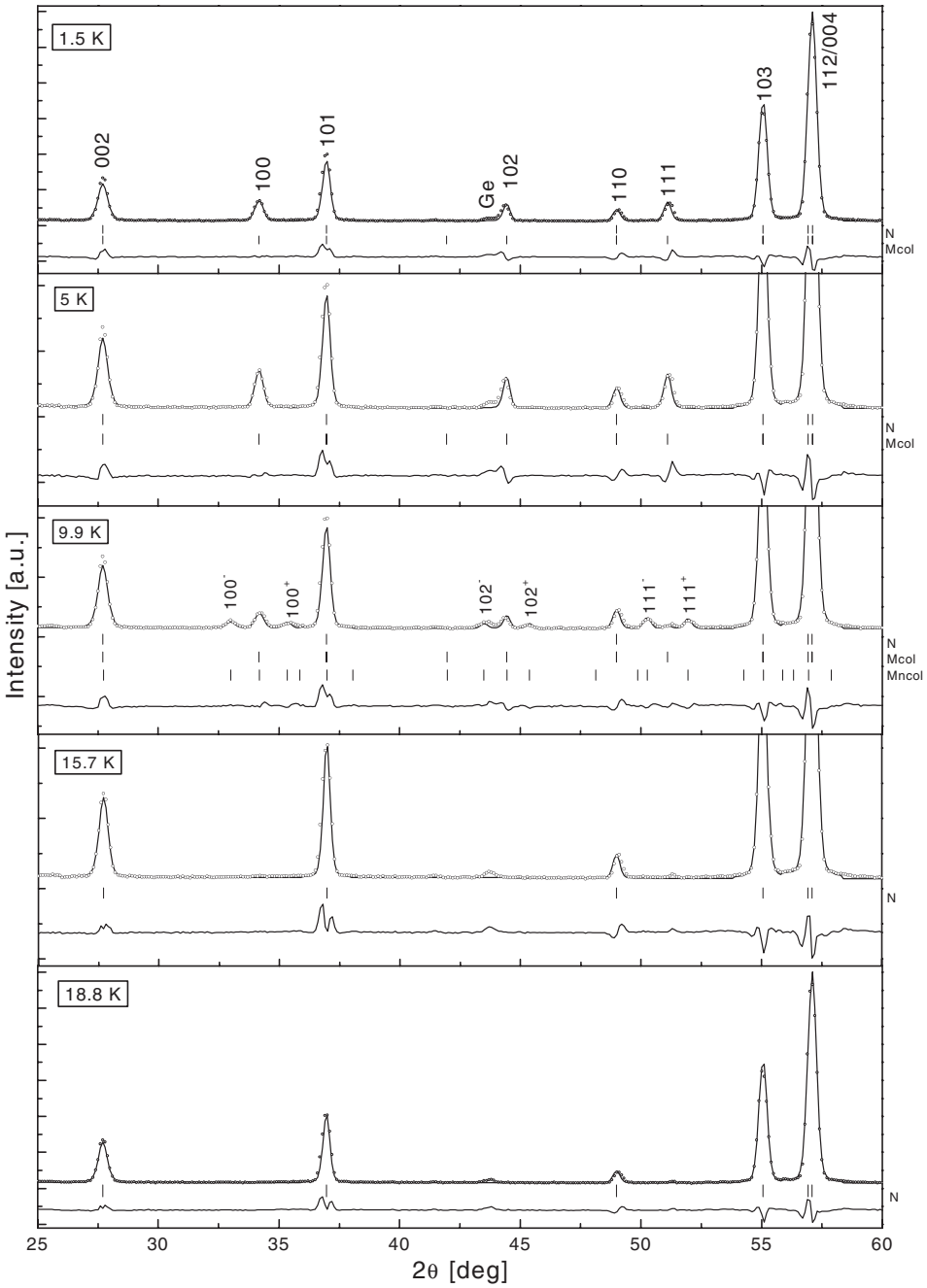

Figure 2. Part of the neutron diffraction patterns of $PrCu_2Ge_2$ collected at 1.5, 5, 9.9, 15.7 and 18.8 K (the 25 °–66 ° 2θ range). The circles represent the experimental points; the solid curves are the calculated profiles for the model crystal and magnetic structures described in the text and the differences between the observed and calculated intensities (at the bottom of each diagram). The vertical bars indicate the Bragg peaks of nuclear (N) and magnetic (Mcol and Mncol for the collinear and non-collinear phases, respectively) origin. The small peak about 43.5 ° originates from the impurity phase (<0.5% of pure Ge). In the three middle sections the vertical scale has been extended in order to show the magnetic peak details.

Table 1. The refined structural and magnetic parameters of $PrCu_2Ge_2$ (space group $I4/mmm$, No 139) obtained from the neutron diffraction data collected at different temperatures. Standard deviations are given in brackets.

T (K)	1.5	3.3	5.0	9.9	15.7	18.8
a (Å)	4.1759(9)	4.176(1)	4.176(1)	4.1763(8)	4.176(1)	4.176(1)
c (Å)	10.268(3)	10.268(3)	10.268(3)	10.268(3)	10.268(4)	10.269(4)
a/c	0.4067	0.4067	0.4067	0.4067	0.4067	0.4067
z	0.3799(4)	0.3799(5)	0.3799(5)	0.3799(4)	0.3798(6)	0.3799(6)
R_{Bragg} (%)	5.87	5.31	4.87	4.97	5.05	4.90
R_{F} (%)	3.10	2.95	2.59	2.74	2.97	2.56
μ_{col} (μ_{B})	2.25(5)	2.26(5)	2.23(5)	0.74(9)	—	—
R_{mcol} (%)	13.7	13.2	14.1	17.5	—	—
μ_{ncol} (μ_{B})	—	—	—	2.60(8)	—	—
k_x	—	—	—	0.0336(5)	—	—
R_{mncol} (%)	—	—	—	22.8	—	—

A different pattern is observed at 9.9 K. Two satellite reflections around each of the magnetic 100, 102 and 111 peaks indicate a transition to a new magnetic phase. The positions of the satellite reflections are described by the wavevector $\mathbf{k} = (0.0336(5), 0, 0)$. This vector corresponds to the modulation equal to 29.8(5) lattice a parameters. For this pattern two magnetic structure models (except for the collinear contribution) were analysed: a sine wave modulated and a helicoidal one. Best agreement with the experimental data has been achieved for the sine wave modulated structure model. This suggests that at a temperature of 10 K $PrCu_2Ge_2$ exhibits complex magnetic structure with two components: the collinear (with the moment values of 0.74(9) μ_{B}) and the modulated one (with the moment values of 2.60(8) μ_{B}).

Figure 3(a) shows parts of the neutron diffraction patterns of $PrCu_2Ge_2$ collected at different temperatures and the temperature dependences of the magnetic peak intensities are presented in figure 3(b). The transition temperatures can be estimated as equal to about 5.5 and 15 K, which is in agreement with the magnetic data. Between these two values the satellites corresponding to the sine wave modulated phase are observed. The angular position of these satellite peaks does not change with changing temperature, which testifies that the period of the modulation is constant.

3.2. $NdFe_2Ge_2$

The temperature dependence of the magnetic susceptibility χ' of $NdFe_2Ge_2$ has a maximum at 16.7 K (see figure 4), which is in a very good agreement with previous results ($T_{\mathrm{N}} = 16.4$ K) [7]. Below the Néel temperature the maximum at 13.5 K in the temperature dependence of $\mathrm{d}\chi'/\mathrm{d}T$ indicates an additional phase transition reported in the previous paper as well [7].

Figure 5 shows neutron diffraction patterns of $NdFe_2Ge_2$ measured at 1.5, 14.5 and 25 K. All strong intensity peaks observed at 25 K obey the $h+k+l = 2n$ rule, which is characteristic for structures of the $ThCr_2Si_2$ type. Analysis of the intensities performed with the use of the FULLPROF programme [10] has shown that in the unit cell the Nd, Fe and Ge atoms are situated at the 2a, 4d and 4e sites of the space group $I4/mmm$, respectively (an analogous atomic order and stoichiometry analysis, as in the case of the Pr sample, was performed). The refined lattice constants and the z position parameter values are listed in table 2.

In the neutron diffraction pattern measured at 1.5 K additional peaks of magnetic origin are observable (see figure 5). These reflections are indexable in a magnetic unit cell that is equal to the chemical one doubled along the c-axis. Analysis of the intensities of these

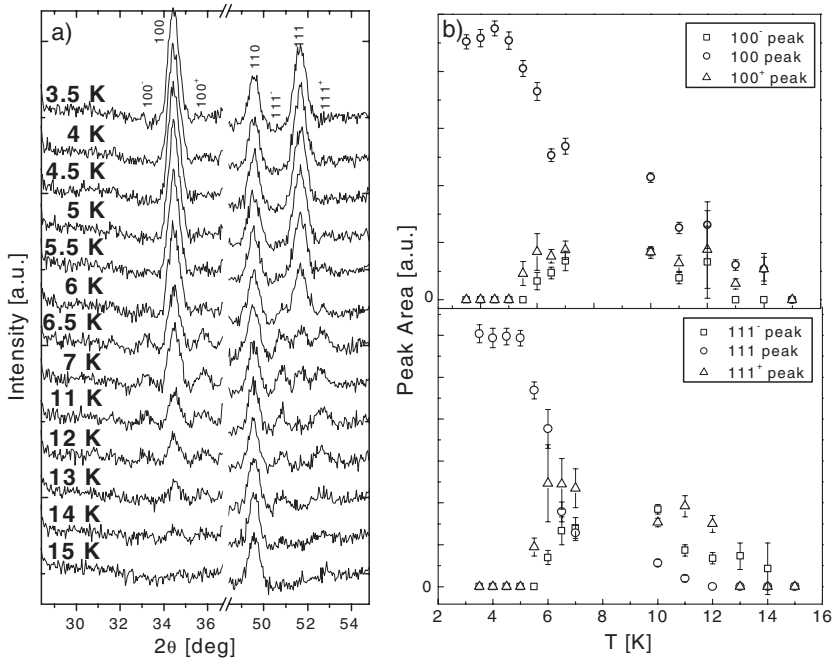

Figure 3. Parts of the neutron diffraction patterns of $PrCu_2Ge_2$ collected at different temperatures (a) and temperature dependences of the magnetic peak intensities (b).

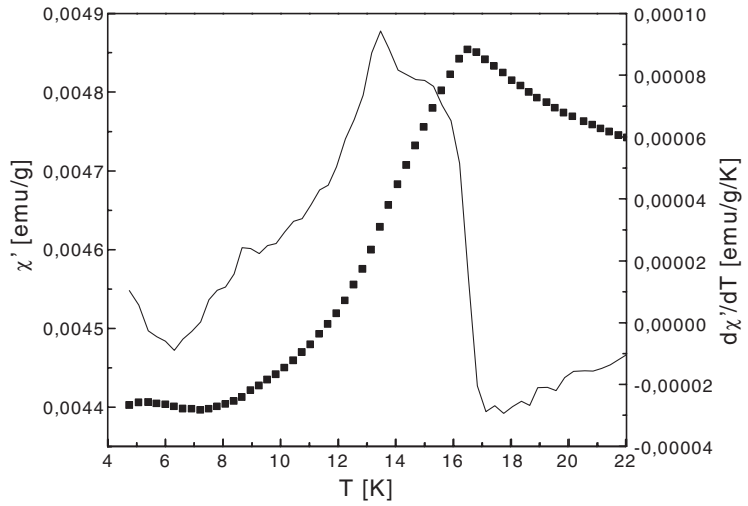

Figure 4. The temperature dependence of the real part of the magnetic susceptibility χ' of $NdFe_2Ge_2$ measured at 2 Oe and at 125 Hz (squares) and its derivative $d\chi'/dT$ (solid curve).

reflections indicates a collinear antiferromagnetic structure of the AFII type, described by the propagation vector $\mathbf{k} = (0, 0, 1/2)$, identical to the one observed previously [8]. The Nd magnetic moment parallel to the c-axis is equal to 3.04(6) μ_B. This value is smaller than the value determined previously (3.38(7) μ_B) [8] but closer to the one determined from the magnetic data (3.2 μ_B) [7].

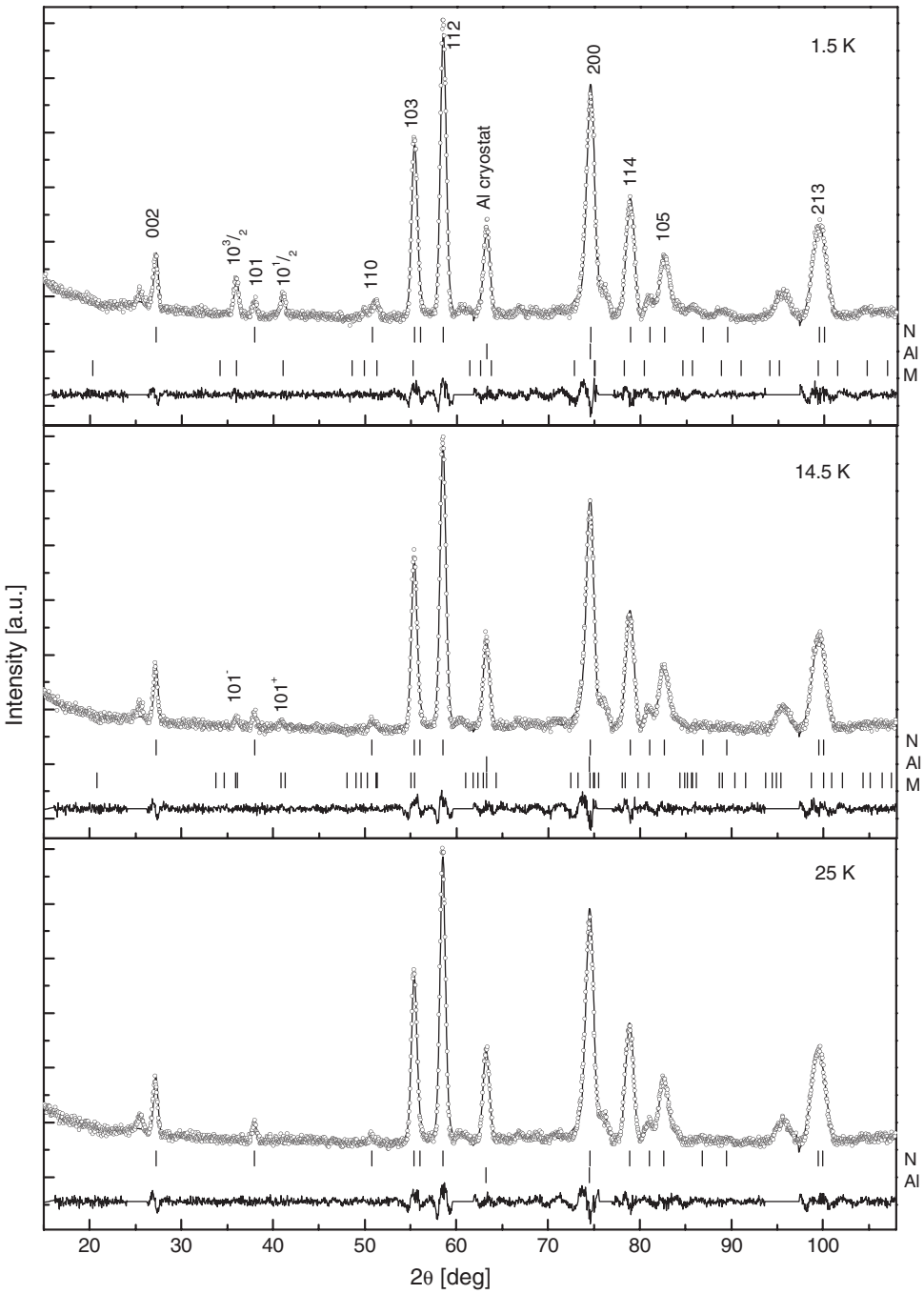

Figure 5. Part of the neutron diffraction patterns of $NdFe_2Ge_2$ collected at 1.5, 14.5 and 25 K. The circles represent the experimental points; the solid curves are the calculated profiles for the model crystal and magnetic structures described in the text and the differences between the observed and calculated intensities (at the bottom of each diagram). The vertical bars indicate the Bragg peaks of nuclear (N) and magnetic (M) origin. The peak about 63 ° originates from the Al cryostat and the peaks at about 25 °, 61 °, 76 ° and 96 ° (excluded in the refinement procedure) from an unidentified impurity phase.

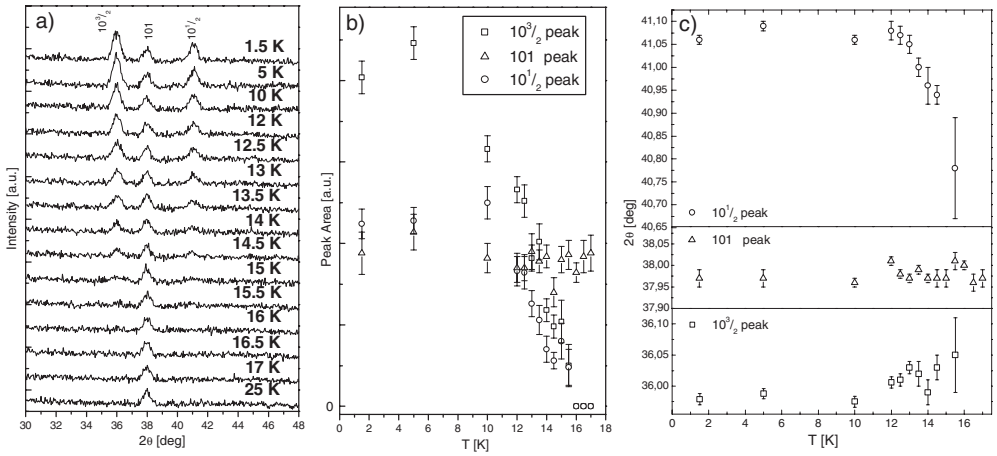

Figure 6. Parts of the neutron diffraction patterns of $NdFe_2Ge_2$ collected at different temperatures (a), temperature dependences of the peak intensities (b) and temperature dependences of the peak positions (c). The $10\frac{3}{2}$ and $10\frac{1}{2}$ peaks change into the 101^- and 101^+ peaks, respectively, at 12 K.

Table 2. The refined structural and magnetic parameters of $NdFe_2Ge_2$ (space group $I4/mmm$, No 139) obtained from the neutron diffraction data collected at different temperatures. Standard deviations are given in brackets.

T (K)	1.5	14.5	25.0
a (Å)	4.0435(5)	4.0437(5)	4.0438(6)
c (Å)	10.435(2)	10.435(2)	10.436(2)
a/c	0.3875	0.3875	0.3875
z	0.3751(3)	0.3748(3)	0.3756(4)
R_{Bragg} (%)	9.85	10.4	9.68
R_{F} (%)	7.52	7.10	5.66
μ (μ_{B})	3.04(6)	2.3(1)	—
k_z	1/2	0.46(1)	—
R_{m} (%)	20.1	35.4	—

The diffraction patterns of $NdFe_2Ge_2$ for 2θ ranging from 30 ° to 48 ° and for temperatures between 1.5 and 25 K are collected in figure 6(a). At 15 K satellites of the 101 nuclear peak appear. With decreasing temperature the intensities of these satellites increase. Temperature dependences of the intensities and angular positions of these satellite peaks are shown in figures 6(b) and (c), respectively. The intensities decrease to zero at 16 K, which corresponds to the Néel temperature value. The angular positions are constant up to 12 K while for higher temperatures they change.

Analysis of the intensities of magnetic reflections at 14.5 K indicates a sine wave modulated structure described by the propagation vector $\mathbf{k} = (0, 0, 0.46(1))$ with the Nd magnetic moments of 2.3(1) μ_{B} parallel to the c-axis.

4. Discussion

Data presented in this work reveal magnetic structure changes related to temperature changes for both compounds investigated. For $PrCu_2Ge_2$ the magnetic moment arrangement changes from a collinear antiferromagnetic structure of the AFI type at low temperatures to a complex

coexistence of the collinear and a sine wave modulated structure near the Néel temperature. For $NdFe_2Ge_2$ the structure changes from a collinear antiferromagnetic of the AFII type at low temperatures to a sine wave modulated structure near the Néel temperature. According to Gignoux and Schmitt [5] such changes arise as a result of the temperature dependences of the free energy for different propagation vectors.

In both compounds, in all magnetic phases the rare earth magnetic moments are parallel to the c-axes. This indicates a very strong anisotropy arising from the crystal electric field effect leading to the Ising-like behaviour. This anisotropy results primarily from the crystal electric field (CEF) splitting of the Hund's rule multiplet. The CEF anisotropy for the tetragonal point symmetry is governed only by the B_2^0 term. The sign of this parameter is connected to the direction of the magnetic moment with respect to the c-axis. For $B_2^0 < 0$ the magnetic moment is parallel to the c-axis whereas for $B_2^0 > 0$ it lies in the basal plane or forms a ϕ angle on the c-axis [12]. Our results concerning the orientation of the magnetic moments parallel to the c-axis suggest that for the compounds investigated B_2^0 is negative.

The values of the rare earth magnetic moments determined in the neutron diffraction experiments differ from the free ion values. In the Pr compound the magnetic moment at 1.5 K equals 2.25(5) μ_B whereas the free ion value equals 3.2 μ_B. The value of the effective Pr magnetic moment determined in this work is equal to 3.61 μ_B, which slightly exceeds the free Pr^{3+} ion value (3.58 μ_B). These discrepancies are most probably connected to the influence of the crystal electric field discussed in detail in [13].

The reduction of the ordered magnetic moment value may be straightforwardly explained by a suitable CEF level scheme. In the PrT_2X_2 compounds the $J = 4$ Hund's rule multiplet of the Pr^{3+} ion is split in the tetragonal environment into three doublets and five singlets. The inelastic neutron diffraction measurements indicate that the ground state is composed of two barely visibly spaced singlets [14, 15]. Different CEF splittings of the low lying singlets observed for $PrFe_2X_2$ (X = Si, Ge) cause large differences in the observed ordered moment values equal to 1.41 μ_B for X = Si and 2.75 μ_B for X = Ge [14]. These results indicate the influence of the X (p electron) elements on the CEF parameter values. In the $PrCu_2X_2$ (X = Si, Ge) compounds the difference in the magnetic moment values is less; they equal 2.51 μ_B for X = Si [8] and 2.25 μ_B for X = Ge (this work). The CEF parameters were determined only for $PrCu_2Si_2$ [16, 17] and are different from those observed in the $PrFe_2X_2$ compounds. The overall energy splitting is of the order of 50 meV for $PrFe_2Si_2$, 25 meV for $PrFe_2Ge_2$ and 11.33 meV for $PrCu_2Si_2$. These results suggest a complex influence of the d (T) and p electron (X) elements on the CEF parameter and magnetic moment values.

The value of the effective Nd magnetic moment in $NdFe_2Ge_2$ was determined in [7] as being equal to 3.4 μ_B, which is near the free Nd^{3+} ion value (3.62 μ_B).

The value of the rare earth magnetic moment determined for the Nd compound at 1.5 K equals 3.04(6) μ_B, which is near the free Nd^{3+} ion value of 3.27 μ_B. A similar value of the Nd moment of 3.02 μ_B at 2 K is observed for isostructural $NdFe_2Si_2$ [18]. The tetragonal crystal field splits the ground state multiplet $^4I_{9/2}$ of the Nd^{3+} ion into five Kramers doublets. The values of the Schottky specific heat data for $NdFe_2Si_2$ suggest that the first excited doublet is at about 90 K above the well-isolated ground state Kramers doublet [18].

The modulation found in $PrCu_2Ge_2$ is different from those discovered in the other PrT_2X_2 compounds, in which the structures are modulated along the c-axes [4, 5] because in $PrCu_2Ge_2$ it is parallel to the a-axis. The existence of the simple AFI-type structure below 5 K agrees with systematics based on the a/c ratio (equal to 0.4067 here) analysis. This analysis performed for a large number of the RT_2X_2 intermetallic compounds [2] indicates that for $a/c > 1/\sqrt{6}$ (=0.4082) an oscillatory magnetic ordering scheme appears while for $a/c < 1/\sqrt{6}$ a collinear order is stable. Our structural data indicate that the a/c ratio

is constant in the measurement temperature range (see table 1). However, in $PrCu_2Ge_2$ an additional commensurate–incommensurate (C–I) transition to the sine wave modulated structure is observed.

In $NdFe_2Ge_2$ the change of the magnetic structure from a collinear form commensurate with the crystal structure at low temperatures to a sine wave modulated incommensurate one near the Néel temperature is similar to that observed in isostructural $PrFe_2Ge_2$ [19, 20] and also in a large number of other rare earth intermetallics [4, 5]. This change can be explained on the basis of the model proposed by Bak and von Boehm [21] which takes into account the interaction between the magnetic moments with first-nearest (J_1) and second-nearest (J_2) planes. The theory gives the following stability condition for a structure of the AFII type, $(+--+)$: $J_2/|J_1| > 1/2$, and in the case of a modulated magnetic ordering with a propagation vector **k** given by $\cos \pi k = -J_1/4J_2$ the ordering is stable if $|J_2/J_1| > 1/4$ [8]. These conditions suggest that the change of the magnetic structure is a result of the change of the interlayer interactions related to temperature changes.

The analysis of the nuclear part of the $PrCu_2Ge_2$ neutron diffraction pattern was performed for two models of the crystal structure:

- firstly, including the mixing of the Cu and Ge atoms between the 4d and 4e sublattices;
- secondly, allowing atomic defects in the Cu and Ge sites.

The minimization procedures carried out for the both models indicated 1:2:2 stoichiometry.

A quality-of-sample estimation is possible, by comparing the lattice parameters: $a = 4.1459(9)$ Å, $c = 10.268(3)$ Å, $a/c = 0.4067(2)$, obtained from our data at 1.5 K to: $a = 4.125(4)$ Å, $c = 10.152(10)$ Å, $a/c = 0.4063(8)$, obtained in [6] at 1.8 K. At 300 K $a = 4.187(2)$ Å, $c = 10.292(8)$ Å, $a/c = 0.4068(5)$ for our sample and $a = 4.149(4)$ Å, $c = 10.211(10)$ Å, $a/c = 0.4063(8)$ at 280 K in [6]. The values of a and c, given in [6], are smaller than those obtained by us. This suggests that that sample did not have a perfect atomic arrangement (unfortunately Sampathkumaran *et al* [6] did not report full structural parameters). According to [12, 22] the changes of the atomic arrangement influences the magnetic interactions.

5. Conclusions

Results of the magnetic and neutron diffraction measurements presented in this work confirm the existence of magnetic phase transitions in $PrCu_2Ge_2$ and $NdFe_2Ge_2$. For $PrCu_2Ge_2$ the AFI-type order transforms to a complex structure at $T_t = 5.5$ K. The values of T_t and T_N are different in different references and equal to 5.5 and 15.0 K in this work, 4.2 and 16 K in [6] and 3.5 and 14.4 K in [23]. This suggests an influence of the preparation procedure (stoichiometry) on the magnetic properties of the samples. In this work a new type of magnetic structure for the RT_2X_2 compounds has been detected: a coexistence of a simple collinear AFI type form and a form sine wave modulated along the a-axis.

For $NdFe_2Ge_2$ the values of T_t and T_N obtained in this work are in agreement with the data in [7] within the limit of the experimental error. In this case the change of the magnetic structure from a collinear commensurate to a sine wave modulated incommensurate one is similar to those observed for other isostructural compounds.

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