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A high-resolution neutron study of $Y_2Fe_{17-x}Ga_x$ ($x = 5, 7$)

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Abstract. The crystallographic and magnetic structures of $Y_2Fe_{17-x}Ga_x$ ($x = 5, 7$) compounds at room temperature have been refined by Rietveld analysis of the high-resolution neutron powder diffraction data. The analysis indicates that $Y_2Fe_{17-x}Ga_x$ ($x = 5, 7$) compounds have Th_2Zn_{17}-type structures ($R3m$). The Ga atoms only occupy preferentially 18h and 18f sites in $Y_2Fe_{12}Ga_5$ compounds, but Ga atoms occupy preferentially 18h, 18f and 6c sites in $Y_2Fe_{10}Ga_7$ compounds. The magnetic moments of all of the Fe atoms of $Y_2Fe_{12}Ga_5$ compounds display ferromagnetic alignment, lie in the plane perpendicular to the sixfold axis and exhibit planar magnetic anisotropy. In contrast, the magnetic moments of all of the Fe atoms of $Y_2Fe_{10}Ga_7$ compounds lie parallel to the sixfold axis and exhibit easy-axis magnetic anisotropy.

1. Introduction

In recent years, various magnetic materials based on R_2Fe_{17} have been extensively investigated in order to improve the magnetic properties. Two drawbacks restrict their application as permanent magnets: none of them exhibits an easy-axis magnetocrystalline anisotropy at room temperature and the magnetic ordering temperature T_c of the compounds is relatively low. Therefore, many efforts have been made with the aim of solving such problems by either substituting other elements for iron in the R_2Fe_{17} structure or introducing interstitial atoms into the materials. It was found that the substitutions of the non-magnetic elements Al and Ga for Fe will increase the Curie temperature and change the planar anisotropy to easy-axis anisotropy [1–5]. Hu *et al* [1] and Yelon *et al* [5] reported on the magnetic properties and structures of $Tb_2Fe_{17-x}Ga_x$ compounds and $Nd_2Fe_{17-x}Al_x$ compounds respectively at room temperature. Cheng *et al* [6] reported that the substitution of Ga has a significant effect on the magnetic anisotropy of $Gd_2Fe_{17-x}Ga_x$ compounds. These papers reported that the easy magnetization directions of those compounds change from the basal plane to the *c*-axis with increasing non-magnetic Ga and Al concentrations (except for when R = Sm). This paper reports on the refined magnetic structures of $Y_2Fe_{17-x}Ga_x$ ($x = 5, 7$) at room temperature obtained from the high-resolution powder diffraction patterns. The refined structures show that the magnetic moments of all of the iron atoms of the $Y_2Fe_{10}Ga_7$ compound are parallel to the sixfold axis and exhibit easy-axis anisotropy. In contrast, the $Y_2Fe_{12}Ga_5$ compounds exhibit planar anisotropy, although the Y sublattices have not made any contribution to the magnetocrystalline anisotropy. This indicates that the Fe–Ga sublattices (9d, 18h, 18f, and 6c) completely determine the

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magnetocrystalline anisotropy (uniaxial or planar) of the compounds. The detailed analysis of the structure demonstrates that the non-magnetic atoms Ga with the larger occupancies on 18f and (or) 6c sites will overcome the drawbacks of R_2Fe_{17} compounds, and exhibit uniaxial anisotropy.

2. Experiments and refinements

The samples $Y_2Fe_{17-x}Ga_x$ were prepared by arc melting high-purity Y (99.9% purity), Fe (99.95% purity) and Ga (99.999% purity) in an argon arc furnace. The ingots were then ground to yield powder samples. The samples were examined by x-ray diffraction. The results show that the samples have Th_2Zn_{17}-type structures (space group $R3m$).

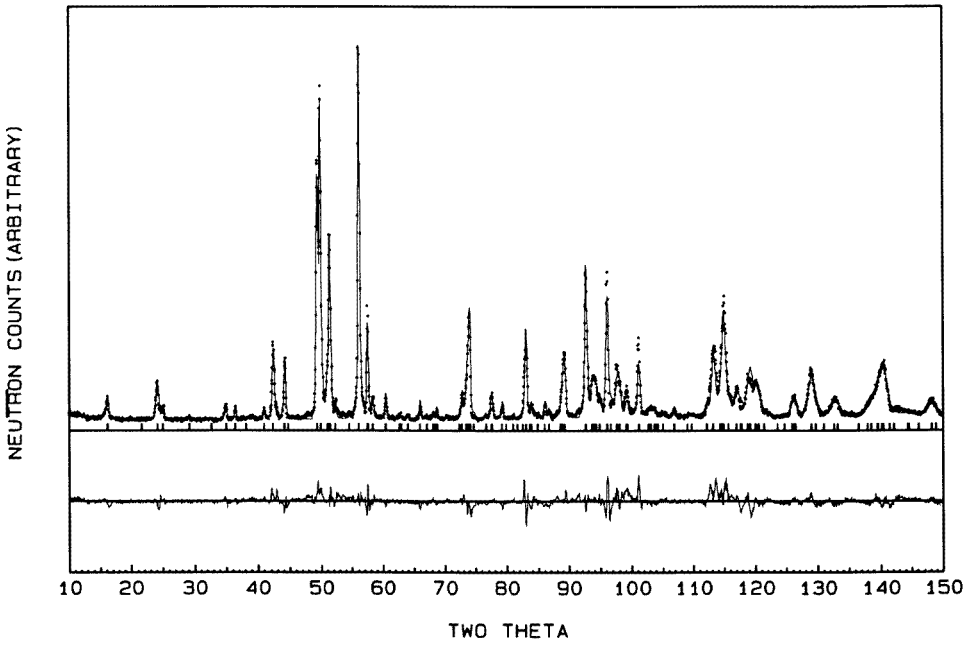

Figure 1. A high-resolution powder diffraction pattern for $Y_2Fe_{10}Ga_7$ at room temperature. The observed and calculated profiles are given by the dotted and solid curves respectively, and the calculated positions are indicated at the bottom. The differences between the observed and calculated data are also given.

The neutron diffraction investigations were performed at room temperature by using a high-resolution powder diffractometer with a multi-detector system installed at Neutron Guide No 2 (NGT-2), at the Material Science Research Centre attached to the Multi-Purpose Research Reactor, GA Siwabessy (RSG-GAS), Serpong, Indonesia. The incident-neutron wavelength was 1.8215 Å monochromatized by the Ge(331) single crystal. The total (3200 points) data were collected by step scanning at 0.05° intervals over the angular range 2.5–162.5° with 32 detectors. The diffraction pattern of the $Y_2Fe_{10}Ga_7$ compound is shown in figure 1. The collected data were analysed via the Rietveld structure refinement program RIETAN [7]. The parameters of the crystallographic structure of Th_2Zn_{17}-type rare-earth–iron compounds [8] were used to start the refinement. It was assumed that Fe and Ga

atoms occupy simultaneously 9d, 18h, 18f, and 6c sites with the linear constraint condition: the sum of the occupancies of Fe and Ga atoms at each of the four sites equals 1.0. The possibilities of alignment of magnetic moments of Fe atoms either parallel or perpendicular to the sixfold axes were checked extensively in subsequent refinements.

Table 1. Crystallographic and magnetic parameters of $Y_2Fe_{12}Ga_5$ (space group $R3m$). a = 8.6694(5) Å; c = 12.634(1) Å.

Atom	Occupancy	x	y	z	B (Å^2)	M (μ_B)
Y	1.00	0	0	0.3415(5)	1.1(2)	
Fe(9d)	1.00	0.5	0	0.5	0.8(1)	1.8(2)
Fe(18h)	0.58(2)	0.5027(5)	0.4973(5)	0.1555(5)	0.7(1)	2.3(2)
Fe(18f)	0.60(2)	0.2894(5)	0	0	0.3(1)	1.6(2)
Fe(6c)	1.00	0	0	0.0972(5)	1.1(2)	1.4(2)
Ga(18h)	0.42(2)	0.5027(5)	0.4973(5)	0.1555(5)	0.7(1)	
Ga(18f)	0.40(2)	0.2894(5)	0	0	0.3(1)	

Table 2. Crystallographic and magnetic parameters of $Y_2Fe_{10}Ga_7$ (space group $R3m$). a = 8.7255(5) Å: c = 12.617(1) Å.

Atom	Occupancy	x	y	z	B (Å^2)	M (μ_B)
Y	1.00	0	0	0.3435(5)	0.5(1)	
Fe(9d)	1.00	0.5	0	0.5	0.3(1)	1.8(2)
Fe(18h)	0.75(2)	0.5007(5)	0.4993(5)	0.1546(5)	0.4(1)	2.1(2)
Fe(18f)	0.35(2)	0.2974(5)	0	0	0.8(1)	1.5(2)
Fe(6c)	0.20(2)	0	0	0.1039(5)	0.6(1)	1.3(2)
Ga(18h)	0.25(2)	0.5007(5)	0.4993(5)	0.1546(5)	0.4(1)	
Ga(18f)	0.65(2)	0.2974(5)	0	0	0.8(1)	
Ga(6c)	0.80(2)	0	0	0.1039(5)	0.6(1)	

The final refined crystallographic and magnetic parameters of both compounds are listed in tables 1 and 2. The magnetic moments of all of the iron atoms display ferromagnetic coupling, and exhibit easy-plane and easy-axis anisotropy for $Y_2Fe_{12}Ga_5$ and $Y_2Fe_{10}Ga_7$ compounds respectively. The occupancies of Ga atoms on 18h, 18f and 6c sites coincide with the Ga contents in the two compounds.

3. Discussion

It is known that the net magnetocrystalline anisotropy in rare-earth–iron intermetallic compounds is determined by the sum of the iron sublattice and rare-earth sublattice anisotropies. In R_2Fe_{17} compounds, the total magnetization of the four iron sublattices exhibits planar anisotropy. The rare-earth sublattice anisotropy can be described by the product of the second-order Stevens coefficient α_J and the second-order crystal parameter A_{20} on the basis of the single-ion model [9]. A negative value of the product $\alpha_J A_{20}$ will give a uniaxial contribution from the rare-earth sublattice to the total anisotropy. In the case of R_2Fe_{17} compounds, the second-order crystal parameters $A_{20} < 0$ and the second-order Stevens coefficients $\alpha_J < 0, \alpha_J > 0, \alpha_J = 0$ correspond to the rare-earth elements R = (Nd, Tb, ... except for Sm), R = Sm, R = Y respectively. Therefore Y_2Fe_{17}, Ho_2Fe_{17} and

Tb_2Fe_{17} exhibit easy-plane anisotropy; in contrast $Sm_2Fe_{17-x}Ga_x$ (with a small Ga content x) exhibits easy-axis anisotropy [10].

The substitutions of non-magnetic Ga atoms for Fe atoms exhibit a very strong effect on the magnetocrystalline anisotropy in $R_2Fe_{17-x}Ga_x$ compounds [1, 6, 10]. There is a phase transition of the magnetic anisotropy: from easy-plane to easy-axis anisotropy in these alloys when the Ga content is more than 6.0 (except for when R = Sm) [1, 6, 11]. A reasonable explanation is that the substitution of the larger Ga contents could produce two changes. (1) In $R_2Fe_{17-x}Ga_x$ compounds, the larger Ga contents (for example $x = 7.0$) might imply a reversal in the sign of the second-order crystal parameter A_{20} from negative to positive. So the product $\alpha_J A_{20}$ (rare-earth sublattice anisotropy) might initiate a reversal in the sign of the value. This could explain the uniaxial anisotropy in rare-earth–iron $R_2Fe_9Ga_8$ compounds (except for when R = Sm). (2) In $Y_2Fe_{17-x}Ga_x$ compounds, non-magnetic Y sublattices have not made any contribution to the total magnetocrystalline anisotropy ($\alpha_J A_{20} = 0$). Our refined results show that the larger Ga contents (for example $x = 7.0$–8.0) strongly affect the exchange interactions of the Fe–Ga sublattices; therefore the effect implies a reversal in the sign of the sum of the total anisotropies of the four Fe–Ga sublattices, and they exhibit uniaxial anisotropy. So Y_2Fe_{17} and $Y_2Fe_{12}Ga_5$ compounds exhibit planar anisotropy, but $Y_2Fe_{10}Ga_7$ displays uniaxial anisotropy. In rare-earth–iron $R_2Fe_{17-x}Ga_x$ compounds, the larger Ga contents affect the rare-earth sublattices and the four Fe–Ga sublattices simultaneously. This implies not only a reversal in the sign of the value of the rare-earth sublattice magnetocrystalline anisotropy $\alpha_J A_{20}$, but also a reversal in the sign of the sum of the magnetic anisotropies of the four Fe–Ga sublattices, and indicates uniaxial anisotropy. Our refined magnetic structures for $Y_2Fe_{17-x}Ga_x$ ($x = 5.0, 7.0$) compounds show that the changes of the total magnetic anisotropies of the four Fe–Ga sublattices with increasing non-magnetic Ga contents play a predominant role. The refined results [1, 11] and this report show that the larger Ga contents (for the phase transition of the magnetic anisotropy), which could change the magnetic anisotropy from planar to uniaxial, are almost independent of which rare-earth element (except for when it is Sm) is contained in the compound. It is proved also that the Fe–Ga sublattices make the predominant contributions to the total magnetic anisotropy in the 2:17-type rare-earth–iron compounds.

Tables 1 and 2 show that the Ga atoms occupy 18h and 18f sites with occupancies of 0.42 and 0.40 respectively for $Y_2Fe_{12}Ga_5$ compounds, but the Ga atoms occupy 18h, 18f and 6c sites with the occupancies 0.25, 0.65 and 0.80 respectively for $Y_2Fe_{10}Ga_7$ compounds. The differences between the occupancies of Ga atoms at 18h, 18f and 6c sites in the two compounds show that the larger occupancies of Ga atoms at 18f and 6c sites are necessary to change the magnetic anisotropy from planar to uniaxial anisotropy. That could indicate a way of making the anisotropy of the compounds uniaxial—by using the larger occupancies of non-magnetic atoms at 18f and (or) 6c sites preferentially.

Our refined occupancies of Ga atoms at 18h, 18f and 6c sites in $Y_2Fe_{10}Ga_7$ compounds are different from those for $Tb_2Fe_9Ga_8$ compounds [1]. For example, the Ga atoms occupy 18h sites with the occupancy 0.42 in $Y_2Fe_{12}Ga_5$ compounds, but this decreases to 0.25 in $Y_2Fe_{10}Ga_7$ compounds. The same results were obtained for $Ho_2Fe_{17-x}Ga_x$ compounds [11]. However, [1] showed that the occupancies of Ga atoms at 18h sites are approximately constant (0.41). In principle, the occupancies of Ga atoms on Fe–Ga sublattices must be independent of which rare-earth element is included in the compound. The results of the high-resolution neutron study of $Ho_2Fe_{17-x}Ga_x$ ($x = 5.0, 8.0$) compounds [11] and $Y_2Fe_{17-x}Ga_x$ ($x = 5.0, 7.0$) compounds are more precise and reasonable.

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