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The crystal and magnetic structures of $\text{UFe}_x\text{Al}_{12-x}$ intermetallic compounds

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Abstract. The powder samples of $\text{UFe}_x\text{Al}_{12-x}$ ($x = 4, 5$ and 6) alloys have been measured by means of magnetization, Mössbauer effect, x-ray and neutron diffraction techniques in the temperature range 1.5–400 K. Both neutron and x-ray diffraction experiments showed that the positions f in all samples are occupied by iron, while the iron atoms in the samples with $x = 5$ and 6 locate also at j sites. A little f–j disorder exists in UFe_4Al_8 . The paper shows a change of the magnetic structure with an increase of iron content. The magnetic moments lie in a basal a – b plane. Iron atoms in UFe_4Al_8 alloy order antiferromagnetically. UFe_5Al_7 and UFe_6Al_6 alloys exhibit in general a ferromagnetic behaviour. However a detailed distribution of magnetic moments among different sites could not be determined unambiguously from the neutron data only. Nevertheless, combining information from neutron and Mössbauer experiments, one can infer that the ordering among iron atoms must be non-collinear. A magnetic ordering among uranium atoms has been found in UFe_5Al_7 only. Low-temperature thermal expansion varies nonlinearly with temperature and in all samples is negligible at low temperatures.

1. Introduction

Despite the great efforts of many experimentalists (Baran *et al* 1984, Ptasiewicz-Bąk *et al* 1988, Schäfer *et al* 1989a, b, Gal *et al* 1990, Suski 1990, Suski *et al* 1992, Andreev *et al* 1992, Paixão *et al* 1994, Vagizov *et al* 1995, Godinho *et al* 1995, Dobrzyński *et al* 1996; for a recent review, see Suski 1996) who studied magnetic properties of the intermetallic compounds $\text{UFe}_x\text{Al}_{12-x}$, the information concerning this type of alloy with x ranging from 4 to 6 is very incomplete. All these compounds crystallize in a body-centred tetragonal crystal structure ThMn_{12} which belongs to the $I4/mmm$ space group, figure 1. There are 26 atoms (two molecules) per unit cell. The thorium site (2a) is occupied by uranium atoms and the manganese sites (8f, 8i, 8j) can be occupied by iron and aluminium atoms.

The presence of two different atomic species, namely uranium and iron, which can carry magnetic moments, and a certain freedom of choice of site by an iron atom, which may create natural disorder in the lattice, can lead to complicated magnetic moment distributions. The magnetization measurements (Andreev *et al* 1992, Vagizov *et al* 1995) strongly suggest a change of magnetic structure with an increase of the iron concentration. Our paper confirms this expectation. Based on the magnetization measurements at high magnetic fields, up to

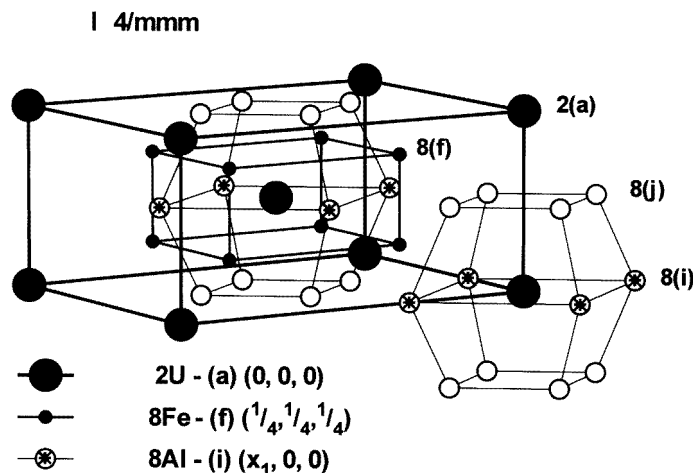


Figure 1. Crystal structure of $\text{UFe}_x\text{Al}_{12-x}$ compounds.

45 T, Lagutin (1989) suggested that ‘noncompensated antiferromagnetic structure’ appears at $x < 4$, antiferromagnetic structure sets in when $x = 4$, and there is a ‘coexistence of pure antiferromagnetic and ferromagnetic ordering at $x > 4$ ’.

The iron sublattice in the most frequently studied UFe_4Al_8 alloy has indeed been reported (Schäfer *et al* 1989a, b, Gal *et al* 1990, Paixão *et al* 1994) to order antiferromagnetically, whereas the situation with uranium atoms is much less clear. Single crystal investigations by means of the polarized neutron diffraction technique (Paixão *et al* 1994, 1997) indicate that canting of iron moments takes place. The model proposed, however, does not seem to explain fully very unusual results shown recently by Bonfait *et al* (1996) for a change of magnetization vector under an applied magnetic field.

The magnetic measurements carried out on a powder (Andreev *et al* 1992) and on a single crystal (Godinho *et al* 1995) of UFe_4Al_8 , as well as the measurements on a single crystal of $\text{UFe}_{5.8}\text{Al}_{6.2}$ (Gonçalves *et al* 1996), show that these compounds have an easy a - b plane while the c -axis is a hard magnetization axis. However, the literature cited above brings many controversial data concerning the values of the saturation magnetization, the iron and uranium magnetic moments and their orientations, etc (see also the article by Dobrzyński *et al* (1996), who showed what an important role is played by the magnetic and mechanical history of the sample). Therefore, in principle, only single-crystal measurements should offer hope for obtaining a solution to the problem. Unfortunately, a strong anisotropy of the compound, and a possibility that the magnetic structure may depend on the magnetic field applied to the sample, complicate this problem in any case. It is apparent that the antiferromagnetism of iron moments cannot be reconciled with the non-zero saturation magnetization reported for UFe_4Al_8 . Seemingly, the easiest explanation is that the uranium atoms carry the appropriate moments which become ordered ferromagnetically under an applied magnetic field only. In fact, uranium atoms surrounded by eight iron atoms oriented antiferromagnetically would exhibit magnetic frustration if the U-Fe nearest-neighbour interaction were the dominant one. On the other hand, canted iron spins may create a sufficiently strong molecular field which orders uranium moments (Paixão *et al* 1997). Our zero-field studies do not confirm the ordering of uranium moments.

In order to approach the solution of this intriguing situation our group decided to make a series of investigations which would start from the iron-rich alloys, namely UFe_5Al_7 and UFe_6Al_6 whose magnetization character strongly suggests a regular ferromagnetism (Andreev *et al* 1992, Vagizov *et al* 1995) with Curie temperatures of 280 and 350 K (Vagizov *et al* 1995), respectively. The idea was that once the magnetic structure of these two alloys was determined, one could study the magnetic phase transition in the region of x below 5. Our first experiments (Rečko *et al* 1996), however, showed that even iron-rich alloys are showing quite a complicated magnetic structure. A detailed report from our magnetization, x-ray, neutron and Mössbauer studies presented below describes this situation in detail.

2. Experimental details

All three samples with $x = 4, 5$ and 6 were prepared by arc-melting in the W Trzebiatowski Institute of Low Temperatures and Structure Research in Wrocław, Poland. The melting itself was carried out under a protective argon atmosphere. The lumps obtained were next crushed into a powder and underwent prolonged annealing at about 800°C for 2 weeks.

The magnetization measurements were carried out using a conventional magnetic balance installed in the Institute of Physics in Białystok. The measurements have been performed in the temperature region of $77\text{--}300$ K and magnetic fields up to 1.4 T, and their main target was to compare the results with the ones published earlier (Andreev *et al* 1992, Vagizov *et al* 1995).

The Mössbauer measurements were also carried out in Białystok. A standard constant-acceleration mode spectrometer was used. ^{57}Co in Rh matrix was used as a source. The width of Lorentzian lines measured with an iron foil was 0.25 mm s^{-1} . The temperature was varied in the cryostat working in closed-cycle refrigeration mode, in the temperature range from 12 K up to room temperature.

The x-ray measurements were carried out on an HZG-4C diffractometer in Białystok at room temperature only. The standard wavelength used was $\lambda = 1.5406$ Å.

The neutron diffraction experiments have been performed at Laboratoire Léon Brillouin in Saclay (France). The diffractometer G4.1 ($\lambda = 2.4249$ Å) was used for magnetic structure determination. The measurements were carried out in the temperature range of $1.5\text{--}270$ K. The crystal structure was measured above the phase transition temperatures on the 3T2 diffractometer ($\lambda = 1.2272$ Å) at $T = 300$ K for the samples of UFe_4Al_8 and UFe_5Al_7 and at $T = 400$ K for UFe_6Al_6 .

3. Results and discussion

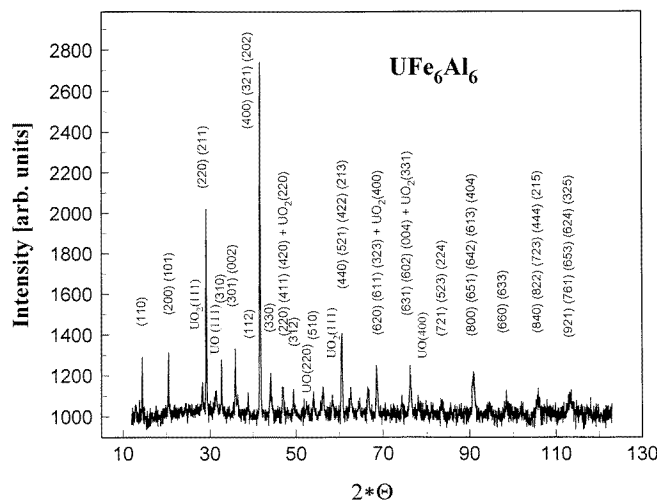
3.1. Magnetization measurements

The results of measurements at liquid nitrogen and room temperatures for all three samples do not deviate much from those published earlier by e.g. Andreev *et al* (1992) and are presented in table 1. Table 1 presents also transition temperatures estimated from our magnetization measurements. They agree well with the ones published by Vagizov *et al* (1995).

Table 1. The magnetic moments and phase transition temperatures measured for $\text{UFe}_x\text{Al}_{12-x}$ samples.

Alloy	77 K ($\mu_B \text{ fu}^{-1}$)	295 K ($\mu_B \text{ fu}^{-1}$)	Phase transition temperature
UFe_4Al_8	1.93	0.05	$T_N \approx 153 \text{ K}$
UFe_5Al_7	6.21	0.80	$T_C \approx 280 \text{ K}$
UFe_6Al_6	6.70	3.87	$T_C = 350 \text{ K}$

(Vagizov *et al* 1995)

**Figure 2.** X-ray powder diagram of UFe_6Al_6 at room temperature. Only some of the reflections have been indexed for better readability.

3.2. X-ray diffraction

The x-ray powder diagrams show that the UFe_4Al_8 sample is single phase, while the remaining two samples contain an extra phase (of FeAl type). The content of this phase increases with iron concentration and we estimate that it is $15 \pm 5\%$ in UFe_6Al_6 . In spite of this high content, the essential part of the diagram is interpreted well within the model of the sample with stoichiometric composition. This is easily explained because appropriate diffraction lines of UO and UO_2 are clearly visible, see figure 2. Both oxides appear in almost equal proportions. Their presence shows that together with FeAl, uranium precipitates as well. A substantially lower quantity of the extra FeAl-type phase in UFe_5Al_7 cannot be unambiguously determined because the peaks belonging to this phase are much less clearly seen.

The diagrams have been analysed by means of the Rietveld (DBWS *Rietveld Refinement Programs*) and FULLPROF (Rodriguez-Carvajal 1993) routines. The lattice constants, occupational and positional parameters obtained are almost the same as obtained from more precise neutron diffraction data, which will be described later. The agreement factors, R , for x-ray data are on the level of 4 to 5%. The values of parameters for UFe_4Al_8 agree with those published earlier by Dobrzyński *et al* (1996). The lattice parameters themselves agree with the data of Andreev *et al* (1992) and Suski *et al* (1992).

3.3. Neutron diffraction

The results of the neutron experiments carried out at temperatures above the phase transition points were analysed by the FULLPROF routine. The parameters of interest are listed in table 2. Both sets of x-ray and neutron data agree very well, which shows, in particular, that the presence of Fe–Al phase, especially in UFe_6Al_6 , is not posing any serious problem, at least as far as the nuclear part of the scattering is concerned. The agreement factors are as a rule much lower than in the case of x-ray diagrams. The only difference between the two sets of data is seen in the Debye–Waller factors determined by the two methods: the values obtained from neutron diffraction are larger. However, the essential trend, i.e. an increase of this factor with the iron content, is seen in both sets of results. We also note that if the atomic distribution model allows for some f–j disorder, one obtains the result that about 4% of iron resides at j-sites of UFe_4Al_8 .

Table 2. Results obtained from neutron experiment at 300 K for both UFe_4Al_8 and UFe_5Al_7 and at 400 K for UFe_6Al_6 .

Parameters	UFe_4Al_8	UFe_5Al_7	UFe_6Al_6	
Position	2(a)	0.996(4)	0.992(8)	1.008(8)
	8(f)	Fe 0.992(6)	Fe 0.999(6)	Fe 1.000(8)
Occupation		Al 0.008(6)	Al 0.001(6)	Al 0.000(8)
	8(j)	Fe 0.008(6)	Fe 0.250(6)	Fe 0.500(8)
		Al 0.992(6)	Al 0.750(6)	Al 0.500(8)
	8(i)	Fe 0.004(2)	Fe 0.000(5)	Fe 0.013(5)
		Al 0.986(2)	Al 1.000(5)	Al 0.987(5)
i ($x_1, 0, 0$)	$x_1 = 0.3443(2)$	$x_1 = 0.3432(4)$	$x_1 = 0.3445(5)$	
j ($x_2, \frac{1}{2}, 0$)	$x_2 = 0.2802(2)$	$x_2 = 0.2775(4)$	$x_2 = 0.2769(3)$	
Lattice constants	$a = 8.7365(2)$	$a = 8.6949(3)$	$a = 8.6583(4)$	
a and c (Å)	$c = 5.0302(2)$	$c = 5.0195(3)$	$c = 5.0070(4)$	
Debye–Waller factors	0.390(9)	0.477(14)	0.552(17)	
Agreement factor—R	0.011	0.030	0.040	
Contribution β -FeAl	—	0.008(4)	0.09(1)	

A typical neutron powder diagram of UFe_5Al_7 is shown in figure 3. The results of measurements carried out at various temperatures allowed one to investigate both the nuclear and magnetic scatterings. Within the accuracy of the experiment the nuclear part, i.e. all important structure parameters, is temperature independent except of the lattice constants. It is of interest to note that the lattice parameters depend non-linearly on temperature, and a typical dependence is presented in figure 4 for UFe_4Al_8 . One notes that the thermal expansion is negligible at low temperatures.

An interpretation of the magnetic part of the scattering requires separate presentation for every alloy studied. After the experience of Dobrzyński *et al* (1996), where irreversibilities were seen even in the neutron diffraction patterns, we have been particularly careful and made the measurements with the temperature decreasing as well as with the temperature increasing. No differences have been found for the diagrams measured at the same temperature, and therefore we treat our data as representative for the alloys of interest. In general the most difficult problem encountered in the magnetic structure determination consisted in a small level of magnetic contributions to predominantly nuclear peaks. No purely magnetic peak was detected for any of the samples. In such a situation we could hardly count on unique determination of the magnetic structures. A common feature in all

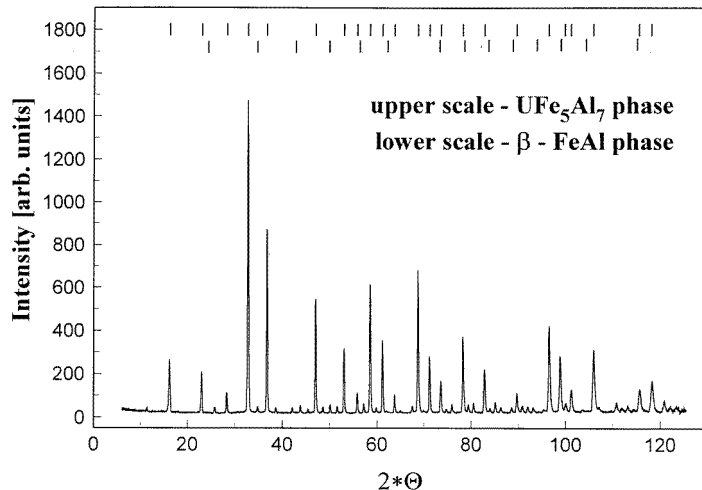


Figure 3. Neutron powder diagram obtained for UFe_5Al_7 at room temperature.

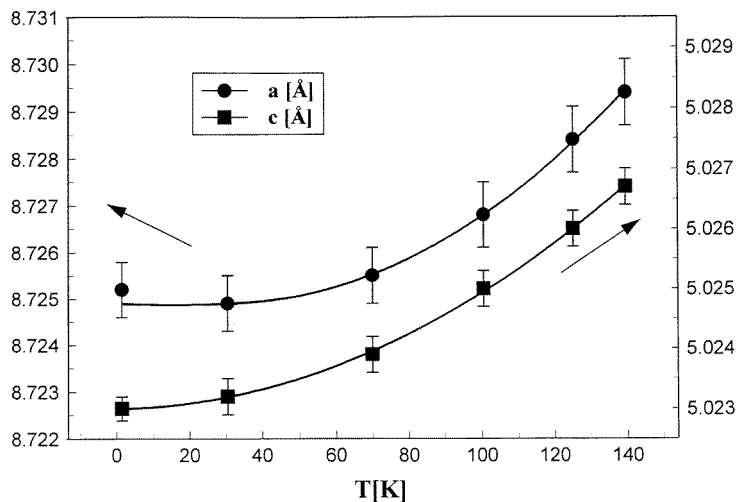


Figure 4. Lattice constants (in Å) against temperature measured for UFe_4Al_8 .

samples is that the magnetic moments lie, as expected, in the a - b plane. When the moments are directed along the $[001]$ direction, the quality of the fits substantially deteriorates.

3.3.1. UFe_4Al_8 . Because the intensities measured in our experiment were much higher than in any of the previously reported experiments on this compound (Ptasiewicz-Bąk *et al* 1988, Gal *et al* 1990, Dobrzyński *et al* 1996), we could attain much better statistical accuracy of the parameters of interest as well.

A typical value obtained for the magnetic moment of uranium atom was $\mu_U = 0.09(12) \mu_B$ at $T = 1.5$ K, and the nuclear (R_N) and magnetic (R_M) factors of agreement were equal to 0.014 and 0.050, respectively. Therefore, in further data treatments the

magnetic moments of uranium atoms have been set to zero (this resulted in $R_N = 0.016$ and $R_M = 0.052$ at the same temperature). The antiferromagnetically coupled Fe(f) moments with identical sequences on both parallel iron sheets above and below uranium (see figure 1) explain the neutron diffraction pattern well. The values of the magnetic moments obtained at various temperatures are presented in figure 5. The errors shown indicate a high degree of reliability of these values.

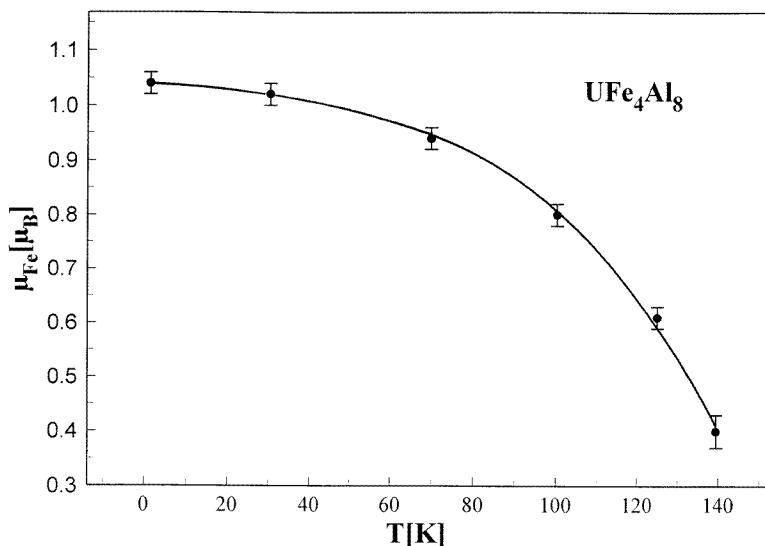


Figure 5. Iron magnetic moment against temperature measured in UFe_4Al_8 . The solid line is to guide the eye only.

A good fit of the model to the neutron data poses a natural question about the origin of the magnetic saturation moment of $1.93 \mu_B \text{ fu}^{-1}$ seen in magnetization measurements. Apparently, the postulated antiferromagnetic structure is not able to explain it, so a reasonable explanation is that the measured saturation moment is due to uranium atoms which become oriented in an external magnetic field. Without such a field the magnetization measurements are not possible.

Models in which a weak ferromagnetic moment of iron would be formed perpendicularly to the axis along which antiferromagnetic arrangement shows up neither improve the quality of fits nor bring magnetic ordering within the uranium sublattice. Nevertheless, a possibility of weak ferromagnetism within the a - b plane cannot be excluded. Such an ordering would be able to explain the origin of the spontaneous magnetization of the alloy.

3.3.2. UFe_5Al_7 . The analysis of the neutron diffraction pattern for this compound is biased by the results of the magnetization measurements which quite strongly indicate that we should deal with a regular ferromagnetic structure in this case. Indeed, the neutron diffraction patterns are well explained by the ferromagnetic structure with all moments lying in the a - b plane. An unambiguous description of magnetic order at j positions turned out to be not possible, so we assumed the same values of iron moments in both sites. This model works very well and the agreement factors are $0.022 \leq R_N \leq 0.025$ and $0.022 \leq R_M \leq 0.025$ in the whole temperature range. The temperature dependences of

magnetic moments obtained in this model are presented in figure 6. The obtained resultant magnetic moments per formula unit is equal to $6.0(4) \mu_B$, which is not far from the value measured in the magnetization measurements, see table 1. However, in light of the results of Gonçalves *et al* (1996) we can expect that the true saturation moments may be larger than inferred from the measurements on powder samples and the suggested structure may turn out to be inadequate. We shall return to this problem by the end of the next section.

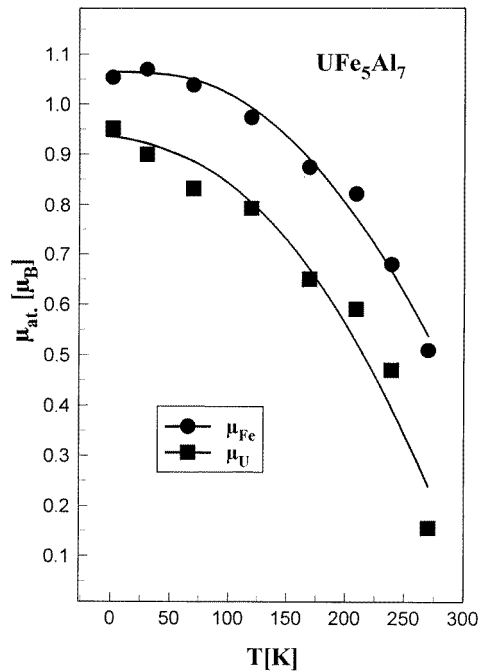


Figure 6. Iron and uranium magnetic moments against temperature measured in UFe_5Al_7 . The solid line is to guide the eye only. A typical error is $0.15 \mu_B$.

The magnetic moment of uranium at 12 K was found to be $0.96(12) \mu_B$, while the iron magnetic moment is $1.06(13) \mu_B$. The latter value agrees with the result of Mössbauer spectroscopy to be described in detail in the next section.

3.3.3. UFe_6Al_6 . Judging from the magnetization data, and the suggestion following from Andreev *et al* (1992) and Vagizov *et al* (1995) it was expected that the magnetic structure of this alloy should not be different from the one described above. To our surprise, the magnetic reflections, which in the case of a ferromagnetic ordering should strongly depend on magnetic moments per uranium located at a positions and iron located at j positions, turned out to be amazingly insensitive to the temperature in the whole covered temperature range of 5–240 K, see figure 7. Were the iron moments in the f sublattice ordered ferromagnetically, the only contribution to the (110) and similar type reflections would come from uranium and j iron sublattices. In order to reduce the number of parameters fitted we assumed again that the moments of Fe(f) and Fe(j) are the same. The agreement factors obtained were $0.020 \leq R_N \leq 0.023$ and $0.018 \leq R_M \leq 0.028$ in the whole temperature range. But even in this model the ratio of magnetic structure factor to the

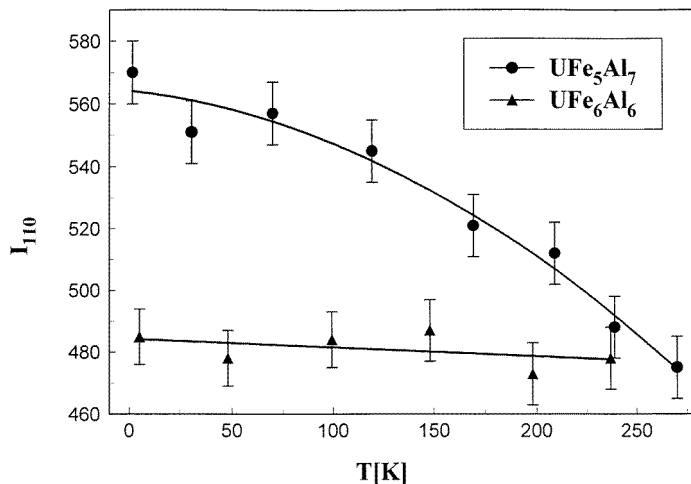


Figure 7. Integrated intensity of (110) against temperature measured in UFe_5Al_7 and UFe_6Al_6 . Solid lines are to guide the eye only.

nuclear one is very small, which leads to an impression that the (110) type reflections are temperature independent. The average magnetic moment of iron at low temperatures turned out to be $0.9(1) \mu_B$.

Within the model used no moment is seen on uranium atoms. The total magnetic moment observed by neutrons at the lowest temperature is about $5.4 \mu_B \text{ fu}^{-1}$ while $7.2 \mu_B \text{ fu}^{-1}$ is expected from Andreev *et al* (1992). Comparing the results obtained for all three samples we see that the uranium moment is $1.0(1) \mu_B$ but its ordering in zero field is seen in UFe_5Al_7 only. The saturation moment measured by Gonçalves *et al* (1996) for the composition of $UFe_{5.8}Al_{6.2}$ is as high as about $10.5 \mu_B \text{ fu}^{-1}$. Such a high value can hardly be reconciled with the existing neutron data.

3.4. Mössbauer spectroscopy

The spectra measured at room temperature for $x = 4$ and 5 samples show nonmagnetic quadrupole doublets, while for $x = 6$ a broad magnetically split pattern with a high intensity in the central part of the spectra was revealed. The spectra measured at 12 K with and without external magnetic field oriented parallel to beam direction are shown in figure 8. All spectra are magnetically split. The spectrum for $x = 4$ exhibits essentially a well resolved single sextet. Spectra for $x = 5$ and 6 are complex, which can be expected on the basis of a variety of possible local surroundings of iron atoms, which are found at f and partly at j sites. In order to obtain more information about positions of second and fifth lines in these samples, additional measurements with an external magnetic field of 1 T oriented parallel to the beam direction were carried out at 12 K.

In the analysis of the spectra of UFe_4Al_8 it has been initially assumed that the iron atoms locate exclusively at f positions, so the shape of Mössbauer spectrum can be described using one narrow component only. The spectra measured from room temperature down to about 155 K show nonmagnetic doublets with the quadrupole splitting independent of temperature and equal to 0.30 mm s^{-1} . From 80 K down to 12 K a well resolved single sextet appears. The temperature behaviour of the isomer shift can be ascribed to the

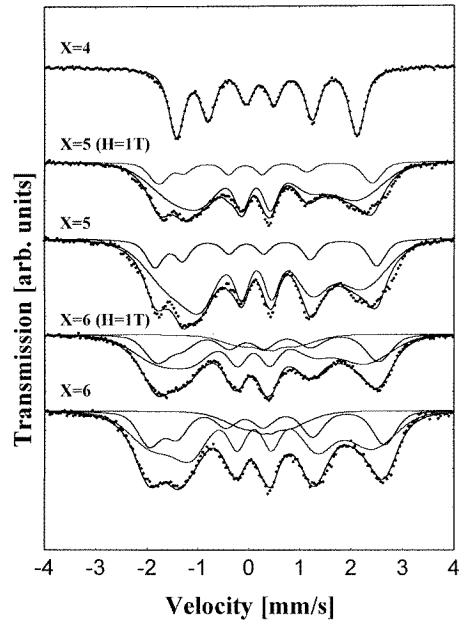


Figure 8. Mössbauer spectra measured at 12 K. Solid lines show the fitted curves.

second-order Doppler shift. The quadrupole splitting of the magnetic part of the spectra is independent of temperature and equal to 0.125 mm s^{-1} . The value of the hyperfine magnetic field (h.m.f.) decreases with temperature and reaches zero at about 155 K in good agreement with the data published by Gal *et al* (1990), Dobrzyński *et al* (1996) and with our neutron experiment. If we assume a linear relation between h.m.f. and magnetic moments of iron ($H = A\mu_{Fe}$), the coupling constant, A , estimated from low-temperature Mössbauer and neutron results is equal to $(10.6 \pm 0.1) \text{ T } \mu_B^{-1}$ for this class of alloys. This value of A was next used for estimation of magnetic moments of iron for samples with $x = 5$ and 6 (table 3).

Table 3. Mössbauer parameters for f and j iron sites at 12 K.

x	h.m.f. (T)	IS (mm s^{-1})	QS (mm s^{-1})	μ_{Fe} (μ_B)
f position				
4	(11 ± 0.2)	(0.285 ± 0.005)	(0.125 ± 0.002)	$(1.04 \pm 0.01)^a$
5	(11.6 ± 0.7)	(0.22 ± 0.02)	(0.19 ± 0.04)	(1.1 ± 0.1)
6	(12.5 ± 0.7)	(0.18 ± 0.02)	(0.17 ± 0.04)	(1.18 ± 0.10)
j position				
5	(13.4 ± 0.7)	(0.13 ± 0.03)	(0.48 ± 0.08)	(1.26 ± 0.10)
6	(14.3 ± 0.7)	(0.14 ± 0.03)	(0.41 ± 0.08)	(1.35 ± 0.10)

^a Iron magnetic moment from neutron data.

More complicated situations have been observed for UFe_5Al_7 and UFe_6Al_6 . The analyses of shapes of their spectra are based on the results of iron occupation obtained from x-ray and neutron measurements. Because in these alloys the excess iron locates predominantly in the j site, one can expect that the shapes of spectra must be described by

a superposition of at least two components. One component, with broad h.m.f. distribution, comes from iron at f positions (five different local surroundings) and the second one, narrower, from iron located at j positions (three different surroundings). The relative intensities of these components must correspond to the occupation of sites by iron: 80% at the f and 20% at the j site in UFe_5Al_7 , while these numbers for UFe_6Al_6 are 66% and 33%, respectively. Each component corresponds to a distribution of h.m.f., IS and QS. These distributions are not well resolved. Moreover, for UFe_6Al_6 one can expect the presence of a weak third component due to the extra phase seen in diffraction experiments.

The spectra for samples with $x = 5$ and 6, measured without and with external magnetic field (figure 8), were fitted with two Gaussian distributions of h.m.f. and for each sample were analysed simultaneously. The intensity ratios between f and j components were kept constant and equal to 4 and 2 for $x = 5$ and 6, respectively. An additional third component with a relative area of 12% was added in the case of $x = 6$. Such a broad component in the central part of the spectrum was also found by Vagizov *et al* (1995) for a sample of $UFe_{3.2}Al_{8.8}$ and was ascribed to the presence of an extra Fe–Al phase. The parameters obtained from the analysis are presented in table 3. As expected, the distributions connected with f positions are broader than the ones for j positions.

The h.m.f. of iron in the j position is slightly higher than observed for the f site. The isomer shift shows the opposite trend. The quadrupole splitting obtained for the j position of iron is much higher than the one for the f site. From the previously derived hyperfine constant, A , we estimated average magnetic moments for iron at j and f positions (see table 3). The results show that magnetic moments of iron both in f and j positions increase with iron concentration.

The total magnetic moments estimated from iron moments only in UFe_5Al_7 and UFe_6Al_6 are very close to the values of saturation moments obtained from magnetization data on powders. The situation, however, is far from simple. The moments listed in table 3 do not fit the neutron diffraction data, nor do the latter produce sensible values for an individual iron site if the magnetic moment at another site is fixed: the neutron powder diffraction pattern brings too limited information to solve the magnetic structure.

Remembering the differences between magnetization values obtained on a single crystal and on powders for UFe_6Al_6 we can say that the magnetic moments of iron which follow from the Mössbauer data analysis of the UFe_5Al_7 spectra, plus the magnetic moment of uranium found from the neutron data, sum up to a much more reliable value of the saturation moment per formula unit ($6.6(3) \mu_B$) than given by the neutron data alone. We are also fully aware of the fact that the hyperfine constant A , estimated for UFe_4Al_8 , which essentially exhibits antiferromagnetism, may not suffice to describe the hyperfine field–magnetic moment relation in the case of a ferromagnet, for which an extra term due to the diffuse moment may be needed. Therefore the somewhat higher magnetic moments of iron obtained in our analysis of the Mössbauer spectra cannot serve as valid information about the inconsistency of Mössbauer and neutron results.

We should finally remember that the saturation moment measured along a and b axes for the UFe_6Al_6 single crystal (Gonçalves *et al* 1996) is about $10.5 \mu_B \text{ fu}^{-1}$. Such a high value can hardly be reproduced by our neutron and Mössbauer data. One has to conclude again that the magnetic structure of UFe_6Al_6 must be complicated and contain probably canted iron moments. Due to the different characteristics of neutron and Mössbauer probes the sensed magnetic moments may be different. We believe that this happens in our sample.

Ending this section we want to comment on the differences between fitted and measured spectra for UFe_5Al_7 , see figure 8. In our opinion the reason for such deviations is that the shape of the distribution was assumed to be Gaussian and thus symmetric. Assuming a

binomial distribution of iron atoms in the *j* positions for given *x*, one can calculate the probability of particular surroundings for *f* and *j* iron positions. For the sample in question the distributions of probabilities in both *f* and *j* positions are apparently asymmetrical while for UFe_6Al_6 they are symmetrical (iron atoms occupy half of the *j* positions). The analysis using eight sextets (maximum number of sextets) with fixed intensities of subspectra gives very good agreement of fitted and experimental spectra, but we doubt the physical reliability of so many parameters obtained in this way. For the sample $x = 6$ the agreement between the model and experiment is satisfactory. This is connected with the aforementioned symmetrical distribution of h.m.f. for this sample. More detailed studies with the use of a polarized source (Szymański *et al* 1996) are in progress and results will be published in forthcoming paper.

The hyperfine field parameters obtained for $x = 5$ and $x = 6$ samples were next used for refinement of the spectra observed for $x = 4$ sample. Substantially better description of the UFe_4Al_8 spectra was obtained when a component due to iron at *j* sites was introduced. The amount of iron at this site appeared to be 5.3(5)% which agreed very well with the results of the analysis of the neutron data for this sample.

4. Summary

(1) The paper reports neutron diffraction measurements on UFe_4Al_8 , UFe_5Al_7 and UFe_6Al_6 compounds. The results for the last two samples are published for the first time in the literature. The Mössbauer spectra obtained for these samples with and without the magnetic field are also presented and analysed together for first time.

(2) From the structural features we mention that the increase of the iron content results in an increase of Debye–Waller factor which shows up in a systematic decrease of the lattice rigidity. The other feature of interest is a thermal lattice expansion, which is negligible at low temperatures.

(3) The magnetic structure changes with an increase of the iron concentration from predominantly antiferromagnetic to predominantly ferromagnetic. Neutron results confirm that the *a*–*b* plane is an easy magnetization plane in all three samples.

(4) In UFe_4Al_8 the uranium atoms, which are surrounded by eight iron atoms oriented antiferromagnetically (the antiferromagnetically coupled $\text{Fe}_{(f)}$ moments with identical sequences on both parallel iron sheets above and below uranium) exhibit probably a magnetic frustration. As follows from the neutron data, within the statistical accuracy of our fits, the ordered magnetic moment on uranium atoms indeed appears to be practically zero. An apparent disagreement between the total magnetic moments obtained from magnetization and neutron data can, however, easily be explained if it is assumed that even a weak magnetic field applied to the sample orders uranium moments. A canting of iron magnetic moments with the *a*–*b* plane cannot be excluded. A small disorder between *f* and *j* sites was detected by both neutron and Mössbauer techniques. Its role remains unclear as yet.

(5) UFe_5Al_7 seems a regular ferromagnet. As follows from neutron data, the ferromagnetic coupling exists between magnetic moments of iron atoms located at *f* sites and uranium atoms located at *a* sites, with a similar value of magnetic moment on uranium and iron atoms of the order of $1 \mu_B$ at low temperatures. The simplest model with assumed equality of magnetic moments of iron at *f* and *j* positions, and the ferromagnetic order in both sites, turned out to be most reasonable in the light of the neutron data. The Mössbauer data show that the hyperfine fields at both iron sites are different by about 14% (see table 3). This can be reconciled with the neutron diffraction results: equally good fit is obtained when

the ratio of magnetic moments at f and j sites is set to the value found from the Mössbauer studies.

(6) No magnetic moment on uranium atoms was detected in UFe_6Al_6 . This, however, can be accidental and due to the low magnetic signal observed in neutron diffraction. Again, the Mössbauer data show that the hyperfine field coupling with the magnetic moments of iron at f positions is by about 15% lower than that at j sites (see table 3).

(7) The presented data indicate that even in ferromagnetic-like samples there might be many non-collinearities and/or frustrations, especially in the uranium sublattice. In this context more magnetization and neutron data should be collected on single crystals.

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