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MAGNETIC STRUCTURE OF Mn_3Ni_2Si

M. KOLENDA, A. SZYTUŁA

Institute of Physics, Jagellonian University, Cracow, Poland

J. LECIEJEWICZ, A. PAWLUKOJĆ and H. PTASIEWICZ-BĄK

Institute of Nuclear Chemistry and Technology, Warsaw, Poland

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Magnetic susceptibility, X-ray and neutron diffraction measurements have been carried out on Mn_3Ni_2Si – a η' phase with fcc cubic crystal structure. The sample is antiferromagnetic below 215 K exhibiting type II magnetic structure similar to that found in MnO. The total magnetic moment of $(0.57 \pm 0.05)\ \mu_B$ at 78 K was found to be localized on manganese ions only.

1. Introduction

Experimental evidence from neutron diffraction studies on a number of intermetallic systems containing manganese indicates that the ordering of magnetic moments localized on Mn is strongly dependent on the magnitude of the shortest Mn–Mn interatomic distance. This effect is particularly distinct in RMn_2X_2 systems (R = lanthanide or actinide element; X = Si, Ge; tetragonal crystal structure of $ThCr_2Si_2$ type) [1]. A "critical" value of $d_{Mn-Mn} = 0.285$ nm emerged from these studies: when $d_{Mn-Mn} < 0.285$ antiferromagnetic coupling between Mn moments is favoured; when $d_{Mn-Mn} > 0.285$ ferromagnetic order is highly probable.

We turned our attention to a compound Mn_3Ni_2Si which belongs to a group of intermetallic systems called η' phase, exhibiting characteristic cubic crystal structure [2]. It is described by the space group Fd3m. Mn ions are located in 48(f) sites with the following positional parameters:

$x_1, 0, 0;\ 0, x_1, 0;\ 0, 0, x_1;\ \bar{x}_1, 0, 0;\ 0, \bar{x}_1, 0;$

$0, 0, \bar{x}_1;\ \ \frac{1}{4} + x_1, \frac{1}{4}, \frac{1}{4};\ \frac{1}{4}, \frac{1}{4} + x_1, \frac{1}{4};\ \frac{1}{4}, \frac{1}{4},$

$\frac{1}{4} + x_1;\ \frac{1}{4} - x_1, \frac{1}{4}, \frac{1}{4};\ \frac{1}{4}, \frac{1}{4} - x_1, \frac{1}{4};\ \frac{1}{4}, \frac{1}{4}, \frac{1}{4} - x_1;$

+ face − centering translation.

Manganese ions are thus grouped in eight octahedra with short Mn–Mn distances $d_{Mn-Mn} = ax_1\sqrt{2}$ (a is the lattice constant). The octahedra form a diamond type lattice, with nickel ions located in 32 (e) sites and Si in 16 (d) sites filling empty holes in the network of Mn octahedra (see fig. 1). Structural parameters reported in ref. [2] give the shortest Mn–Mn distance to be 0.281 nm, suggesting antiferromagnetic interactions in Mn_3Ni_2Si. It became of interest to undertake a study of its magnetic properties. The results we present in this paper.

2. Experiment

The sample was synthesized by arc melting of stoichiometric quantities of high purity elements in an argon atmosphere. The pellets were afterwards annealed at 800 °C for 100 h and subsequently cooled slowly to room temperature. X-ray diffractograms of a powdered sample taken with CoK_α radiation showed that the crystal structure is cubic and the lattice constant amounts to 1.0756(5) nm, in good agreement with its value reported in ref. [2].

Magnetic measurements were carried out by the Faraday method using an electronic balance

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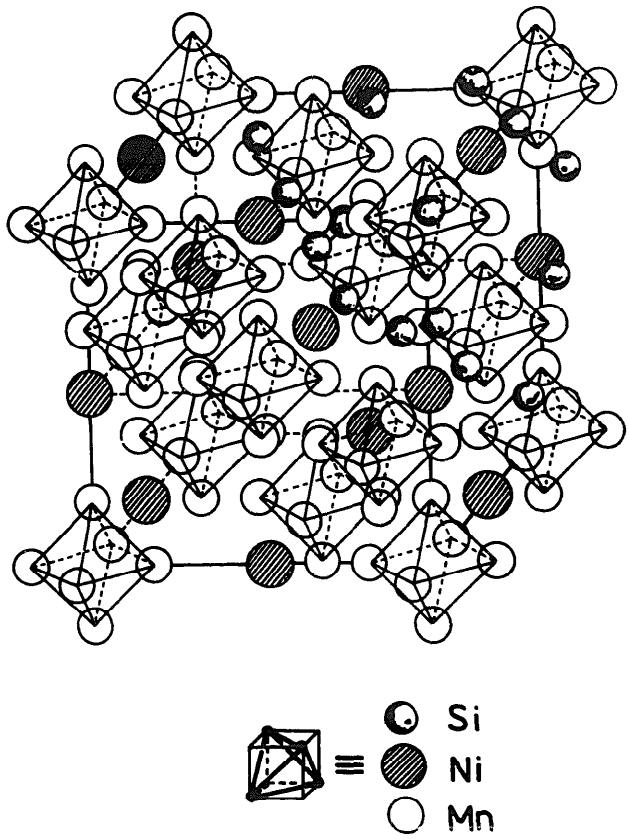

Fig. 1. Crystal structure of Mn_3Ni_2Si.

with the maximum field of 1.25 T. The temperature range covered was from 78 to 300 K.

Neutron diffraction patterns were obtained at 78 and 300 K (see fig. 3). The neutron wavelength was 0.1326 nm. The refinement of nuclear and magnetic intensities was done using a Rietveld line profile analysis method. Nuclear scattering lengths were taken after ref. [3], magnetic form factors of the Mn^{2+} ion after ref. [4].

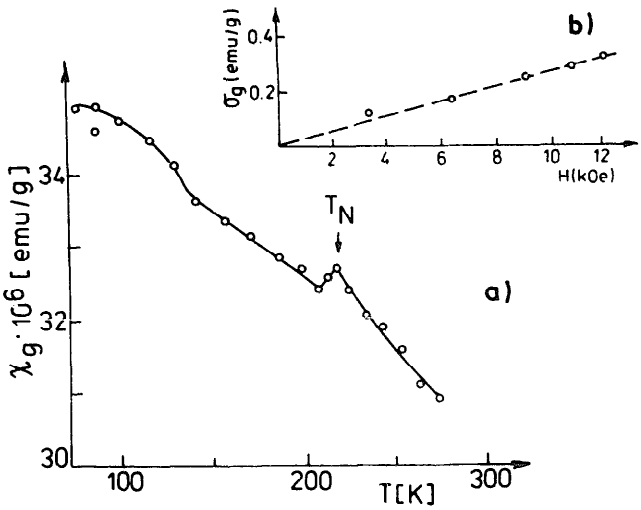

Fig. 2. Magnetic susceptibility against temperature graph (a) and magnetization versus H plot at 80 K for Mn_3Ni_2Si.

3. Results

Our neutron diffraction data confirm the results of earlier X-ray studies [2] that the crystal structure of Mn_3Ni_2Si is face-centered cubic (space group Fd3m). The refinement of neutron intensities taken at 300 K gave:

$$x_{Mn} = 0.185 \quad \text{and} \quad x_{Ni} = -0.835,$$

in fair agreement with the values reported in ref. [2].

The magnetic susceptibility versus temperature plot (fig. 2) displays a peak at 215 K which indicates a transition to an antiferromagnetic state. Neutron diffraction patterns obtained at 78 K confirms this indication – magnetic peaks indexable as $h/2$, $k/2$, $l/2$ are observed suggesting a superstructure resulting from the presence of antiferromagnetic interactions.

The magnetic peaks, observable on the difference pattern at 78 K, were readily indexed on

Table 1
Calculated and observed on neutron diffraction pattern at 80 K magnetic reflection positions for Mn_3Ni_2Si and a comparison of their calculated and observed intensities

h	k	l	Reflection position θ		Magnetic intensity	
			calculated	measured	calculated	measured
$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	3.06	–	0.0	0
$\frac{3}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	5.86	5.8	8.0	8.6
$\frac{3}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	7.72	7.7	34.3	34.6
$\frac{5}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	9.21	9.3	9.4	8.6
$\frac{3}{2}$	$\frac{3}{2}$	$\frac{3}{2}$				
$\frac{5}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	10.55	10.5	23.2	26.9
$\frac{5}{2}$	$\frac{3}{2}$	$\frac{3}{2}$	11.67	11.7	1.5	1.5
$\frac{5}{2}$	$\frac{5}{2}$	$\frac{5}{2}$	12.70	12.8	18.5	21.0
$\frac{7}{2}$	$\frac{1}{2}$	$\frac{1}{2}$				
$\frac{5}{2}$	$\frac{5}{2}$	$\frac{3}{2}$	13.70	13.8	2.3	3.3
$\frac{7}{2}$	$\frac{3}{2}$	$\frac{1}{2}$				
$\frac{7}{2}$	$\frac{3}{2}$	$\frac{3}{2}$	14.60	14.5	7.4	8.6

basis of a magnetic unit cell doubled in three directions i.e. 2a, 2a, 2a (fig. 4), suggesting MnO (type II) magnetic structure. Similar magnetic ordering was found earlier in related Heusler alloys Pd_2MnIn and CuMnSb [5]. From the analysis of magnetic intensities the following model of magnetic ordering was deduced:

- magnetic moments in the octahedra consisting of 6 Mn ions are coupled ferromagnetically;
- the octahedra situated in the (111) plane are ordered ferromagnetically, but are coupled antiferromagnetically to those in the adjacent planes;
- the absence of M $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ reflection indicates

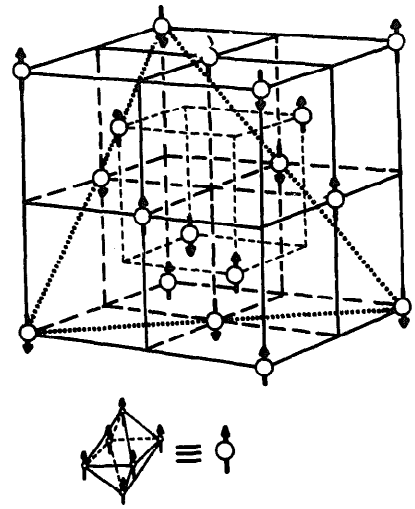

Fig. 4. Magnetic structure of Mn_3Ni_2Si.

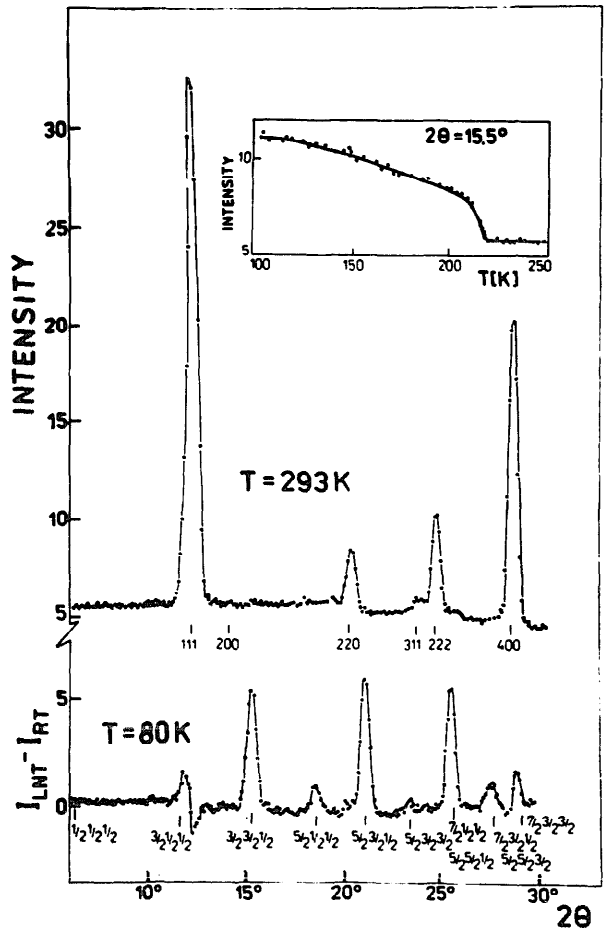

Fig. 3. Neutron diffraction pattern of Mn_3Ni_2Si taken at 300 K and a difference pattern at 80 K showing the magnetic reflections only. The temperature dependence of the $\frac{3}{2}, \frac{3}{2}, \frac{1}{2}$ magnetic peak height is also shown.

that the moments corresponding to octahedra in (0, 0, 0) and $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ lattice points are coupled antiferromagnetically;

- the moments are aligned in the (111) planes;
- each Mn ion carries a magnetic moment of $(0.57 \mp 0.05)\ \mu_B$;
- there is no magnetic moment on the Ni ion.

The magnetic structure of Mn_3Ni_2Si is displayed schematically in fig. 4. Table 1 lists the comparison of magnetic peaks' positions and their intensities observed in experiment and calculated assuming the above model of magnetic order. The plot of temperature against $\frac{3}{2}, \frac{3}{2}, \frac{3}{2}$ peak height gave the Néel point at 215 K (fig. 3).

References

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