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Abstract. The crystallographic and magnetic structures of the compound $Er_2Fe_{14}Mn_3C$ have been investigated by high-resolution powder neutron diffraction at 10 K and 300 K. The results show that this compound crystallizes in the Th_2Ni_{17}-type hexagonal phase (space group $P6_3/mmc$), and the occupancy factors of manganese atoms are 0.082, 0.224, 0.075, 0.478 at the 12k, 12j, 6g and 4f positions respectively. $Er_2Fe_{14}Mn_3C$ is paramagnetic at room temperature, but it shows magnetic uniaxial anisotropy at 10 K. The magnetic moments of all of the magnetic atoms are parallel to the sixfold axis, and the moments of the Er sublattice are ferrimagnetically coupled to that of the transition-metal (Fe, Mn) sublattice.

1. Introduction

Since the nitrides [1, 2] and carbides [3, 4] of R_2Fe_{17} intermetallic compounds (R = rare earth) were obtained with significant hard magnetic properties—hence looking promising for use as permanent magnets—the effect of substitution of non-magnetic elements such as Ga, Al and Si for Fe in R_2Fe_{17} compounds or their nitrides (or carbides) has been widely studied. The addition of these elements has the benefit of crystallizing the 2:17-type phase, and improving the Curie temperature and the anisotropy field, but it reduces the saturation magnetization. Neutron diffraction [5, 6, 7] has been adopted for analysing the occupancy of these substituting atoms and the relationship between the occupancy and magnetic properties. The results show that neither Ga nor Al occupy the 9d site, and that both prefer to occupy the 18h site for low Ga or Al concentration while they prefer to occupy 18f and 6c sites for higher concentrations.

Our recent investigation revealed that the substitution of manganese in R_2Fe_{17} carbides results in rapid decreases of the Curie temperature and the saturation magnetization. In this work, we studied the crystallographic and magnetic structures of the compound $Er_2Fe_{14}Mn_3C$ in detail by using high-resolution powder neutron diffraction.

2. The experiment and refinement

The alloy of $Er_2Fe_{14}Mn_3C$ was prepared by arc melting the appropriate amounts of Er, Fe, Mn and Fe–C alloy according to the chemical composition of $Er_2Fe_{14}Mn_{3.3}C$. The raw

materials were at least 99.9% pure, and the extra 10 at.% of manganese were provided to compensate for the losses during melting. The ingot was then sealed in an evacuated quartz tube and annealed at 600 °C for about a week. The sample was characterized after annealing by chemical analysis, x-ray diffraction and magnetic measurements. The results show that the specimen crystallizes into a Th_2Ni_{17}-type hexagonal phase (space group $P6_3/mmc$) with the Curie temperature $T_C = 225$ K. It should be noted that the sample contains about 2% by volume of secondary phases.

The powder neutron diffraction patterns were collected at 10 K and 300 K on a multi-detector system installed at Neutron Guide Tube No 2 (NGT-2), at the Materials Science Research Centre attached to the Multi-Purpose Research Reactor, G A Siwabessy (RSG-GAS), Serpong, Indonesia. Neutrons with a wavelength of 1.8215 Å were monochromatized by using 'hot-pressed' Ge(331) single crystal. About 70 mg of the powdered sample were placed into a vanadium can of diameter 5 mm and height 6 mm and then this was stored in a cryostat. The temperature was kept at a certain constant value (±0.1 K) during the measurements. All 2800 of the readings (with 2θ from 10° to 150°) were collected by a stepped counting method (with 0.05° per step). The absorption correction for a cylindrical sample was applied to the measured data.

The data were analysed using the Rietveld method implemented via the program RIETAN [8]. The parameters of the crystallographic structure of Th_2Ni_{17}-type rare-earth–iron compounds [9] were used as the initial input in the refinement, and it was assumed that the carbon atoms [10] were at 6h sites. The magnetic form factors were calculated according to equations given in the *International Tables* [11]. As is usual in Rietveld analysis, the global parameters including the background parameters and scale factor were refined in the first cycle, and this was followed by the refinement of the profile parameter and lattice constants in the second cycle. In the subsequent cycles, all of the parameters including the magnetic moments also were refined.

Data collected at 300 K were used to determine the occupancy factors of all of the atoms. These factors should be very accurate because the magnetic diffraction is absent at room temperature, and the neutron diffraction length of the manganese atom is especially negative. Manganese atoms were assumed to replace Fe at four sites, 12k, 12j, 6g and 4f, and the occupancy factors were refined according to chemical concentration and with the linear constraint condition. The total occupancy factor of Fe and Mn atoms on each of the four sites equals 1. Data collected at 10 K were then applied to determine the magnetic structure with the occupancy factors derived from the refined results for data recorded at 300 K.

3. Results and discussion

In figure 1 the observed neutron diffraction intensities (dots) and the fitting-calculated intensities (lines) of $Er_2Fe_{14}Mn_3C$ are shown. The crystallographic and magnetic parameters are listed in table 1. The rather large value of R_{wp} (11.41% at 300 K; 13.93% at 10 K) could be caused by the large value of R_e (5.53% at 300 K, 5.85% at 10 K). We find that there are Mn atoms on all four transition-metal (TM) sites with the different occupancy factors of 0.082, 0.224, 0.075, 0.478 for the sites 12k, 12j, 6g and 4f respectively. Mn obviously 'prefers' to replace iron on the dumb-bell site 4f, with 12j being the second choice. Er atoms occupy 2b and 2d sites with the same occupancy factor of 1.000.

There is no additional peak in the diffraction pattern at 10 K as compared with the pattern at 300 K (see figure 1), which means that there is no complex magnetic structure in $Er_2Fe_{14}Mn_3C$. Therefore, we can assume that the magnetic moments of all of the magnetic
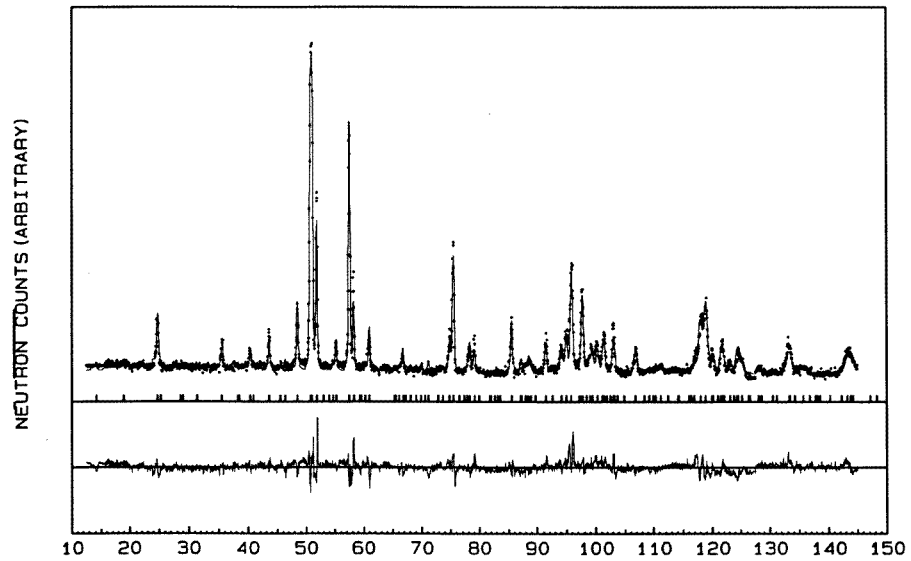
(a)
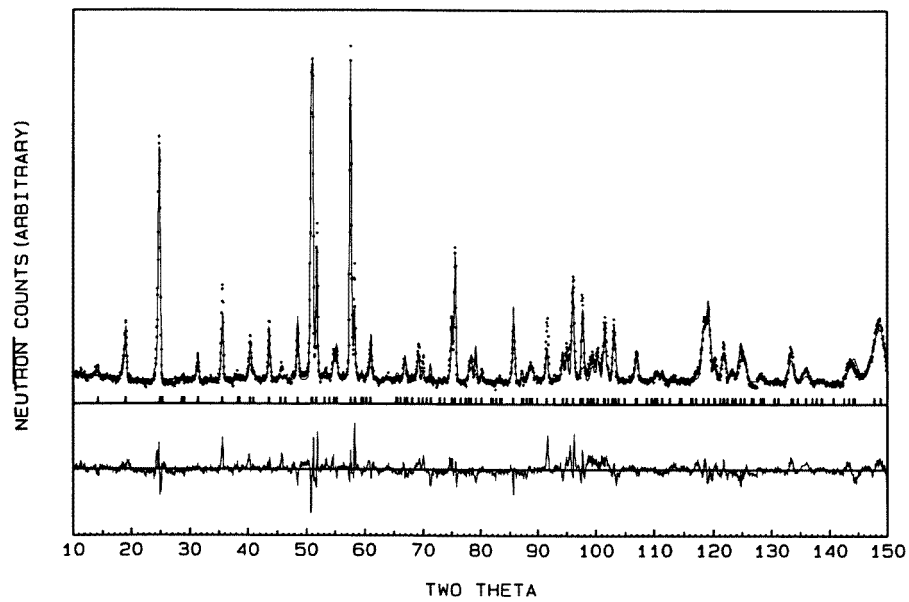
(b)

Figure 1. The collected (dots) and calculated (lines) patterns of the high-resolution powder neutron diffraction (a) at 300 K, and (b) at 10 K. The calculated positions are indicated at the bottom. The differences between the observed data and calculated values are also given.

atoms align collinearly. The refined results show that the moments of all of the magnetic atoms are parallel to the sixfold axis; in other words, the compound $Er_2Fe_{14}Mn_3C$ possesses magnetic uniaxial anisotropy at 10 K. The moments of the Er sublattice are ferrimagnetically

Table 1. The crystallographic and magnetic parameters of $Er_2Fe_{14}Mn_3C$ investigated by high-resolution powder neutron diffraction at 10 K ($R_{wp} = 13.93\%$, $R_e = 5.85\%$). The occupancy factors were determined by the refinement of the data collected at 300 K, with paramagnetism. $a = b = 8.4865(9)$, $c = 8.3384(8)$.

Wyckoff symbol	Atom	Occupancy factor	x/a	y/b	z/c	B (Å^2)	M (μ_B)
2b	Er	1.00	0	0	1/4	0.35(1)	−7.58(69)
2d	Er	1.00	1/3	2/3	3/4	0.35(9)	−8.82(72)
12k	Fe	0.91(8)	0.168(0)	2x	0.985(7)	0.45(8)	1.34(42)
	Mn	0.08(2)					
12j	Fe	0.77(6)	0.329(1)	0.955(2)	1/4	0.45(4)	0.52(07)
	Mn	0.22(4)					
6g	Fe	0.92(5)	1/2	0	0	0.45(8)	1.77(02)
	Mn	0.07(5)					
4f	Fe	0.52(2)	1/3	2/3	0.110(5)	0.45(1)	0.88(05)
	Mn	0.47(8)					
6h	C	0.33(3)	0.841(2)	2x	1/4	0.73(5)	

coupled to that of the TM sublattice. Regrettably, the moment of manganese cannot be distinguished from the average moment of the TM site in the program because Mn and Fe randomly occupy the same site. Thus, the moments of the TM sites shown in table 1 are the average moments of atoms composed of Fe and Mn.

Acknowledgments

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