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Journal of Alloys and Compounds 323–324 (2001) 531–533
Journal of ALLOYS AND COMPOUNDS
www.elsevier.com/locate/jallcom

Magnetism of $UFe_{4-x}Al_{8+x}$ ($x=\langle -0.4, 0.4\rangle$) intermetallics

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Abstract

Results of X-ray, neutron diffraction, magnetization and Mössbauer measurements with circularly polarised beam on polycrystalline samples of the ternary intermetallic alloys $UFe_{4-x}Al_{8+x}$ with x in the range (-0.4, 0.4) are presented. The alloys crystallise in the structure of $ThMn_{12}$ type, belonging to the space group *I*4/*mmm* (no. 139). X-ray and neutron diffraction confirmed the phase homogeneity of all samples but that one with $4-x=4.4$. Monochromatic Circularly Polarised Mössbauer Source (MCPMS) measurements reveal spin-canted magnetic structures in randomly oriented powders exposed to a field of 1 T at T=12 K. Magnetization measurements in an external field of 0.01 T allowed disclosing ferromagnetic-like behaviour vs. temperature and quite dissimilar magnetization and hysteresis curves against the fields in the alloys of interest. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Intermetallics; Magnetisation; Phase transitions; Neutron scattering, diffraction; Magnetic measurements.

1. Introduction

The magnetism of $UFe_{4-x}Al_{8+x}$ intermetallic compounds is raising a lot of interest [1,2] because of rather unusual properties of these alloys. In light of the cited papers we find it interesting to report our results of the X-ray, unpolarized neutron diffraction as well as magnetization and Mössbauer investigations of the magnetic properties of $UFe_{4-x}Al_{8+x}$ alloys with iron concentration ranging from 3.6 to 4.4. The crystal structure of these samples is well known: they crystallise in the body-centred tetragonal $ThMn_{12}$ type structure (space group *I*4/*mmm*). The thorium site *2a-(0,0,0)* is fully occupied by uranium atom at the origin and center of the unit cell. The manganese sites *8i-*$(x_1,0,0)$ are occupied by aluminium atoms only and *8f-(1/4,1/4,1/4)* as well as *8j-*$(x_2,1/2,0)$ are occupied by iron and aluminium according to the sample chemical composition. Weak ferromagnetism discovered in the single crystal sample of UFe_4Al_8 [3] disproved a simple magnetic model, deduced from powder neutron diffraction in the zero magnetic field, in which it was assumed that the uranium magnetic moments are frustrated while the iron ones are antiferromagnetically oriented perpendicularly to [001] direction [4]. A small ferromagnetic component found along, say, [100] direction in the field of 4.6 T [3] indicates that the atomic iron moments are inclined by 25° with respect to [010]. The authors estimated this angle in zero field to be 16°, resulting in ferromagnetic component of the iron magnetic moment close to 0.3 μ_B. This model was shown to work well also for our earlier powder data [5]. Recently, the magnetic phase diagram, based on the ac-susceptibility, magnetisation and ^{57}Fe Mössbauer-effect measurements of single crystals UFe_yAl_{12-y}, with y ranging from 3.8 to 5.8, was proposed [2]. In a range of y a little above $y=4$, two transition temperatures were discovered. This was already noticed for unannealed UFe_4Al_8 [6] contrary to only one reported in Refs. [1–3].

2. Sample characterisation

The $UFe_{4-x}Al_{8+x}$ compounds of compositions $x=0.4$, 0.2, 0.0 and -0.4 were prepared by melting in an induction furnace. The melting itself was carried out under a protective argon atmosphere. The lumps obtained were

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PII: S0925-8388(01)01137-9

next crushed into powders and underwent prolonged annealing at 800°C for 2 weeks. X-ray RT measurements were carried out on HZG-4C diffractometer. The standard wavelength used was $\lambda=70.926$ pm. The neutron diffraction experiments on the $UFe_{3.6}Al_{8.4}$ have been performed at HMI, BENSC (Germany). In order to determine the magnetic structure, the diffractometer E2 ($\lambda=240.56$ pm) was used. The measurements were carried out in the temperature range of 1.5–140 K. The neutron studies of UFe_4Al_8 and $UFe_{4.4}Al_{7.6}$ samples have been carried out at LLB (France). The crystal structures were measured above the phase transition temperatures on 3T2 diffractometer ($\lambda=122.72$ pm). The neutron measurements, which have been performed on G4.1 diffractometer ($\lambda=242.49$ pm) in the temperature of 1.5–230 K, allowed to determine the magnetic structures of the alloys of our interest. The diffraction spectra were analysed by means of the Rietveld refinement method with the aid of the computer program Fullprof [7]. The magnetization measurements were carried out using vibrating sample magnetometer (VSM) under magnetic fields up to 1.2 T in the temperature range 10–300 K, both with zero-field-cooling (ZFC) and field-cooling (FC) scenarios. The ^{57}Fe Monochromatic Circularly Polarised Mössbauer Source (MCPMS) measurements were curried with commercial ^{57}Co source in Cr matrix with an initial activity of 50 mCi. The details of the technique can be found in Ref. [8]. During these measurements the polycrystalline samples were cooled in the field of 1 T.

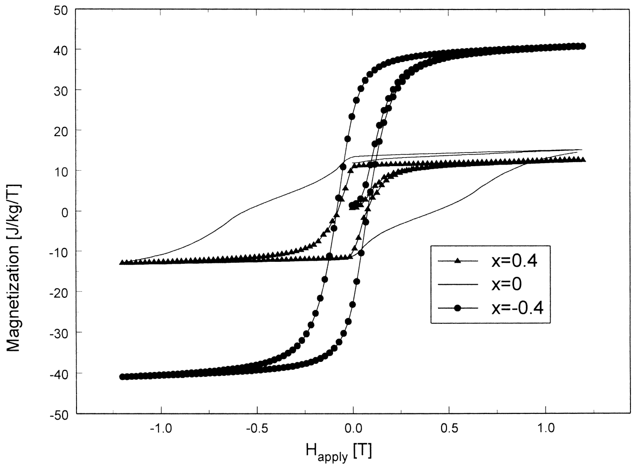

Fig. 1. Magnetization and hysteresis curves measured on $UFe_{4-x}Al_{8+x}$ samples at 10 K.

3. Results and conclusions

The X-ray powder diagrams show that $UFe_{4-x}Al_{8+x}$ compounds with the $x\geq 0$ are all single phase, while that one with the $x=-0.4$ contains an extra phase of β-FeAl type. Its quantity cannot be unambiguously determined because of the small intensity of peaks belonging to this phase. Substitution of Fe atoms by larger Al atoms results in an expansion of the $a=b$ lattice parameter while c remains intact. All recent experiments on powder [4,5,9,10] as well as single crystal samples of UFe_4Al_8 [1–3] indicate that the uranium and iron moments are oriented in the easy ab-plane with iron moments oriented almost antiparallel to each other and their ferromagnetic component parallel to the uranium moment. Our magnetisation and MCPMS results, to be discussed below, show certain disagreement with the results presented recently by Waerenborgh et al. (see Ref. [1]). In accordance with these authors, the magnetic saturation in *ab*-plane is achieved in the magnetic field exceeding 2 T, the value only a little larger than the coercive field. Fig. 1 illustrates magnetization and hysteresis curves obtained for our powder alloys at 10 K. In the case of UFe_4Al_8 the shape of the magnetization curve is more complicated than for other samples. However, the saturation field is a little above 1 T. For the remaining samples, it is below 1 T and the coercive field is about 0.2 T only, diminishing on both sides of $x=0$.

The magnetization measurements indicate that the actual phase diagram is more complicated than that one reported in Ref. [2]. Fig. 2 illustrates ZFC and FC magnetization of UFe_4Al_8 and $UFe_{4.4}Al_{7.6}$ powder samples in magnetic field B = 0.01 T. Both presented compositions exhibiting two magnetic transitions, at ~153 K, ~35 K for UFe_4Al_8 and ~209 K, ~143 K for $UFe_{4.4}Al_{7.6}$. The presence of two transition temperatures turned out to be independent on the annealing treatment and magnetic history. In contrast, in the case of $UFe_{3.6}Al_{8.4}$ and $UFe_{3.8}Al_{8.2}$ one transition temperature only was recorded at ~171 and ~157 K, respectively. The results of MCPMS indicate certain magnetic domain distribution in all samples under study. From the analysis of MCPMS spectra one can infer individual canting angles (α) at iron sites. The values

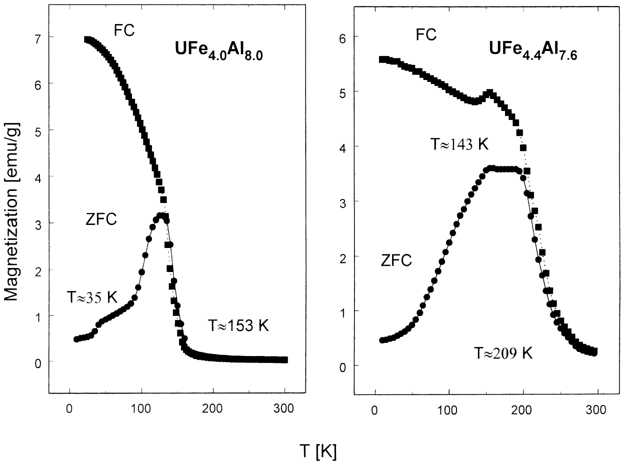

Fig. 2. Magnetization against temperature measured in an applied field of 0.01 T.

obtained vary from ~9 to ~62 degrees with the lowest value for the $UFe_{3.8}Al_{8.2}$ sample. In particular, the iron-canting angle found for UFe_4Al_8 is close to 30 degrees, the value higher than that given in [3]. The conclusion on canted spin structure is not so obvious if one relies on the powder neutron diffraction data only. As was mentioned in the previous paper [4,5] the relatively weak magnetic scattering with respect to the nuclear one makes the analysis exceedingly difficult. In order to separate out the magnetic scattering from thermal effects as well as from the contribution of the second order scattering, the procedure described by Szymanski et al. [5] was used. The appropriate Debye-Waller factors were calculated and fixed. The neutron diffraction data confirms the lack of pure magnetic reflections in all samples. However, it is worth noting that during our unpolarized neutron experiments all crystallites were randomly oriented an applied magnetic field. The neutron diffraction data show that in the case of $UFe_{3.6}Al_{8.4}$ the uranium magnetic moment is $0.62(15)\mu_B$, while iron moments oriented antiferromagnetically equal $0.93(4)\mu_B$ at $T=1.4$ K. These moments are $0.32(14)\mu_B$ and $1.05(4)\mu_B$ at $T=10$ K, respectively. Such values were obtained for preassumed zero canting angle. The nuclear (R_N) and magnetic (R_M) agreement factors as well as χ^2 are 0.028, 0.037 and 64. Changing the canting angle to $\alpha=11°$ as given by MCPMS experiment worsens the R_M by 1.5% only, without change of χ^2 and R_N factor. Both uranium and iron magnetic moments disorder at the same temperature ~160 K. The neutron diffraction pattern for $UFe_{4.4}Al_{7.6}$ may be interpreted in terms of a simple ferromagnetic model with the same value of iron magnetic moment at *f* and *j* sites. The values obtained are $\mu U=1.20(16)\mu_B$ and $\mu Fe=1.10(16)\mu_B$ at 1.4 K with the $R_N=0.028$, $R_M=0.037$ and $\chi^2=36$. If an antiferromagnetic iron component is included as a free parameter we obtain $\mu U=0.92(25)\mu_B$, $\mu Fe_{(F)}=1.16(14)\mu_B$ and $\mu Fe_{(AF)}=0.13(8)\mu_B$ under unchanged χ^2 as well as R_N and better about 2.8% R_M. Such a structure is compatible with our MCPMS results. According to neutron measurements, ordering on the uranium site disappears above 130 K. In light of MCPMS data we think that the low temperature transitions in both, $UFe_{4.4}Al_{7.6}$ and UFe_4Al_8 samples are due rather to the uranium sublattice disordering.

Acknowledgements

The work was sponsored by the State Committee of Scientific Research through grant No 2 PO3B 038 19 and by Contract HPRI-CT-1999-00020.

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