Neutron powder diffraction study of $Fe_3Ga_{1.7}As_{0.3}$ and $Fe_3Ga_{1.15}Sb_{0.85}$ B8$_2$-type compounds

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Abstract. High-resolution neutron powder diffraction measurements on $Fe_3Ga_{1.7}As_{0.3}$ and $Fe_3Ga_{1.15}Sb_{0.85}$ are reported. Substantial differences are observed in the refined crystal and magnetic structures of the two compounds. The compound $Fe_3Ga_{1.15}Sb_{0.85}$ is a disordered defect structure, B8$_2$ isotypic with the Ni_2In ($P6_3/mmc$) structure, whilst $Fe_3Ga_{1.7}As_{0.3}$ is a B8$_2$ variant ordered superstructure with a doubled basal plane lattice constant of the parent Ni_2In subcell. The Fe site occupancies have been refined for the subcell and the superstructure. A low-temperature magnetic structure, with moments oriented in the hexagonal basal plane, is found to give best agreement with the observed data for both compounds. The refined Fe magnetic moments for the superstructure are higher than those for the subcell.

1. Introduction

Recent investigations of $Fe_3Ga_{2-x}As_x$ compounds have shown that they display a B8$_2$ defect structure (for example, Ni_2In, $P6_3/mmc$ space group (Castelliz 1943, Laves and Wallbaum 1941, Wallbaum 1943 and Kubaschewski 1982)) and exist over a rather wide range of compositions $0.21 \leqslant x \leqslant 1.13$ (Ellner and El-Boragy 1986, Harris *et al* 1989, Greaves *et al* 1990). They are fairly representative of similar materials that have either a full or partially filled Ni_2In structure, such as Fe_2Ge (Stolc *et al* 1963, Malaman *et al* 1980), Co_2Ge (Agalakova *et al* 1973), Ni_3Ge_2 (Ellner *et al* 1971, Ellner 1976) and Fe_3Ge_2 (Kanematsu 1965). Refinement of x-ray powder diffraction data measurements report a distribution of Fe atoms in octahedral 2a sites, 2 Ga(As) atoms in 2c sites and 1.2 Fe atoms in the trigonally bipyramidal defect site 2d. As a further complication, a superstructure develops for large Ga content, with a doubled basal plane lattice parameter and additional Fe defect sites (Harris *et al* 1989, Greaves *et al* 1990). No synthesis of large single-crystal material has yet been reported in the literature, and thus far only powder materials have been studied.

Macroscopic magnetization and Mössbauer investigations indicate that these compounds are ferromagnetic in nature with a marked rise in Curie temperature with increasing Ga composition. Curie points as high as 644 K have been reported (Harris *et al* 1989, Greaves *et al* 1990). These materials are hence of possible interest for magnetic applications. A powder neutron diffraction study of $Fe_3Ga_{0.85}As_{1.15}$ at 1.5 K has been previously reported (Moze *et al* 1993). The magnitude and orientation of the Fe magnetic moments at low

temperature was deduced from the data. Additionally, the reasonable degree of contrast between the neutron scattering lengths of Fe and Ga(As) atoms, $b_{Fe} = 0.954 \times 10^{-12}$ cm, $b_{Ga} = 0.720 \times 10^{-12}$ cm and $b_{As} = 0.673 \times 10^{-12}$ cm (Sears 1992), was used to determine the presence of additional ordering of Ga(As) atoms on Fe sites as well as the Fe occupancies at defect sites. The structural model was in good agreement with x-ray studies. The proposed magnetic structure gave best agreement for ferromagnetic coupling with moments parallel to the hexagonal c-axis, but a structure with moments in the basal plane was found to give only a slightly worse agreement and hence the magnetic structure remained ambiguous. A non-collinear magnetic structure deduced for FeGe alloys was also used for analysis of the powder data for this material. This model, however, gave unconvincing agreement with the observed intensities due to inherent limitations of the neutron powder diffraction technique in determinations of complex non-collinear magnetic structures.

Compounds with Sb are easier to synthesize than those corresponding to As and, since Sb has similar atomic properties to As, it is of interest to investigate changes in magnetic properties already observed for $Fe_3Ga_{2-x}As_x$ compounds. The neutron scattering length for Sb, $b_{Sb} = 0.571 \times 10^{-12}$ cm, is also favourable for determination of site occupations of Ga and Sb atoms. Preliminary details of neutron diffraction measurements together with bulk magnetic measurements on polycrystalline $Fe_3Ga_{1.7}As_{0.3}$ and $Fe_3Ga_{1.15}Sb_{0.85}$ have been previously reported (Moze *et al* 1994) in a preliminary form. The former compound is representative of the structural modulation observed for $Fe_3Ga_{2-x}As_x$ compounds rich in Ga, whilst the latter has a crystal structure isomorphous with the simpler $B8_2$ (Ni_2In) subcell. Bulk magnetization measurements have indicated that a planar magnetic anisotropy is present for both materials at low temperatures (Moze *et al* 1994). On the basis of the bulk magnetic data, spin-reorientation transitions from plane to axis (or cone) were also predicted to take place at higher temperatures. The complete results concerning structural and magnetic parameters, including site occupancies and Fe magnetic moments, obtained from refinement of high-resolution powder neutron diffraction measurements on these compounds are now reported here.

2. Experimental details

The two compounds $Fe_3Ga_{1.7}As_{0.3}$ and $Fe_3Ga_{1.15}Sb_{0.85}$ were prepared by melting appropriate proportions of high-purity Fe, Ga and As or Sb, as previously already described in the general preparation of $Fe_3Ga_{2-x}As_x$ compounds (Greaves *et al* 1990). The compounds were then subjected to a homogenization procedure at 800 °C for two weeks and consisted of a uniform single-phase matrix after this treatment with a small amount of GaAs and GaSb at the grain boundaries. The resulting bulk specimens were then ground into fine powders for the neutron measurements, which were carried out using the high-resolution neutron powder diffractometer 3T2 at the Orphée reactor, Laboratoire Léon Brillouin, CEA-Saclay, France. The incident wavelength used was from the (335) reflection of a Ge monochromator, 1.2268 Å, with no $\frac{1}{2}\lambda$ contamination. Data were collected in 0.05° steps of the diffraction angle 2θ, in the range 10–120°. Measurements were carried out at 1.5 K. Typical measuring times were 24 h per data set for approximately 3 g of material.

3. Analysis and discussion

The powder neutron diffraction patterns were analysed by the standard Rietveld technique modified to account for the additional effect of magnetic scattering (Rietveld 1968). Small

traces of GaAs and GaSb impurities were detected in the diffraction patterns, but the samples were overall good single-phase material. The refinable parameters included an asymmetry parameter, a scale factor, a preferred orientation correction, lattice constants, isotropic temperature factors for Fe and Ga(As, Sb) and parameters U, V and W for the full width half-height maximum (FWHM) peak parameter Γ, $\Gamma = U \tan^2 \theta + V \tan \theta + W$ (Cagliotti *et al* 1958) as well as site occupancies and Fe magnetic moments. The preferred orientation correction was found to be insignificant for both compounds. For magnetic powder neutron diffraction, a limiting factor is that only the angle between the magnetic moment direction and the unique crystallographic axis can be obtained (Shirane 1959). The measured Bragg peak intensity for unpolarized neutrons is proportional to the sum of the squares of the nuclear structure factor and the magnetic interaction vector, Q:

$$F^2 = F_{\text{nuc}}^2 + Q^2. \tag{1}$$

For hexagonal symmetry, the latter term is related to the magnetic structure factor by $Q^2 = \langle q^2 \rangle F_{\text{mag}}^2$. The orientation factor $\langle q^2 \rangle$ is expressed in terms of the Miller indices (hkl) of a magnetic Bragg reflection as

$$\langle q^2 \rangle = 1 - \left[\tfrac{1}{2}(h^2 + k^2 + hk)a^{*2} \sin^2 \phi + l^2 c^{*2} \cos^2 \phi \right] d^2 \tag{2}$$

where a^*, c^* are the reciprocal lattice constants, ϕ is the angle between the spin direction and the [0001] axis and d is the d spacing for reflection (hkl). Naturally the maximum information that can be obtained from powder data is the orientation of the spins with respect to the uniaxial axis. All trial structures with moment directions in the basal plane are equally likely.

A number of various trial models of the magnetic moments were tested in the refinements on the basis of the above expressions. These included moments ferromagnetically aligned along the uniaxial direction and in the basal plane. A canted structure reported for FeGe alloys (Adelson and Austin 1965) was previously used to model the diffraction data of Fe₃Ga₀.₈₅As₁.₁₅ but led to no better agreement with observed intensities for this compound. The magnetic form factor dependence of the Fe atoms was taken into account by using the analytic approximations of Lisher and Forsyth (1971).

3.1. Results for Fe₃Ga₁.₁₅Sb₀.₈₅

3.1.1. Crystal structure. A total of 48 separate and overlapping reflections in the 2θ range 20–107° were used for refinement of the crystal and magnetic structure for this compound. A total of seven structural parameters were refined: a scale factor, an Fe temperature factor common for the two sites, a common temperature factor for Ga and Sb, separate Fe occupancy factors for sites 2a and 2d and lattice parameters a and c. The data refined well on the partially filled Ni₂In structural model with the octahedral 2a Fe site fully occupied whilst the trigonally coordinated bipyramidal 2d Fe site is only partially occupied. The refined Fe occupancy for the defect 2d site was 1.0, in contrast with 1.2 for As compounds. This is consistent with the expected stoichiometric ratio of Fe/(Ga + Sb) = 3/2. The refined thermal parameters for all sites are less than those observed for the corresponding As compound whilst the lattice parameters are about 1–2% larger than for the corresponding Fe₃Ga₁.₁₅As₀.₈₅ compound (Greaves *et al* 1990) giving a unit cell volume of 73.9 Å³, in contrast to a unit cell volume of 70.5 Å³ ($a = 4.03$ Å, $c = 5.01$ Å) for the As compound.

Table 1. Calculated and observed intensities for $Fe_3Ga_{1.15}Sb_{0.85}$ at 1.5 K.

h	k	l	2θ (deg)	I_{Nuc}^{Calc}	I_{Mag}^{Calc}	I_{Total}^{Calc}	I_{Obs}
0	1	0	19.95	972	53	1024	1032
0	1	1	24.37	189	81	270	243
0	0	2	27.95	12	0	12	0
0	1	2	34.57	8413	42	8455	8688
1	1	0	34.89	7560	103	7664	7755
0	2	0	40.51	277	5	281	249
0	2	1	43.01	28	13	41	0
1	1	2	45.27	6	0	6	0
0	2	2	49.92	3985	16	4001	3604
1	2	0	54.50	354	2	356	355
1	2	1	56.53	11	7	18	0
0	0	4	57.73	882	0	882	754
0	2	3	60.12	3	1	4	9
0	1	4	61.70	301	0	301	333
1	2	2	62.34	5109	11	5120	5392
0	3	0	62.55	2242	6	2248	2347
0	3	1	64.42	0	0	0	0
1	1	4	69.25	3645	2	3647	3944
0	3	2	69.85	2	0	2	78
1	2	3	71.42	0	1	1	154
0	2	4	72.88	248	0	248	258
2	2	0	73.66	1612	2	1614	1521
1	3	0	77.20	233	0	234	244
0	1	5	77.76	0	0	0	0
0	3	3	78.53	0	0	0	0
1	3	1	78.91	1	1	1	0
2	2	2	80.51	7	0	7	121
1	2	4	83.40	435	0	435	371
1	3	2	83.97	2941	2	2943	2818
0	2	5	88.14	2	0	2	77
0	4	1	89.27	2	0	2	30
0	3	4	90.28	2220	0	2220	2108
1	3	3	92.32	5	0	6	67
0	0	6	92.77	2	0	2	7
0	4	2	94.27	1228	0	1228	1351
0	1	6	96.21	1193	0	1193	1632
2	3	0	97.91	195	0	195	141
1	2	5	98.46	8	0	9	0
2	3	1	99.60	9	0	9	0
2	2	4	100.63	1884	0	1884	1791
0	4	3	102.71	5	0	5	0
1	1	6	103.18	18	0	18	0

3.1.2. Magnetic structure. Refinement of the magnetic structure with moments oriented in the basal plane gave an overall better quality of fit, with sensible moments and smaller errors than a uniaxial model, but a complex non-collinear magnetic structure cannot be excluded. The two different atom co-ordinations that occur around the two Fe atom sites would indicate different magnetic moments at 2a and 2d sites. The larger interatomic distances observed for this Sb compound are consistent with the higher magnetic moments at the two Fe sites, in contrast with the corresponding As compound. The Curie point for this compound is also approximately 18% higher than for the corresponding As compound (Moze *et al* 1993, 1994), pointing to an increased exchange interaction induced by Sb
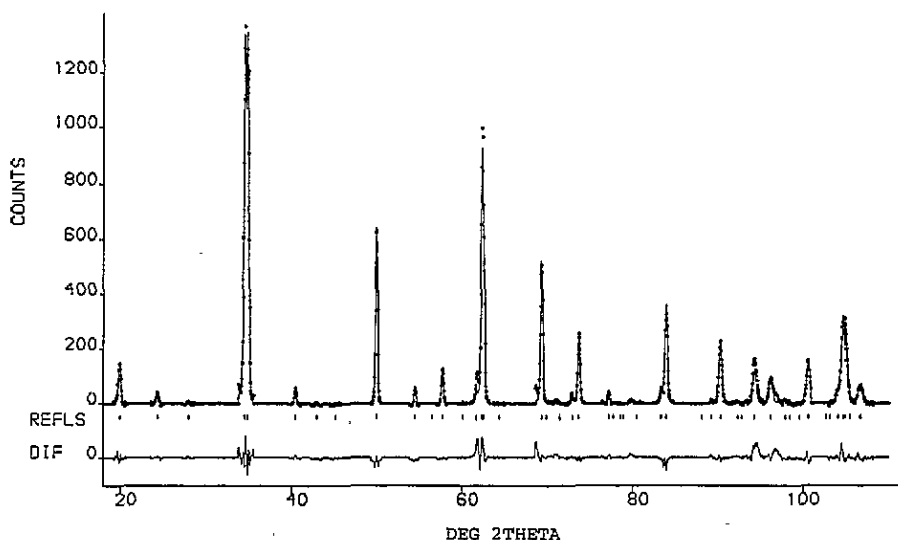
Figure 1. Observed and calculated neutron diffraction pattern at 1.5 K for $Fe_3Ga_{1.15}Sb_{0.85}$. Calculated positions of reflections are also marked together with a difference plot of calculated and observed points, which is on the same scale as the calculated and observed intensities.

substitution. A list of calculated and observed intensities is displayed in table 1, whilst the refined parameters are displayed in table 2. However, close inspection of table 1 clearly shows that for this compound, the neutron powder diffraction measurements are not sufficiently reliable to identify one magnetic model from another or indeed to determine the true microscopic magnetic structure of this material. The magnetic contribution to the overall diffraction pattern is extremely small and renders most difficult an accurate assessment of the magnetic structure for this compound. This is an obvious source of the large errors for the refined magnetic moments at sites 2a and 2d. In the region where there is the largest magnetic contribution to the diffraction pattern i.e. below $2\theta = 60°$, the R factor is more than 100%. The associated standard deviations are large and, taking this into consideration, an average Fe site moment of about $2\mu_B$ would appear to be a more realistic and sensible estimate, despite the underestimated errors yielded by the Rietveld analysis. This example points out the general difficulty of determining the magnetic structure of complex materials from powder data alone. The observed and calculated diffraction pattern for $Fe_3Ga_{1.15}Sb_{0.85}$ are displayed in figure 1. A schematic layout of the structure is shown in figure 2. No constraints have been imposed in the refinements on the magnitudes of the Fe moments but, nevertheless, the average Fe moment obtained from this neutron study is in good agreement with previously reported bulk magnetization data on the same sample. This point is amplified in the discussion below.

Table 2. Refined structural and magnetic parameters for $Fe_3Ga_{1.15}Sb_{0.85}$ at 1.5 K.

Atom	Site	x/a	y/a	z/c	$B(\text{Å}^2)$	Occupancy	$\mu_y(\mu_B)$
Fe_{2a}	2a	0	0	0	0.49(5)	2	1.5(3)
Fe_{2d}	2d	1/3	2/3	3/4	0.49(5)	0.90(5)	2.0(3)
Ga	2d					0.04(1)	
Sb	2d					0.06(1)	
Ga(Sb)	2c	1/3	2/3	1/4	2.13(20)	2	

○ Fe 2a
● Ga/Sb 2c
◉ Fe , Ga/Sb 2d
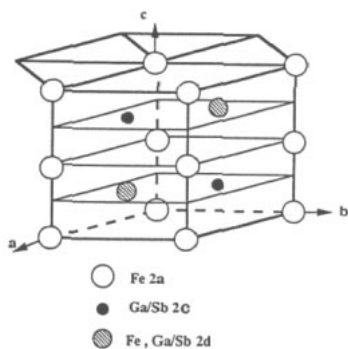
Figure 2. Disordered Ni_2In-type structure observed for $Fe_3Ga_{1.15}Sb_{0.85}$.

3.2. Results for $Fe_3Ga_{1.7}As_{0.3}$

3.2.1. Crystal structure. A total of about 180 separate and overlapping reflections in the 2θ range 15–120° were used for refinement of the crystal and magnetic structure for this compound. Indexing of strong reflections immediately pointed to a cell with a doubled lattice parameter a with respect to As-rich compounds (Greaves *et al* 1990). Additional peaks not belonging to this phase were also observed and were immediately excluded from the refinement. The most intense peak of this second phase occurred at 35°. The data were refined on the structural model previously reported for powder x-ray measurements of the same compound (Greaves *et al* 1990), based on a similar $B8_2$ ($P6_3/mmc$) type variant of a η-phase reported for the Fe–Ge system (Kanematsu 1965). The overall composition was constrained to $Fe_3Ga_{1.7}As_{0.3}$. In terms of this large cell, the 2a, 2d and 2c sites for the subcell transform into sites (2a, 6g), (2c, 6h, $6h_1$) and (2d, $6h_2$) respectively. The good quality of the data set, with a large number of intense reflections even at high scattering angles, allowed an adequate refinement to be carried out of the Fe site occupancies, fractional atomic co-ordinates and temperature factors. The atomic co-ordinates and site occupancies used in a previous x-ray refinement were used as starting parameters.

The initial refinements indicated a full occupancy for Fe site 2a and zero occupancy for Fe site 2c. These occupancies were hence subsequently fixed. A total of 13 structural parameters were refined: a scale factor, lattice parameters a and c, x and y fractional co-ordinates for sites $6h_1$ (Fe) $6h_2$ (Ga, As), common temperature factors for sites Fe (2a, 6g, $6h_1$), Ga(As) (2d, $6h_2$), occupancy factors for sites 2a, 6g and 6h. Final refined parameters are displayed in table 2. The ordering scheme here is very similar to the least probable structure postulated by Kanematsu (1965) who proposed, instead, an alternative for $Fe_{1.4}Ge$ with only a 56% occupancy of the $6h_1$ five-coordinated site and a 60% occupancy of the octahedral 2a site. In contrast, however, with the x-ray powder refinement for this compound, the 2a site is only 83% occupied.

3.2.2. Magnetic structure. Initial refinements with different magnetic moments on the three Fe sites 2a, 6g and $6h_1$ led to extreme correlations between structural and magnetic parameters. The moments were thus constrained to be equal on these sites. Shown in table 3 is a list of calculated and observed intensities, which for convenience are listed only up to $2\theta = 60°$. The refined parameters for this compound are displayed in table 4. The refined moments are in agreement with the bulk measurements reported for the series $Fe_3Ga_{2-x}As_x$ (Harris *et al* 1989) where an enhanced Fe moment is reported with increasing

Table 3. Calculated and observed intensities for $Fe_3Ga_{1.7}As_{0.3}$ at 1.5 K.

h	k	l	2θ (deg)	I_{Nuc}^{Calc}	I_{Mag}^{Calc}	I_{Total}^{Calc}	I_{Obs}
0	1	1	17.38	368	71	439	558
1	1	0	17.41	755	242	996	1180
0	2	0	20.13	774	928	1702	1613
0	2	1	24.68	84	625	709	733
1	2	0	26.71	542	1	543	609
0	0	2	28.52	635	0	635	651
0	1	2	30.30	89	6	95	96
1	2	1	30.35	4269	134	4403	4157
0	3	0	30.36	712	52	765	734
1	1	2	33.59	173	9	182	220
0	3	1	33.64	3031	20	3052	3397
0	2	2	35.14	24135	453	24589	24178
2	2	0	35.19	20464	1048	21513	22033
1	3	0	36.68	54	8	62	376
1	2	2	39.45	969	2	970	953
1	3	1	39.49	1372	19	1391	1301
0	4	0	40.86	713	86	800	764
0	3	2	42.12	464	8	472	322
0	4	1	43.44	248	93	341	96
0	1	3	44.60	64	0	65	0
2	3	0	44.71	9	20	29	0
2	2	2	45.88	308	14	321	221
1	3	2	47.09	0	10	10	6
2	3	1	47.12	146	4	150	100
1	4	0	47.13	75	14	89	61
0	2	3	48.21	45	13	58	34
1	4	1	49.45	1539	8	1547	1721
0	4	2	50.56	9316	176	9492	10000
1	2	3	51.62	1528	5	1533	1224
0	5	0	51.72	186	1	188	154
0	3	3	53.81	1192	1	1193	945
2	3	2	53.87	15	19	34	28
0	5	1	53.90	1116	13	1129	924
3	3	0	53.91	108	2	110	90
2	4	0	54.98	689	44	733	719
1	4	2	55.99	18	4	22	390
2	4	1	57.07	1280	46	1326	1550
1	3	3	58.02	655	1	657	565
1	5	0	58.11	83	0	83	59
0	0	4	58.97	3281	0	3281	3302
0	1	4	59.98	1	0	1	1

Ga content. For $Fe_3Ga_{1.7}As_{0.3}$ an average Fe moment of $2.12\mu_B$ at 77 K is quoted in comparison with $1.50\mu_B$ reported for $Fe_3Ga_{0.88}As_{1.12}$. The observed and calculated neutron diffraction pattern for this compound is displayed in figure 3 whilst a schematic layout of the refined superstructure is shown in figure 4. As can be inferred from table 3, the magnetic contribution to the total diffraction pattern is, in this case, larger than for the Sb compound. The larger number of observations imply that the Fe moments refined for this compound are certainly more reliable. Thus it appears that even from this powder data alone that the Fe moments are considerably higher in the ordered superstructure.
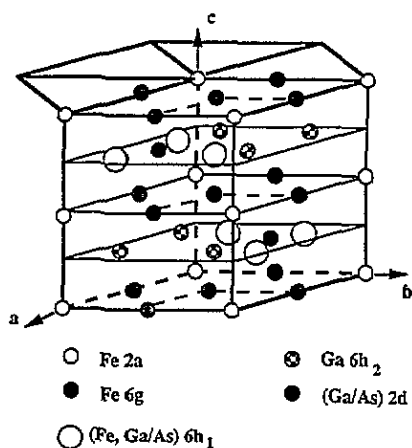
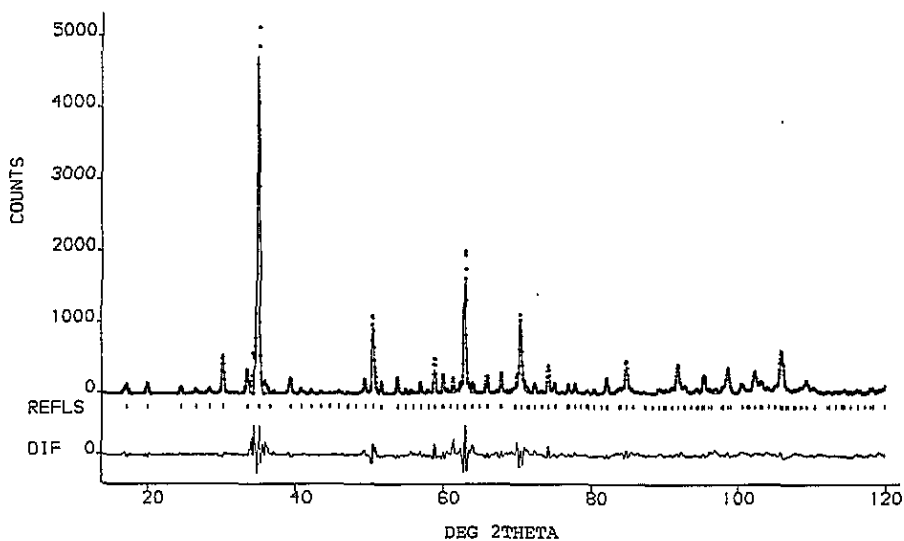Figure 3.   The $B8_2$ superstructure observed for $Fe_3Ga_{1.7}As_{0.3}$.

Figure 4. Observed and calculated neutron diffraction pattern at 1.5 K for $Fe_3Ga_{1.70}As_{0.30}$. Calculated positions of reflections are also marked together with a difference plot of calculated and observed points, which is on the same scale as the calculated and observed intensities.

Table 4. Refined structural and magnetic parameters for $Fe_3Ga_{1.7}As_{0.3}$ at 1.5 K.

Atom	Site	x/a	y/a	z/c	$B(\text{Å}^2)$	Occupancy	$\mu_y(\mu_B)$
Fe_{2a}	2a	0	0	0	0.26(2)	1.65(8)	2.13(15)
Fe_{6g}	6g	1/2	0	0	0.26(2)	5.85(9)	2.13(15)
Fe_{6h1}	$6h_1$	0.8428(3)	0.1572(3)	1/4	0.26(3)	5.40(2)	2.13(15)
Ga(As)	$6h_1$			1/4	0.26(3)	0.51(2)	
Ga(As)	2d	2/3	1/3	1/4	0.86(8)	2	
Ga(As)	$6h_2$	0.1993(5)	0.8007(5)	1/4	0.86(8)	6	

4. Discussion

No constraints were imposed on the individual Fe magnetic moments for the Sb compound. The refined Fe magnetic moments are in good agreement with the bulk magnetization data reported for both compounds. In table 5 we summarize the relevant bulk magnetic data for the two compounds reported here, as well as data for the previously reported Fe$_3$Ga$_{0.85}$As$_{1.15}$ compound (Moze *et al* 1993). Also included in this table are refined lattice constants for all three compounds, as well as the average Fe moment obtained from the neutron data. The agreement for all three compounds between bulk and microscopic magnetic data is satisfactory, with the possible exception of the Sb compound, where the disagreement between the refined average Fe moment and the bulk magnetization is larger. In table 6 we display the quality factors for the refinements of the compounds presented in this work, as well as for Fe$_3$Ga$_{0.85}$As$_{1.15}$. A planar magnetic structure gives an overall better quality of fit as well as realistic Fe magnetic moments, which are in agreement with bulk data, for Fe$_3$Ga$_{1.7}$As$_{0.3}$ and Fe$_3$Ga$_{1.15}$Sb$_{0.85}$ whereas for Fe$_3$Ga$_{0.85}$As$_{1.15}$, the distinction between a planar and axial structure is less clear. Bulk magnetic anisotropy measurements report a basal plane easy magnetization direction for all three compounds at low temperatures.

Table 5. Curie points, T_c and bulk magnetization, σ_s at 77 K, average magnetic moment and lattice parameters for Fe$_3$Ga$_{0.85}$As$_{1.15}$, Fe$_3$Ga$_{1.7}$As$_{0.3}$ and Fe$_3$Ga$_{1.15}$Sb$_{0.85}$. The bulk magnetization data at 77 K were taken from Harris *et al* 1989. Average Fe moments reported at 1.5 K have been calculated from the refined magnetic moments.

	Fe$_3$Ga$_{0.85}$As$_{1.15}$	Fe$_3$Ga$_{1.7}$As$_{0.3}$	Fe$_3$Ga$_{1.15}$Sb$_{0.85}$
T_c (K)	375	600	588
σ_s (77 K) (emu gm^{-1})	81	127	98
μ_B/Fe (77 K)	1.50	2.20	1.90
μ_B/Fe (1.5 K)	1.4(1)	2.1(0.2)	1.7(0.3)
a (Å)	3.9810(3)	8.1260(1)	4.0943(4)
c (Å)	5.0312(1)	4.9883(3)	5.0843(2)

Table 6. Refinement quality factors for Fe$_3$Ga$_{0.85}$As$_{1.15}$ (A), Fe$_3$Ga$_{1.7}$As$_{0.3}$ (B) and Fe$_3$Ga$_{1.15}$Sb$_{0.85}$ at 1.5 K (C).

	A		B		C	
Moment direction	z	y	z	y	z	y
R_{wp} %	13.1	13.2	26.0	26.0	16.2	16.1
R_{exp} %	11.6	11.6	13.0	13.0	15.3	15.3
R_{nuc} %	8.9	9.1	14.2	14.2	9.2	9.3
R_{mag} %	8.8	13.7	17.4	17.6	39.4	21.0
μ_B, 2a	1.24(14)	1.40(19)	1.88(14)	2.13(15)	0.8(5)	1.5(3)
μ_B, 2d	1.55(13)	1.41(16)	1.88(14)	2.13(15)	1.5(6)	2.0(3)

A theoretical description of the magnetic properties of these materials is lacking. For the Fe$_3$Ge$_2$ system where the magnetic structure has definitively been shown to be of a canted type (Adelson and Austin 1965), it has been suggested that the de Gennes theory of double exchange (de Gennes 1960) is applicable. In this case, a possible antiferromagnetic

exchange via Ge atoms, gives rise to a canting of Fe moments at 2d sites. Similar arguments also aptly apply for the $Mn_{1-x}Cr_xSb$ system (Reimers *et al* 1982) which crystallizes in the hexagonal NiAs structure. In the present case it has not been possible to directly verify canting of moments at 2d sites but given the nature of the significant changes which occur in saturation magnetization, Curie temperature and magnetic anisotropy with substitution of non-magnetic ions it would appear that the de Gennes model may also be applicable for these materials. The observation of transitions from a planar to axial or cone anisotropy at higher temperatures (Moze *et al* 1994) is a strong indication for competing exchange interactions and strongly temperature-dependent competing anisotropies. The magnetic contribution to the overall powder diffraction pattern is very small (typically 5%) and magnetic information is intrinsically restricted to the first three or four peaks. Polarized neutron measurements on single crystals are obviously desirable for a more accurate determination of the magnetic structure of this series of interesting materials.

Acknowledgments

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