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MAGNETIC STRUCTURES OF PrSi AND NdSi INTERMETALLIC ALLOYS

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The magnetic structure of the PrSi and NdSi intermetallic compounds is reported. It corresponds to a non-collinear ferromagnetic order resulting from the crystal field anisotropy. The magnetic moments lie in the $(a-c)$ miroir plane and their value is $2.8 \pm 0.3\mu_B$/atom and $2.6 \pm 0.3\mu_B$/atom respectively for PrSi and NdSi.

1. INTRODUCTION

THE EQUIATOMIC intermetallic compounds between rare earth and silicon atoms crystallize in the CrB type structure with the space group *Cmcm* or in the FeB type with the space group *Pnma* [1, 2].

Some compounds of this series like TbSi, DySi or HoSi cristallize in both types of structure and the CrB type corresponds to the low temperature phase.

The CrB type RSi compounds with R = Tb to Tm have been already studied by neutron diffraction as well as magnetic and Mössbauer effect measurements [3–5].

However among the RSi (R = La to Er) compounds with the FeB type structure only the magnetic properties of TbSi have been published [6].

In this paper we report the neutron diffraction results for the compounds NdSi and PrSi. The remaining compounds of this series are being studied and their results will be published later.

2. CRYSTALLOGRAPHIC PROPERTIES

The PrSi and NdSi alloys were synthesized by an induction method in a high frequency furnace [4].

The unit cell parameters refined by X-rays measurements are reported in Table 1.

Table 1. Unit cell parameters of NdSi *and* PrSi *refined from X-rays measurements*

	NdSi	PrSi
a (Å)	8.158 ± 0.03	8.243 ± 0.003
b (Å)	3.918 ± 0.003	3.941 ± 0.003
c (Å)	5.887 ± 0.003	5.918 ± 0.003

In the FeB type structure the orthorhombic unit cell contains four formula units, the rare earth ions are located in the (4c) site of the *Pnma* space group and are labelled as follows

(1) in $(x, 1/4, z)$; (2) in $(-x, -1/4, \bar{z})$

(3) in $(1/2-x, -1/4, 1/2+z)$;

(4) in $(1/2+x, 1/4, 1/2-z)$.

Neutron diffraction measurements in the paramagnetic state at room temperature (Fig. 1) allowed us to refine the atomic position parameters x and z of neodymium and silicon atoms in NdSi. The results are given in Table 2.

For the PrSi compound, the refinement was less precise because the praseodymium and silicon atoms have a nuclear scattering length of about the same order of magnitude. The same values of position parameters as in NdSi were then used in all the calculations for the PrSi compound.

3. MAGNETIC PROPERTIES

All the equiatomic rare earth–silicon compounds are antiferromagnetic except PrSi and NdSi which order ferromagnetically at 54 and 46 K respectively [7].

In a magnetic field of 28 kOe, the measured magnetisation on a polycrystalline sample at $T = 4.2$ K is $1.85\mu_B$/atom for NdSi and $1.94\mu_B$/atom for PrSi.

4. MAGNETIC STRUCTURE OF NdSi

The neutron diffraction pattern obtained at 4.2 K (Fig. 1) shows an important enhancement of the nuclear intensities indicating the existence of a ferromagnetic component; in addition new weak magnetic peaks such as (110) (012) forbidden by the space group *Pnma* were also found which implies the coexistence of a small antiferromagnetic component. Under these conditions, the magnetic order described by a propagating vector $\mathbf{k} = 0$, corresponds to a non-collinear ferromagnetic structure.

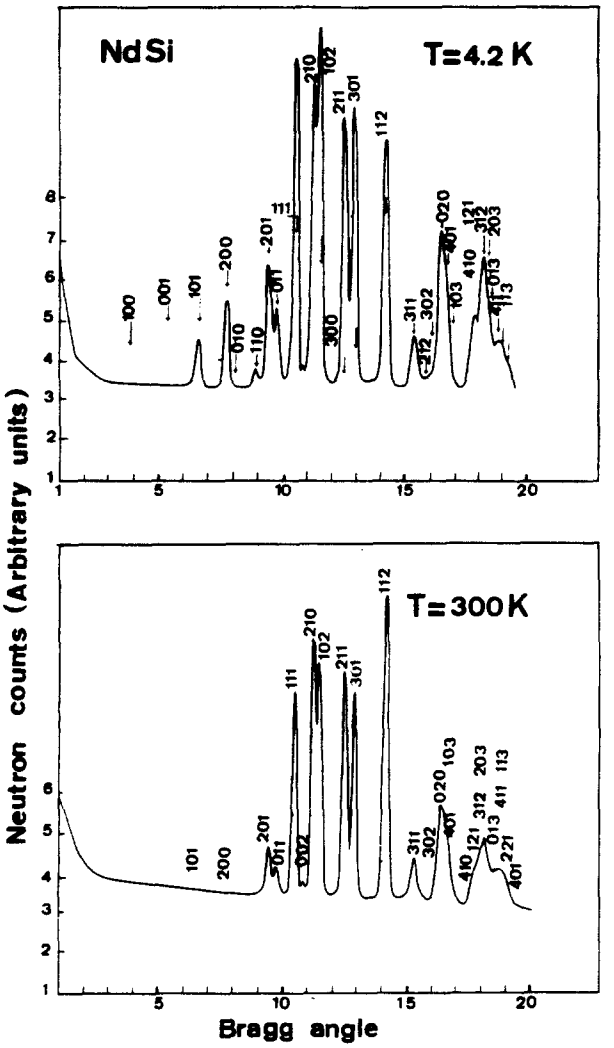

Fig. 1. Neutron diffraction pattern of NdSi at 300 and 4.2 K.

It can be shown, using the representation theory developed by Bertaut [8] that within the *Pnma* space group such a non-collinear ferromagnetic structure exists only for the two models: *CzFx* and *FzCx* where *F* and *C* represent respectively a ferromagnetic (+ + + +) and an antiferromagnetic (+ + − −) arrangement of the four rare earth magnetic moments in the unit cell. The antiferromagnetic component implies that the (010) peak is zero but not the (100) and (001) peaks. The presence of a weak (001) peak and a non-observed (100) peak imply that the *C*-antiferromagnetic component is parallel to the *a*-axis. The magnetic structure observed in NdSi is then of the type *FzCx*. The calculated magnetic intensities are compared to the observed values in Table 2. The magnetic moments lie in the (*a*, *c*)plane and make an angle of 20° ± 2° with the *c*-axis; the moment value at 4.2 K is 2.6 ± 0.3μ_B. This non-collinear

Table 2. Neutron intensities (Barns/unit cell) in the paramagnetic and the ordered states for NdSi

hkl	$I_{N(obs)}$ (300 K)	$I_{N(calc)}$ (300 K)	$I_{M(obs)}$ (4.2 K)	$I_{M(calc)}$ (4.2 K)
100	n.o.	0.0	0.0	0.0
001	n.o.	0.0	Weak	0.91
101	n.o.	0.19	2.45	3.9
200	n.o.	0.05	5.34	4.67
010	n.o.	0.0	n.o.	0.0
110	n.o.	0.0	1.25	2.31
201 011	8.5	10.22	12.01	13.32
111	20.79	19.74	14.05	14.47
002	1.25	0.04	0.0	0.0
210 102	59.3	56.9	19.2	19.46
211 301	62.85	66.1	24.0	23.92
R	6.5%		7%	

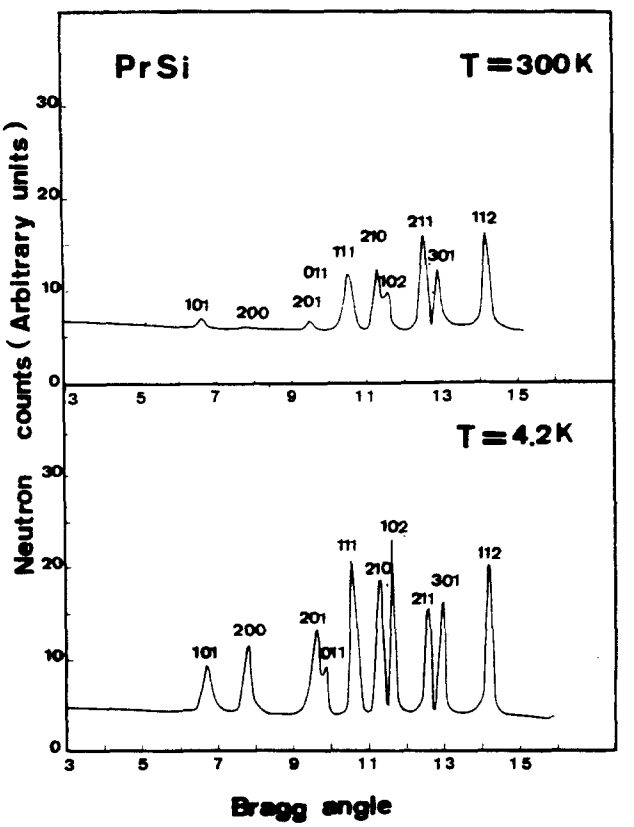

Fig. 2. Neutron diffraction pattern of PrSi at 300 K and 4.2 K

(Fig. 3) gives a reliability factor of 4.5%. The associated magnetic space group *Pn'm'a* has been also found in other compounds such as ErNi and TmNi [9].

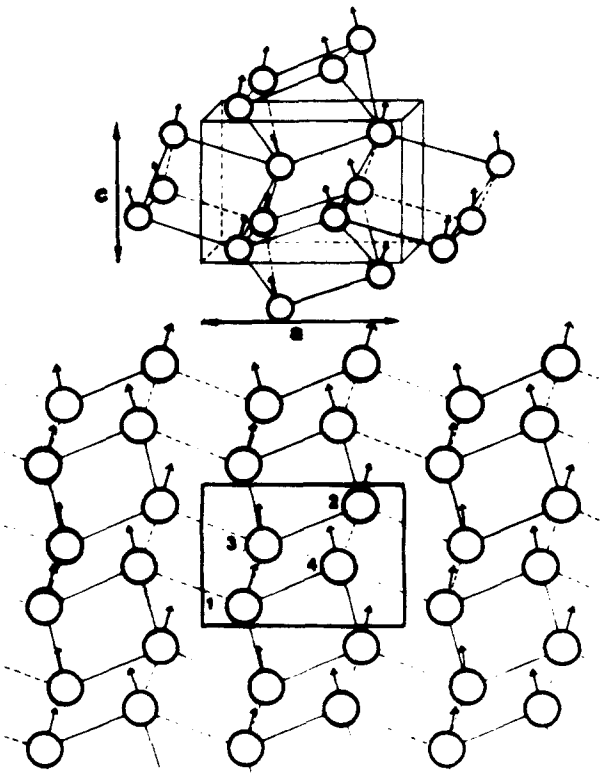

Fig. 3. Magnetic structure of NdSi.

5. MAGNETIC STRUCTURE OF PrSi

For PrSi, a similar diffraction pattern was obtained (Fig. 2) but the additional weak peaks were not observed as in NdSi. This means that if a non-collinear ferromagnetic structure exists, the antiferromagnetic component would be too weak to be observed. All the ferromagnetic models given by the representation analysis were tested. Only the *FxCz* model is in agreement with the observed intensities. The magnetic moments lie in the (a, c) plane but near the a-axis (about 6°) and the moment value at 4.2 K is $2.8 \pm 0.3 \mu_B$. The magnetic space group is in this case *Pnm'a'* like for the DyNi compound (10).

6. DISCUSSION

The above neutron diffraction results have shown evidence for a non-collinear ferromagnetic structure in both PrSi and NdSi intermetallic compounds. Such a magnetic order can only be accounted for by a large anisotropic exchange interactions or (and) a strong crystal field anisotropy. For these compounds a rough crystal field estimation based on a point charge model shows that the single ion anisotropy is sufficient to interpret this non-collinear ferromagnetic order.

As in rare earth perovskites [11] and RNi rare earth–nickel intermetallics [9] the Cs [(a, c)plane] low symmetry of the rare earth sites gives rise to high second order C.E.F. parameter implying that the moments lie along either the b-axis or a direction in the (a, c) plane. In fact for these two compounds the sign of the second order C.E.F. parameters indicates that the magnetic moments lie in the (a, c) plane along the same direction with respect to the corresponding local axes.

As the ions (1) and (2), (3) and (4) are related by inversion symmetry their moments are parallel to each other; whereas (1) and (3), (2) and (4) are related by the two-fold axis parallel to the c-axis, their moment direction are then symmetric with respect to the c-axis.

So we have shown that the non-collinear ferromagnetic ordering, resulting from the crystal field and symmetry considerations, is in good agreement with the neutron diffraction experiments.

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