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Neutron diffraction study of the magnetic structures of $PrMn_{2-x}Co_xGe_2$ (x = 0.4, 0.5 and 0.8) with a new refinement procedure

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Abstract

The magnetic structures of $PrMn_{2-x}Co_xGe_2$ (x = 0.4, 0.5 and 0.8) with $ThCr_2Si_2$-type structure have been investigated by means of neutron diffraction measurements between 2 and 312 K. We introduced a new refinement procedure to determine the magnetic moments of Pr and Mn sublattices below the rare-earth ordering temperature T_C^{Pr} because of the overlapping of the magnetic reflections of the Pr and Mn sublattices. Rietveld refinements demonstrated that above the Curie temperature an intralayer antiferromagnetic ordering within (001) Mn layers is observed in $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$, while the intralayer antiferromagnetic ordering within (001) Mn layers is found over the whole temperature range for $PrMn_{1.2}Co_{0.8}Ge_2$. Below the Curie temperature the $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$ compounds have a canted ferromagnetic structure with the canting angles 62° and 65° at 2 K, respectively. Below 75 and 70 K, a ferromagnetic ordering of the Pr sublattice is observed along the c-axis for these compounds. Below 70 K, a ferromagnetic ordering of Pr sublattices is found (not detected by magnetic measurements) along the c-axis in $PrMn_{1.2}Co_{0.8}Ge_2$.

1. Introduction

The magnetic properties of ternary rare-earth–transition metal silicides and germanides have been intensively studied over the last 25 years. There has been special interest in compounds

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of the general formula RT_2X_2 (R = rare-earth, T = transition metal, X = Si or Ge) because of the numerous combinations of magnetic interactions [1]. The crystal structure of the naturally layered RT_2X_2 compounds is the body centred tetragonal $ThCr_2Si_2$-type structure with space group $I4/mmm$. The crystal structure is formed by the stacking of atomic layers perpendicular to the c-axis with the sequence R–X–T–X–R. All of the atoms are located at special positions: R at 2a (0, 0, 0), T at 4d (0, 1/2, 1/4) and X at (0, 0, z_X) with z_X always close to 0.38 and consequently the distances mainly depend on the a and c lattice constants [2].

The magnetic properties of these compounds are relatively well known. Neutron diffraction experiments showed that only Mn atoms carry magnetic moments in RT_2X_2 compounds [1], and other transition metals are nonmagnetic. However, the R sublattice orders magnetically at low temperatures (below ∼100 K). The nature of the magnetic coupling within and between the Mn layers in these compounds is closely related to the intralayer Mn–Mn spacing d^a_{Mn-Mn}. Roughly, if $d^a_{Mn-Mn} > 2.87$ Å ($a > 4.06$ Å), the intralayer coupling is antiferromagnetic, and the interlayer coupling is ferromagnetic. When 2.84 Å $< d^a_{Mn-Mn} < 2.87$ Å (4.02 Å $< a < 4.06$ Å), the intralayer in-plane coupling is again antiferromagnetic, but the interlayer coupling is, in this case, antiferromagnetic. In the case of $d^a_{Mn-Mn} < 2.84$ Å ($a < 4.02$ Å), there is effectively no intralayer in-plane spin component, and the interlayer coupling remains antiferromagnetic [3–7].

Due to the strong dependence of the interlayer Mn–Mn exchange interaction on the lattice constant a, studies of compounds such as the $R(1)_{1-x}R(2)_xMn_2X_2$ system or the $RMn_{2-x}T_xX_2$ system provide more information on how the magnetic interactions evolve from one end compound to the other.

When R is a light rare earth (R: Pr, Nd and Sm) in RMn_2Ge_2 compounds, the magnetic moments of the rare-earth sublattice order ferromagnetically below the rare-earth transition temperature T_C^R, and are aligned parallel to the ferromagnetic components of the Mn sublattice. When R is a heavy rare earth (R: Gd, Tb, Dy and Er), the ferromagnetic ordering of the rare-earth sublattice is antiparallel to the ferromagnetic ordering of the Mn sublattice below T_C^R. For RMn_2Ge_2 compounds with a light or heavy rare earth, the magnetic contributions to the (hkl) lines from the rare-earth and Mn sublattices overlap below T_C^R. Owing to this overlapping of the magnetic contributions, one cannot refine the values of the rare-earth moments and Mn moments independently. In this paper, we introduced a new refinement procedure to determine the magnetic moments of the rare-earth and Mn sublattices for RMn_2Ge_2-type compounds. The $PrMn_{2-x}Co_xGe_2$ system was chosen as a model system since the Pr and Mn sublattices order magnetically below T_C^R. Recently, the crystal structure and magnetic behaviour of $PrMn_{2-x}Co_xGe_2$ ($0 \leqslant x \leqslant 1$) have been investigated by means of x-ray powder diffraction and DC magnetic measurements [8]. We focused on the samples with $x = 0.4$, 0.5 and 0.8 to find the magnetic structures on the Pr and Mn sublattices independently and especially to check for the presence or lack of magnetic ordering on the Pr sublattice for the samples with $x = 0.8$.

2. Experimental details

Polycrystalline samples of $PrMn_{2-x}Co_xGe_2$ ($x = 0.4$, 0.5 and 0.8) were synthesized by arc melting on a water-cooled Cu hearth under purified argon gas. The samples were prepared from high purity constituent elements (Pr 99.9%, Mn 99.98%, Co 99.5% and Ge 99.9999%). The starting materials contained ∼3% Mn excess to compensate for the loss due to evaporation during melting and the ingot was melted four times for improved homogeneity. As our argon arc furnace can only load relatively small charges ∼2 g, the samples consisted of a combined mixture of fine powders from four ingots. The quality of the samples was checked

by means of x-ray powder diffraction. The x-ray powder patterns confirm the existence of the tetragonal $ThCr_2Si_2$-type structure. To check the magnetic properties of the prepared samples, magnetic measurements were carried out using a SQUID magnetometer in the temperature range 5 K $\leqslant T \leqslant$ 350 K in an external field of 1.2 kOe.

Neutron diffraction experiments were carried out on powder samples of $PrMn_{2-x}Co_xGe_2$ ($x = 0.4$, 0.5 and 0.8) between 2 and 312 K. The data were collected with the double-axis multicounter diffractometer D1B at the ILL in Grenoble, using a wavelength of $\lambda = 2.52$ Å. The step increment of the diffraction angle 2θ was 0.2°. Measurements were performed at atmospheric pressure in a standard vanadium sample holder. The neutron spectra were analysed with a Rietveld method using the FULLPROF program [9], allowing the simultaneous refinement of structural and magnetic profiles. The scale factor, the background, the lattice constants, a positional parameter z_{Ge} and the Mn and Pr magnetic moment values were refined.

3. Results and discussion

3.1. Details of the Rietveld refinements

Figures 1(a) and (b) show the thermal evolution of the neutron diffractograms of $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.2}Co_{0.8}Ge_2$ as a three-dimensional plot. In all patterns, the peak at the angle of 72° is attributable to the (111) reflection of the vanadium sample holder.

Rietveld refinements on neutron diffraction data confirmed that the samples crystallize in the body centred tetragonal form with space group $I4/mmm$ as expected. In this crystal structure, the rare-earth Mn atoms and Ge atoms occupy the positions 2a (0, 0, 0), 4d (0, 1/2, 1/4) and 4e (0, 0, z_{Ge}) with $z_{Ge} \sim 0.38$. The special position of the Mn atoms on the 4d sites means that nuclear scattering, and consequently scattering due to ferromagnetic ordering of the Mn atoms, only contributes to reflections obeying the (hkl) conditions $h + k = 2n$ and $l = 2n$. For an antiferromagnetic arrangement of the Mn moment within the (001) Mn layers the (hkl) lines obey the conditions $h + k = 2n + 1$ and l is odd. For the conditions $h + k = 2n$ and $l = 2n + 1$, the Mn moments order antiferromagnetically along the c-axis.

Before proceeding to discussing the results, we review briefly the refinement procedure. For $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$, large changes in the local Mn magnetic moments have been detected below 100 K. This is related to the appearance of magnetic ordering in the Pr sublattice. Since the magnetic reflections, according to the ordering of Pr and Mn sublattices, overlap below 100 K, one cannot refine the magnetic moments of Pr and Mn independently. The compounds with $x = 0.4$ and 0.5 show ferromagnetism below the Curie temperature T_C^{inter} and the Pr sublattice orders below T_C^{Pr} [8]. To distinguish the magnetic moments of Pr and Mn sublattices below T_C^{Pr}, we refined the magnetic moments of Mn above 190 K for $x = 0.4$ and above 160 K for $x = 0.5$, in a temperature range where the Pr sublattice does not order and, thereby, determined total magnetic moments of Mn at different temperatures. The temperature dependences were fitted by the following low temperature approximate $M(T)$ functions:

$$M = 2.704 - 0.095 \times 10^{-4}T^2 \qquad \text{for } PrMn_{1.6}Co_{0.4}Ge_2 \tag{1}$$

and

$$M = 2.533 - 0.096 \times 10^{-4}T^2 \qquad \text{for } PrMn_{1.5}Co_{0.5}Ge_2. \tag{2}$$

These functions allow an extrapolation to lower temperatures. For $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$ the total Mn moments were taken from equations (1) and (2), and these values were not refined below 190 and 160 K, respectively. The component of the Mn moment along the c-axis, the canting angle (between the c-axis and the total Mn moment), the Pr moment and the structural parameters were refined.

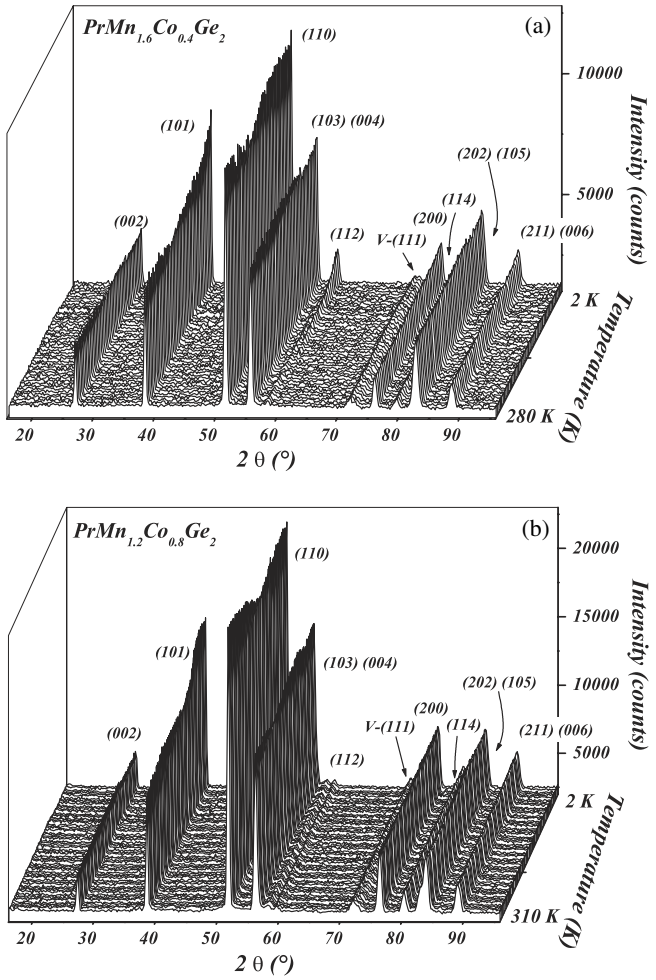

Figure 1. Neutron thermograms of the $PrMn_{1.6}Co_{0.4}Ge_2$ (a) and $PrMn_{1.2}Co_{0.8}Ge_2$ (b) compounds recorded between 2 and 300 K. The principal reflections are indexed. The reflection labelled V(111) is from the vanadium sample holder.

A similar procedure was used for the refinement of $PrMn_{1.2}Co_{0.8}Ge_2$. The $PrMn_{1.2}Co_{0.8}Ge_2$ compound shows antiferromagnetic properties over the whole temperature range [8]. A sudden increase of refinement values of the Mn moments was observed for this compound. This increase is related to the magnetic ordering of the Pr sublattice. First, we refined only Mn moments above 80 K and determined total magnetic moments of Mn at different temperatures. After that, we fitted the Mn moment values obtained, from the refinement. For this compound, the fit equation is

$$M = 2.791 - 0.141 \times 10^{-4} T^2 \qquad \text{for } PrMn_{1.2}Co_{0.8}Ge_2. \tag{3}$$

Below 80 K the total Mn moment was taken from equation (3), and this value was not refined. The Pr moment and structural parameters were refined.

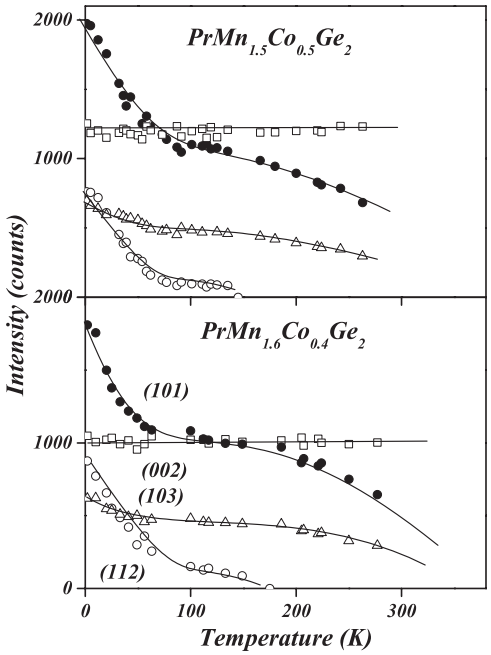

Figure 2. The variations of the intensities of the (101), (103) and (112) magnetic reflections and the (002) nuclear reflections of $PrMn_{1.6}Co_{0.4}Ge_2$ (bottom) and $PrMn_{1.5}Co_{0.5}Ge_2$ (top) with temperature. The lines act as guides to the eye.

3.2. $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$

The two samples show quite similar neutron diffraction patterns. The temperature dependences of the intensities of four characteristic lines are depicted in figure 2. According to the special atomic positions of the Mn atoms (see section 3), the magnetic contributions to the intensities of the (101) and (103) lines are characteristic for the occurrence of an antiferromagnetic component within (001) Mn layers. The increase in the intensity of the (112) line indicates that ferromagnetic (001) Mn layers couple ferromagnetically along the *c*-axis. As shown in figure 2, this intensity increase correlates quite well with the Curie temperature observed by means of magnetic measurements at about 170 K for $PrMn_{1.6}Co_{0.4}Ge_2$ and 140 K for $PrMn_{1.5}Co_{0.5}Ge_2$. The magnetic contribution of the intensity of the (112) line remains larger than the purely nuclear contribution. The second sudden increases in the (112), (101) and (103) lines are probably related to the magnetic contribution of the Pr sublattice at around 75 and 70 K for these two samples. The absence of any contribution to the intensities of the (002) (see figure 2) and the (00*l*) lines with $l = 2n + 1$ clearly show that the ferromagnetic components of the Pr and Mn moments are along the *c*-axis.

The magnetic structures resulting from our refinements of the neutron diffraction patterns are shown in figure 3. These structures are described using the notation developed by Venturini *et al* [4] for the magnetic ordering of the Mn sublattice in RMn_2X_2. Above the Curie temperatures 170 and 140 K for the samples with $x = 0.4$ and 0.5, there is purely antiferromagnetic ordering (AF*l*) within (001) Mn layers (figure 3(c)). A canted ferromagnetic structure (F *mc*) is found below the Curie temperatures. In these samples, below $T_C^{Pr} = 75$ and 70 K, the magnetic moments of the Pr sublattice order ferromagnetically along the *c*-axis and are parallel to the ferromagnetic component of the Mn moments. The values of the Pr and

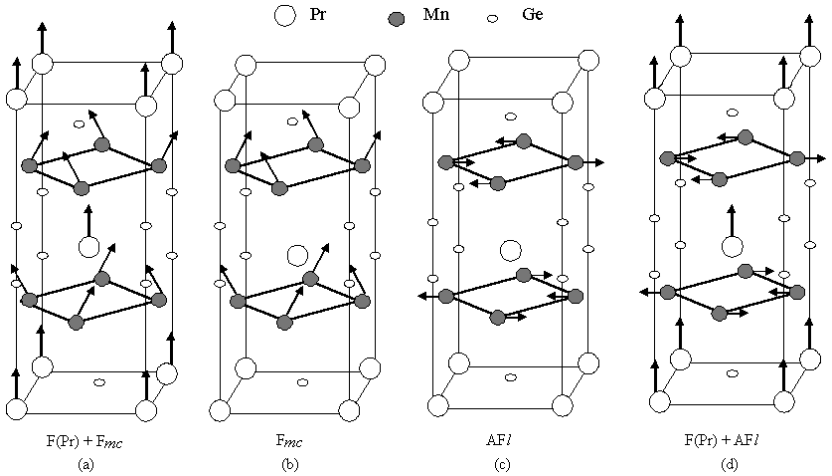

Figure 3. (a) Below T_C^{Pr}, the ferromagnetic structures of the Pr and Mn sublattices of $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$; (b) the canted ferromagnetic structure F *mc* of the Mn sublattice for $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$ below T_C^{inter}; (c) the antiferromagnetic structure AF*l* of the Mn sublattice for $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$ above T_C^{inter} and $PrMn_{1.2}Co_{0.8}Ge_2$ above T_C^{Pr}; (d) below T_C^{Pr}, the ferromagnetic structure of the Pr sublattice and the antiferromagnetic structure AF*l* of the Mn sublattice of $PrMn_{1.2}Co_{0.8}Ge_2$.

Mn magnetic moments and the lattice constants *a* and *c* are shown in figures 4 and 5. The details of the structural and magnetic parameters are also given in table 1.

3.3. $PrMn_{1.2}Co_{0.8}Ge_2$

The temperature dependences of the intensities of four characteristic lines are shown in figure 6. The magnetic contributions to the intensities of the (101) and (103) lines show the presence of intralayer antiferromagnetic ordering within (001) Mn layers (see figure 3(c)). There is no magnetic contribution to the intensity of the (112) line above 70 K. This is evidence of the absence of interlayer ferromagnetic ordering between (001) Mn layers along the *c*-axis. Below 70 K, a strong intensity increase of the (101) line and weak intensity increases of the (103) and (112) lines as compared with the (101) line are observed (see figure 6). These increases of the (101), (103) and (112) lines are due to the ferromagnetic ordering of Pr moments. As for $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$, the (002) line intensity remains constant over the whole temperature range 2–312 K. This indicates that the ferromagnetic components of Pr moments are aligned along the *c*-axis, as shown in figure 3(d).

The temperature dependences of the magnetic moment values of the Pr and Mn sublattices and the lattice constants *a* and *c* are plotted in figures 7 and 8, respectively. The structural and magnetic parameters obtained from the Rietveld refinement at different temperatures are summarized in table 1.

4. Conclusion

Neutron diffraction study has allowed us to determine the magnetic structures of $PrMn_{2-x}Co_xGe_2$ with $x = 0.4$, 0.5 and 0.8. Above the Curie temperatures, the samples with $x = 0.4$ and 0.5 are characterized by the occurrence of the antiferromagnetic structure

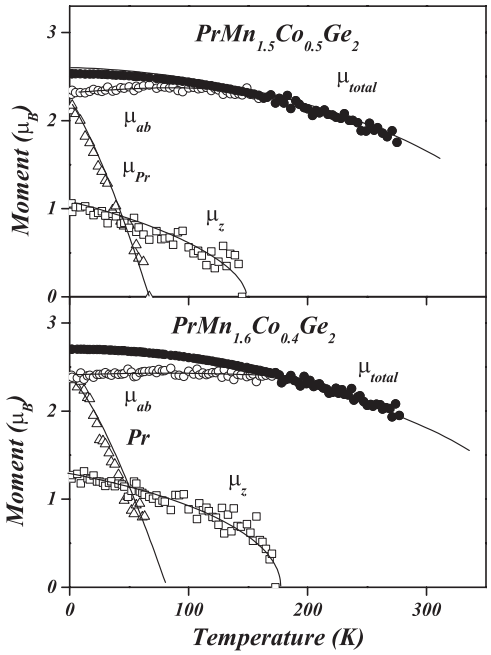

Figure 4. The temperature dependence of the Pr and Mn magnetic moments for the $PrMn_{1.6}Co_{0.6}Ge_2$ (bottom) and $PrMn_{1.5}Co_{0.5}Ge_2$ (top) compounds. μ_{total}: total Mn magnetic moment; μ_{ab}: the Mn moment in (001) Mn layers; μ_z: the Mn moment along the c-axis; μ_{Pr}: the Pr moment along the c-axis.

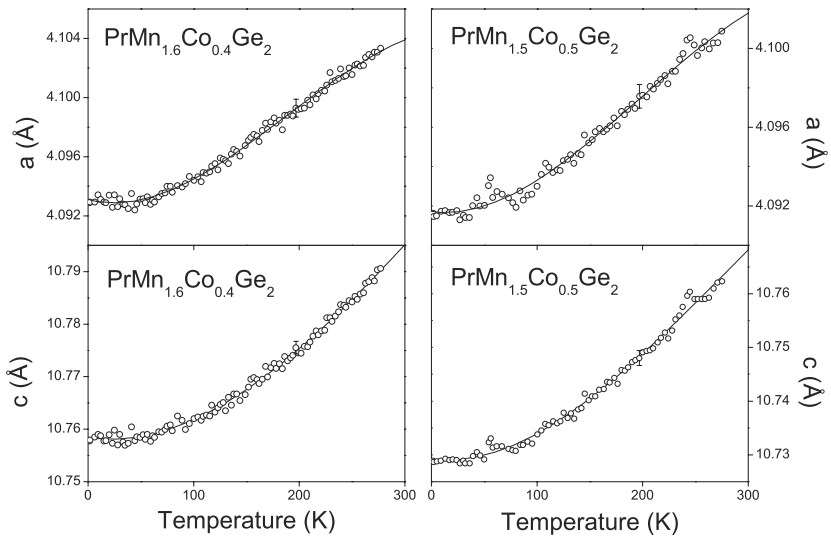

Figure 5. The temperature dependence of the lattice constants a and c for the $PrMn_{1.6}Co_{0.6}Ge_2$ (on left) and $PrMn_{1.5}Co_{0.5}Ge_2$ (on right) compounds determined from the Rietveld refinements.

(AFl) constituted of antiferromagnetic (001) Mn layers. Below the Curie temperatures, the magnetic structure of the Mn sublattice is a canted ferromagnetic structure for these two

Table 1. Structural and magnetic parameters of $PrMn_{1.6}Co_{0.4}Ge_2$, $PrMn_{1.5}Co_{0.5}Ge_2$ and $PrMn_{1.2}Co_{0.8}Ge_2$ as determined from Rietveld refinements of neutron diffraction patterns at different temperatures. The refinements were obtained using the new refinement procedure as discussed in section 3.1. μ_{total}: the total Mn moment; μ_{ab}: the Mn moment in (001) Mn layers; μ_z: the Mn moment along the *c*-axis; μ_{Pr}: the Pr moment along the *c*-axis.

	$PrMn_{1.6}Co_{0.4}Ge_2$					$PrMn_{1.5}Co_{0.5}Ge_2$				
T (K)	277	199	104	52	2	277	200	95	53	2
a (Å)	4.1033	4.0992	4.0947	4.0932	4.0929	4.1012	4.0976	4.0926	4.0930	4.0916
c (Å)	10.7906	10.7746	10.7623	10.7590	10.7579	10.7635	10.7491	10.7322	10.7323	10.7290
z_{Ge}	0.3807	0.3807	0.3804	0.3811	0.3804	0.3807	0.3806	0.3808	0.3801	0.3797
μ_{ab} (μ_{B})	1.353	2.311	2.421	2.444	2.385	1.912	2.137	2.329	2.340	2.300
μ_z (μ_{B})	—	—	0.951	1.096	1.273	—	—	0.749	0.895	1.061
μ_{total} (μ_{B})	1.353	2.311	2.601	2.678	2.704	1.912	2.137	2.446	2.505	2.533
μ_{Pr} (μ_{B})	—	—	—	1.027	2.355	—	—	—	0.819	2.109
Canting angle	90	90	68.559	65.846	61.907	90	90	72.173	69.071	65.243
R_{wp} (%)	12.0	12.2	11.6	13.2	10.6	11.0	10.8	11.2	11.9	9.19
R_{Bragg} (%)	3.32	2.98	2.71	3.08	3.23	3.01	2.57	2.80	2.56	2.07
R_{mag} (%)	1.39	1.87	3.17	3.12	3.97	1.59	1.38	2.08	2.93	2.28
χ^2 (%)	2.08	2.30	2.22	2.74	2.17	2.10	2.29	2.54	3.31	3.95

	$PrMn_{1.2}Co_{0.8}Ge_2$								
T (K)	312	250	201	150	101	79	50	25	2
a (Å)	4.0881	4.0853	4.0838	4.0817	4.0800	4.0791	4.0786	4.0787	4.0793
c (Å)	10.6639	10.6503	10.6426	10.6337	10.6278	10.6244	10.6210	10.6199	10.6213
z_{Ge}	0.3782	0.3778	0.3777	0.3777	0.3780	0.3778	0.3784	0.3783	0.3785
μ_{ab} (μ_{B})	—	1.856	2.247	2.466	2.627	2.813	2.756	2.783	2.791
μ_z (μ_{B})	—	—	—	—	—	—	—	—	—
μ_{total} (μ_{B})	—	1.856	2.247	2.466	2.627	2.813	2.756	2.783	2.791
μ_{Pr} (μ_{B})	—	—	—	—	—	—	1.153	1.337	1.310
Canting angle	—	90	90	90	90	90	90	90	90
R_{wp} (%)	11.6	11.8	11.0	11.1	11.5	11.0	9.79	10.7	9.64
R_{Bragg} (%)	4.41	3.48	3.60	3.55	4.21	4.00	3.39	3.59	3.02
R_{mag} (%)	—	1.45	1.44	1.53	1.52	1.19	4.24	4.70	3.21
χ^2 (%)	4.25	4.07	3.94	3.87	4.37	4.00	3.31	3.98	3.23

samples. At low temperatures, the magnetic ordering of the Pr sublattice is ferromagnetic along the *c*-axis and parallel to the ferromagnetic component of the magnetic moments of the Mn sublattice.

Below 280 K, a conic ferromagnetic structure was observed along the *c*-axis for $PrMn_2Ge_2$ [3]. But the conic ferromagnetic structure is not observed for $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$. It means that the temperature $T_{\text{c/c}}$ of the transition from canted to conic ferromagnetic structure disappears for samples with $x < 0.4$.

In [8], the transition temperatures $T_{\text{C}}^{\text{inter}}$ and T_{C}^{Pr} were obtained from the inflection points of the magnetization curves. The value of the temperature $T_{\text{C}}^{\text{inter}}$ obtained from neutron diffraction measurements is very close to the value obtained from the inflection point. T_{C}^{Pr} is different from the value obtained from the inflection point. According to the neutron diffraction results, the temperature T_{C}^{Pr} is that of the onset of the increase of the magnetization curve when temperature is decreasing.

In the samples of $PrMn_{1.2}Co_{0.8}Ge_2$, the AF*l*-type magnetic structure occurs from 2 to 312 K. The Rietveld refinements demonstrate that below 70 K the magnetic moments of the Pr

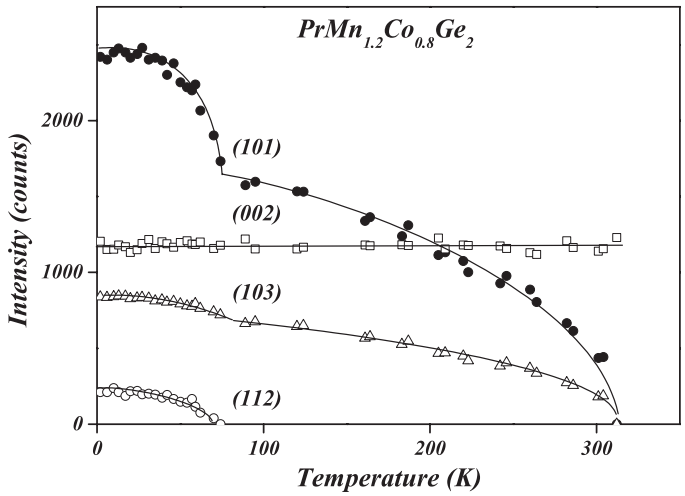

Figure 6. The variation of the intensity for the (101), (103) and (112) magnetic reflections and the (002) nuclear reflection of $PrMn_{1.2}Co_{0.8}Ge_2$ with temperature. The lines acts as a guide to the eye.

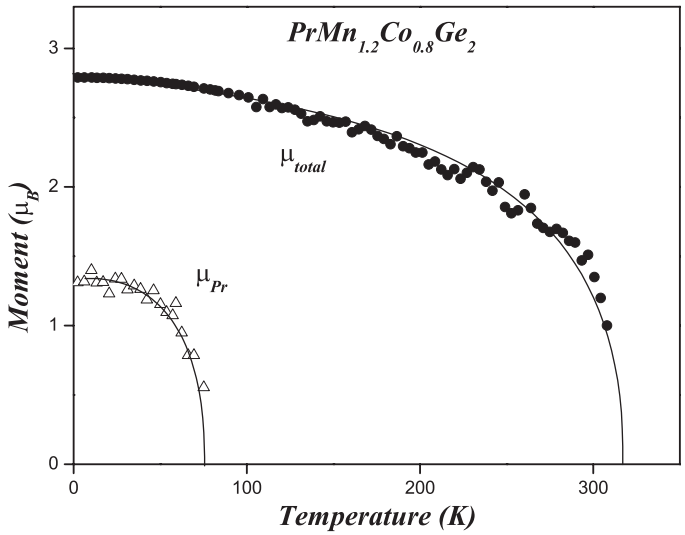

Figure 7. The temperature dependence of the Mn magnetic moment for the $PrMn_{1.2}Co_{0.8}Ge_2$ compounds. μ_{total}: total magnetic Mn moment (equal to μ_{ab}: the Mn moment in (001) Mn layers); μ_{Pr}: the Pr moment along the c-axis.

sublattice order ferromagnetically along the c-axis. This ferromagnetic ordering of Pr moments was not detected from the magnetization measurements [8]. This magnetic structure is a new magnetic structure in the RMn_2X_2 family.

The z_{Ge} values of the compound with $x = 0.8$ versus temperature show anomalies around the transition temperatures T_{C}^{Pr} and $T_{\text{N}}^{\text{intra}}$ as shown in figure 9 while the z_{Ge} values of the compounds with $x = 0.4$ and 0.5 are nearly constant versus temperature. Such anomalies at a magnetic ordering temperature provide information about the underlying magnetostructural couplings in this system, although it is not clear yet how one can explain this behaviour.

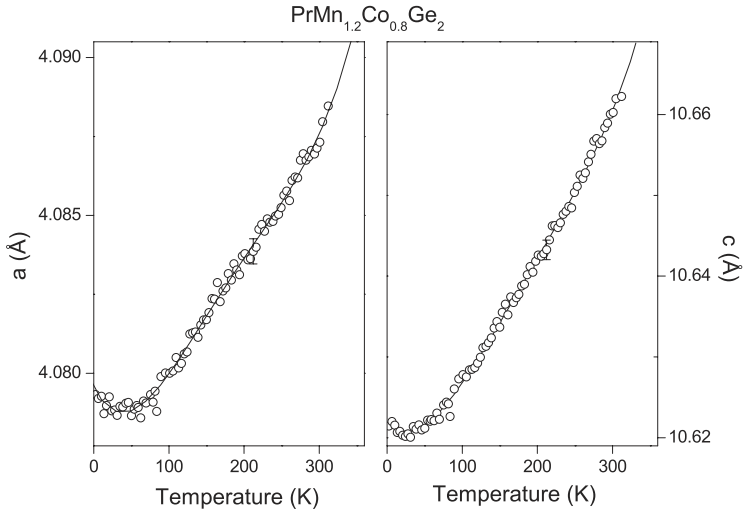

Figure 8. The temperature dependences of the lattice constants a and c for the $PrMn_{1.2}Co_{0.8}Ge_2$ compounds determined from the Rietveld refinements.

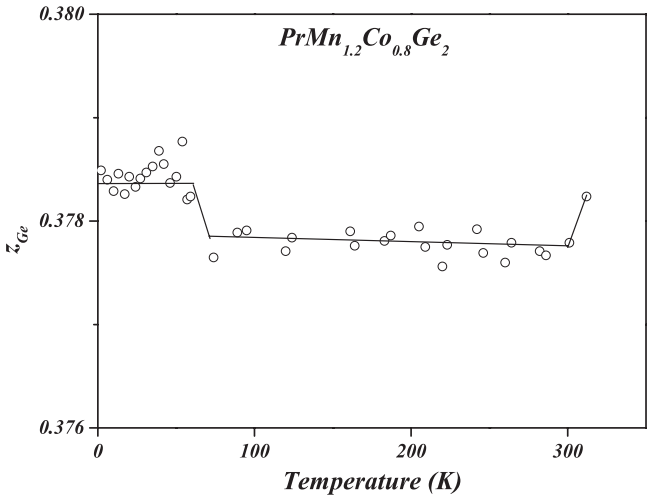

Figure 9. The temperature dependence of the z_{Ge} coordinate obtained from the Rietveld refinements for $PrMn_{1.2}Co_{0.8}Ge_2$.

Recently, the magnetic phase diagrams of $NdMn_{2-x}Fe_xGe_2$ [10], $NdMn_{2-x}Co_xGe_2$ [11], $PrMn_{2-x}Fe_xGe_2$ [12, 13] and $PrMn_{2-x}Co_xGe_2$ [8] were reported. An interlayer ferromagnetic–antiferromagnetic phase transition occurs in $PrMn_{1.4}Fe_{0.6}Ge_2$ and $NdMn_{1.575}Fe_{0.425}Ge_2$, giving rise to the AF$mc$ magnetic structure, while no similar magnetic transition was observed for $NdMn_{2-x}Co_xGe_2$ [11] and $PrMn_{2-x}Co_xGe_2$ [8]. Modifications in chemical bonds are likely to be the source of these differences.

Before applying our new refinement procedure, for $PrMn_{1.6}Co_{0.4}Ge_2$ and $PrMn_{1.5}Co_{0.5}Ge_2$, we observed an unexpected decrease of the Mn moment values below T_C^{Pr} as compared to Mn moment values above T_C^{Pr}. However, for $PrMn_{1.6}Co_{0.4}Ge_2$, we observed an unexpected increase below T_C^{Pr}. These observed increases and decreases of Mn moment

values are not physically correct. A similar decrease of the Mn moment values was also observed for $Nd_xTb_{1-x}Mn_2Ge_2$ below T_C^R [14]. The reasons for these increases and decreases are related to the abundance of the refinable magnetic parameters below T_C^{Pr}. After applying our new refinement procedure, these physically incorrect results were no longer observed. Our results show that the refinement procedure can be used to determine the magnetic structures of rare-earth and Mn sublattices independently of each other for RMn_2Ge_2 compounds.

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