Projected iron moments in UFe_4Al_8 investigated with monochromatic polarized Mössbauer radiation

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Abstract. UFe$_4$Al$_8$ was investigated using a monochromatic, circularly polarized Mössbauer source. The measurements are sensitive to the sign of the hyperfine magnetic field. The iron magnetic moment component along the direction of the applied magnetic field was determined. Quantitative analysis shows that the configuration of iron moments is not collinear and, on average, only about one fourth of the atomic iron moment contributes to the total magnetization in a randomly oriented powder exposed to a field of 1 T at a temperature of 12 K. The presence of ferromagnetic clusters in an otherwise antiferromagnetic ordering is discussed. The conclusions regarding the magnetic structure are compared with the results of powder neutron diffraction experiments. In the course of the interpretation of latter, the Debye temperature $\theta_D = 421(13)$ K was determined.

1. Introduction

Many attempts have been made towards an understanding of the magnetic structure of UFe$_4$Al$_8$ (Suski 1990, Gal *et al* 1990, Andreev *et al* 1992, Godinho 1995, Dobrzyński *et al* 1996, Rećko *et al* 1996, 1997a, b). The UFe$_4$Al$_8$ compounds crystallize in the tetragonal ThMn$_{12}$ type structure (space group $I4/mmm$) where the thorium site $2a - (0, 0, 0)$ is occupied by a uranium atom, while the manganese sites $8f - (\frac{1}{4}, \frac{1}{4}, \frac{1}{4})$ are occupied by iron atoms and $8i - (x_1, 0, 0)$ as well as $8j - (x_2, \frac{1}{2}, 0)$ by aluminium (I), respectively. The magnetic phase transition temperature was found to be close to 155 K (Gal *et al* 1990, Godinho 1995, Dobrzyński *et al* 1996, Rećko *et al* 1996, 1997a, b). The magnetic and nuclear unit cells are identical, so the magnetic intensities (unfortunately weak) occur only at positions of the nuclear Bragg peaks. Only recently, weak ferromagnetism was discovered in the single crystal sample of our interest (Paixǎo *et al* 1997). According to this paper, U atoms have magnetic moment of 0.47(2) μ_B and are oriented ferromagnetically in the basal plane along one of the equivalent directions $a = [100]$ or $b = [010]$. When the magnetic moments of uranium atoms are oriented, say, along a axis, the iron moments of 1.08(2) μ_B are predominantly oriented antiferromagnetically along the b axis. A small ferromagnetic component found along the a direction in the field of 4.6 T indicates that the atomic iron moments are inclined by 25° with respect to b. Keeping the total value of uranium and iron moments fixed, Paixǎo *et al* (1997) estimated this angle in zero field to be 16° which can explain the value of the remanence of magnetization in the easy (ab) plane of 1.6(2) μ_B mol^{-1} at 4.2 K.

On the other hand, Rećko *et al* (1996), who carried out powder neutron diffraction in zero field, found no evidence either of a uranium moment or of a weak ferromagnetic component in the iron sublattice. Comparing the two sets of data, a difference of the magnetic moments

per formula unit reported by Paixǎo *et al* (1997) for the single crystal and by Rećko *et al* (1996, 1997a, b) for the powder samples is to be noticed. The value measured for a powder is 1.93(2) μ_B at $T = 77$ K (Rećko *et al* 1996). An equally high value was also found by Andreev *et al* (1992).

These differences motivated us to carry out an experiment with the use of a monochromatic, circularly polarized Mössbauer source (MCPMS). It was demonstrated earlier that with a ^{57}Co MCPMS and a polycrystalline sample one can easily and unambiguously show the antiparallel ordering of Fe and Ho moments in ferromagnetic HoFe$_2$ alloy (Szymański *et al* 1996). This new technique was applied here for investigation of the magnetic structure of the UFe$_4$Al$_8$ powder sample studied in previous experiments (Rećko *et al* 1996, 1997a, b).

The paper is organized as follows. We start from a re-analysis of our standard Mössbauer results on UFe$_4$Al$_8$ and then the construction of the MCPMS is given. Next, we describe preparation of the UFe$_4$Al$_8$ absorber and a procedure for the determination of absorption line intensities, from which the projection of the Fe magnetic moment on the applied field direction was determined. Finally, we re-analyse the neutron powder diffraction data by Rećko *et al* (1996). Special emphasis is put on the problem of interpretation of weak magnetic scattering amplitudes and thermal vibration effect. We also discuss the influence of the stoichiometry on the magnetic properties of the alloy. The results are compared with those of Paixǎo *et al* (1997) and Rećko *et al* (1996, 1997a, b).

2. Standard Mössbauer results on UFe$_4$Al$_8$

Preliminary Mössbauer results obtained for UFe$_x$Al$_{12-x}$ with $x = 4, 5$ and 6 were presented by Rećko *et al* (1996). It was assumed that the spectrum of UFe$_4$Al$_8$ consists of only one magnetic component, due to iron in the so-called f-positions. Although a reasonable description of the spectrum was obtained, some weak discrepancies between the model and the experimental points have to be kept in mind (see figure 8, Rećko *et al* (1997)) have to be kept in mind. Therefore, in this paper we also consider a possibility of the presence of iron atoms at the so-called j-positions. We also examine consequences of a precipitation of the β-FeAl phase in our UFe$_4$Al$_8$ sample, expecting that both effects must be small.

The Mössbauer spectra measured at 12 K were thus fitted with two magnetic components and one quadrupole doublet. Each magnetic component corresponded to a Gaussian distribution of the hyperfine magnetic field. Because the component corresponding to iron at j-positions is weak, the hyperfine parameters for such iron were first assumed to be the same as found in the samples with $x = 5$ and 6. In the next step these hyperfine parameters were taken as 'free'. The contribution from the β-FeAl phase was fitted by a doublet, with parameters equal to those given by Steiner *et al* (1996). A typical fitted spectrum is shown in figure 1(a) where two weak iron components are drawn separately. The analysis shows that $4 \pm 1\%$ of the iron locates at the j-positions, $3 \pm 1\%$ forms the β-FeAl phase and the remaining iron atoms locate at the f-positions of UFe$_4$Al$_8$.

In the previous investigation of the family of UFe$_x$Al$_{12-x}$ alloys, the x-ray and neutron diffraction patterns showed the presence of about 15% of the β-FeAl phase for $x = 6$ and traces for $x = 5$ (Rećko *et al* 1996). No evidence of this phase was found in the neutron diffraction pattern of UFe$_4$Al$_8$. However, our Mössbauer data also show that in the case of UFe$_4$Al$_8$ the presence of a β-FeAl phase cannot be excluded. This only shows that the sensitivity of the diffraction methods to such small amounts of the other phase may be less than the sensitivity of the Mössbauer technique. The β-FeAl phase creates precipitations of small dimensions so that they are only very weakly visible in the x-ray diffraction, but they can be sufficient for Mössbauer measurements.
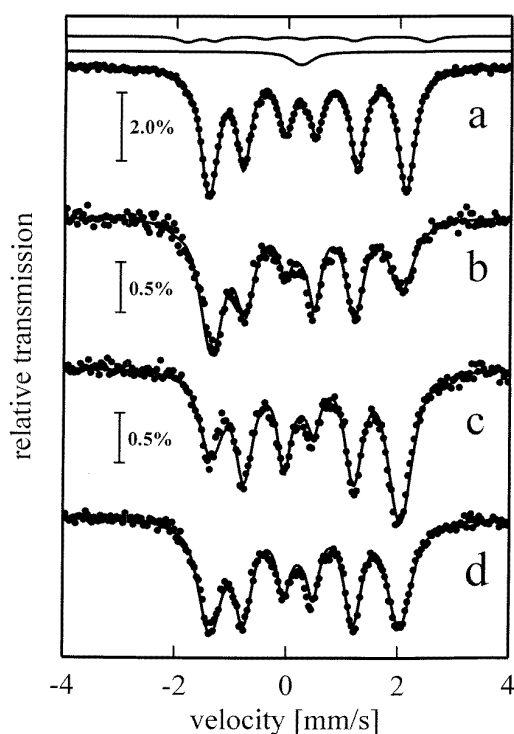
Figure 1. (a) Mössbauer spectrum of UFe$_4$Al$_8$ measured at 12 K in zero magnetic field. Two solid lines above experimental points represent a small fraction of the iron atoms at j-sites and the β-FeAl phase, respectively. (b) and (c) Mössbauer spectra of UFe$_4$Al$_8$ measured with MCPMS for two helicities. (d) The sum of the spectra measured for two helicities.

3. MCPMS measurements

Monochromatic, circularly polarized Mössbauer source (MCPMS) constructed and installed at the Institute of Physics in Bialystok uses a commercial ^{57}Co source in Cr matrix with an initial activity of 50 mCi. The filter technique is applied to absorb-out resonant photons with one polarization. The linewidth of the inner lines of an iron foil with a thickness of 6 μm is 0.25 mm s^{-1}. A full description of the source was given in Szymański *et al* (1996).

The sample used was the same as in the experiment of Rećko *et al* (1996). The sample powder was mixed with epoxy glue and a disc containing UFe$_4$Al$_8$ 7.4 mg cm^{-2} was formed. The disc was placed inside the bore of the permanent magnet. Because of rigidity of the sample its grains could not perform any rotation in the field. The magnetic field of 1 T acting perpendicular to the sample plane (and parallel to the γ-rays), was produced by a rare-earth permanent magnet.

The sample temperature in the experiment, 12 K, was maintained by a commercial closed cycle helium refrigerator, equipped with the Mössbauer shroud preventing sample vibrations. The broadening of the absorption lines caused by refrigerator vibrations was less than 0.01 mm s^{-1}.

The spectra of UFe$_4$Al$_8$ measured with two opposite, incomplete ($80 \pm 2\%$) polarizations are shown in figures 1(b) and (c), while the sum of the spectra for two polarizations is shown

Table 1. The line intensities obtained from fits shown in figures 1(b) and (c) are listed in columns 2 and 3 whereas the intensities corrected for incomplete source polarization are given in columns 4 and 5.

Line no (i)	$I_i \uparrow\uparrow$	$I_i \uparrow\downarrow$	$i_i \uparrow\uparrow$	$i_i \uparrow\downarrow$
1	0.317(4)	0.167(3)	0.352(5)	0.141(5)
2	0.171(3)	0.171(2)	0.171(4)	0.171(4)
3	$1/3I_6$	$1/3I_6$	$1/3i_6$	$1/3i_6$
4	$1/3I_1$	$1/3I_1$	$1/3i_1$	$1/3i_1$
5	I_2	I_2	i_2	I_2
6	0.177(4)	0.327(3)	0.142(5)	0.353(5)

in figure 1(d). The sum is equivalent to the spectrum measured with unpolarized radiation in an external, longitudinal field. The spectrum was analysed in a similar way as described in section 2. The hyperfine magnetic fields of the magnetic components were set to be free parameters in the fit while the isomer shifts and quadrupole splittings were fixed at the values found in the zero field experiment. For the description of the β-FeAl component we used the routine of Blaes *et al* (1985) which describes the shape of Mössbauer spectrum in an external magnetic field under the assumption that the principal axes of the electric field gradient (EFG) tensor are randomly oriented. The resulting fitted line is shown in figure 1(d). Next, keeping all the parameters corresponding to the hyperfine structure fixed, fits of the spectra shown in figures 1(b) and (c) were performed. The only free parameters were the line intensities constrained so that $I_1 = 3I_4$, $I_2 = I_5$ and $I_6 = 3I_3$, see columns 2 and 3 in table 1. Then using equation (20) of Szymański (1998) and the degree of polarization ($80 \pm 2\%$) measured in the way presented by Szymański *et al* (1996), we estimated the line intensities for a fully polarized beam, see columns 4 and 5 in table 1.

4. Configuration of the iron moments

The iron moment arrangement can be described by the magnetic texture function $P(\Omega)$, being the probability of finding a local magnetization vector directed along the direction described by $\Omega = (\theta, \varphi)$. $P(\Omega)$ can be expanded into the sum of real spherical harmonics $y_{lm}(\theta, \varphi)$ multiplied by coefficients A_{lm}. Mössbauer spectroscopy brings some information about $P(\Omega)$, for example in the case of the $I_e = \frac{3}{2} \rightarrow I_g = \frac{1}{2}$ transition, the first nine coefficients A_{lm} can be determined (Pfannes and Fisher 1977). The A_{10} coefficient is proportional to the average projection of the magnetic moment on the z axis, μ_z. By definition

$$\mu_z = \mu_{Fe} \int_{sphere} \cos\theta \, P(\Omega) \, d\Omega \tag{1}$$

where θ is the angle between the moment and the radiation direction and μ_{Fe} is the magnetic moment of iron. It is assumed in equation (1) that the value of μ_{Fe} is independent of the direction of radiation.

In the case of the geometry described in section 3, the line intensities in the sextet measured with MCPMS are equal to (Frauenfelder *et al* 1962)

$i_1 : i_2 : i_3 : i_4 : i_5 : i_6$

$$= \tfrac{3}{16}(1\pm\cos\theta)^2 : \tfrac{1}{4}\sin^2\theta : \tfrac{1}{16}(1\mp\cos\theta)^2 : \tfrac{1}{16}(1\pm\cos\theta)^2 : \tfrac{1}{4}\sin^2\theta : \tfrac{3}{16}(1\mp\cos\theta)^2 \tag{2}$$

where θ is the angle between the spin and the direction of radiation. The upper and lower signs correspond to the two opposite photon helicities. The difference between intensities of the first and sixth lines is particularly interesting because it is proportional to the projection moment onto the direction of the applied magnetic field. In terms of the distribution of the directions of the moments one has

$$i_1 - i_6 = \frac{3}{16} \int_{sphere} 4\cos\theta \, P(\Omega) \, d\Omega. \tag{3}$$

Combining (1) and (3) and inserting appropriate values from table 1 one gets

$$\mu_z = \tfrac{4}{3}(i_1 - i_6)\mu_{Fe} = 0.28(1)\mu_{Fe}. \tag{4}$$

In order to check the sensitivity of result (4) on the introduced assumption concerning the presence of a β-FeAl phase, an additional fit, in which β-FeAl content was set to zero was performed. Within the experimental accuracy the result for μ_z was the same. We thus conclude that the obtained value, from (4) is insensitive to our low β-FeAl content.

The result from (4) informs us about the angular distribution of magnetic moments of iron and, in principle, is independent of the information given by the hyperfine magnetic field. Strictly speaking, the difference of the line intensities i_1–i_6 is proportional to the projection of the iron moment on the direction of the beam. We should also point out that the shape of the measured Mössbauer spectrum clearly indicates that almost all iron atoms have the same value of magnetic moment. Thus the assumption made in (1) is fulfilled. In summary, from measurements with the polarized source, it follows that in the applied field of 1 T the contribution of iron to the magnetization is equal to 0.28(1) of the total magnetic moment of iron.

5. Re-analysis of neutron powder diffraction data

The results obtained from the measurements with polarized Mössbauer source motivated us to carry out a re-analysis of the data published earlier by Rećko *et al* (1996), where pure antiferromagnetic configuration within an iron sublattice and uranium magnetic frustration was claimed. Original neutron powder diffraction experiments were performed at LLB (Saclay, France) using the high-resolution 3T2 diffractometer with $\lambda = 1.2272$ Å and the G4.1 diffractometer with $\lambda = 2.4249$ Å. With the former, the diffraction pattern was recorded at room temperature only, at which pure nuclear scattering was observed. The magnetic contributions to diffraction were analysed from 1.5–218 K by using the data from the G4.1 diffractometer. Obviously, the weak magnetic contributions to the strongest peaks of predominantly nuclear origin are difficult to interpret, so we tried to limit the weight of such an information. Because of generally small magnetic scattering, special care was also taken to correct the neutron diffraction pattern for the $\lambda/2$ contribution. The analysis of the magnetic part of the spectrum was limited to the angular range in which peaks are well resolved (see figure 2). The neutron spectra were analysed by means of the Rietveld refinement method with the aid of the computer program Fullprof (Rodriguez-Carvajal 1993).

Another problem, important for the interpretation of weak magnetic intensities, is connected with correct estimation of the temperature dependence of the Debye–Waller factors. In order to separate out the magnetic scattering from thermal effects, the Debye–Waller factor dependence on temperature had to be accurately determined. The Debye temperature of UFe$_4$Al$_8$ was, to the authors' best knowledge, never reported, so we made an attempt to determine it. From the neutron measurements carried out on G4.1 diffractometer the Debye–Waller factor, obtained at 218 K, was B $= 0.307(59)$ Å^2. This value leads to
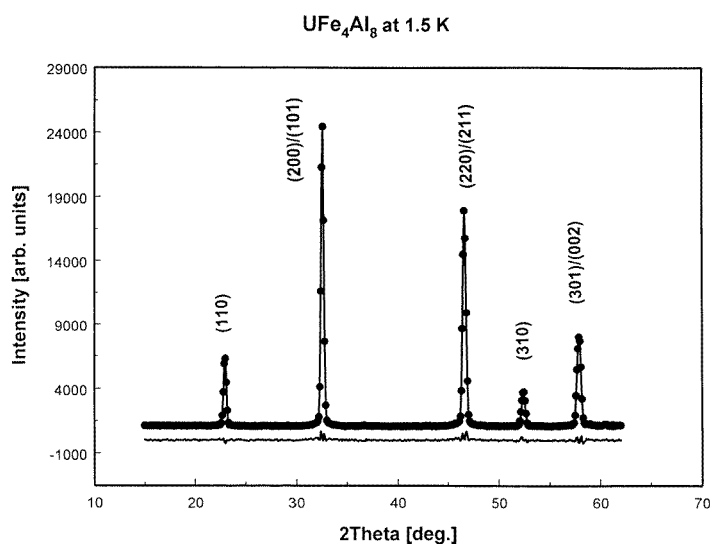
UFe$_4$Al$_8$ at 1.5 K

Figure 2. Observed and calculated neutron diffraction patterns of powdered UFe$_4$Al$_8$ for $\lambda = 2.4249$ Å. In the lower part the difference between experiment and calculations is shown.

$\theta_D = 414(80)$ K. The high-resolution neutron experiment at 300 K allowed to obtain the appropriate B equal to 0.400(9) Å^2 what results in $\theta_D = 420(13)$ K. The analysis of the thermal expansion of the volume of the unit cell should allow for an independent estimation of the θ_D using the Debye function and Thacher approximation (see Thacher 1960), according to the method proposed by Sayetat *et al* (1998) were employed. From the measured dependence of the lattice parameters on temperature, the obtained value of θ_D was 426(39) K. The weighted average of θ_D is 421(13) K. Knowing θ_D, we could calculate thermal corrections to the neutron intensities. Next, we could check to what extent the magnetic structure determined by Paixão *et al* (1997) can explain our data. Rietveld's type refinement, shown in figure 2, allowed one to obtain the best fit with the uranium magnetic moment and ferromagnetic and antiferromagnetic iron moment components equal to: $\mu_U = 0.351(15)$ μ_B; $\mu_{Fe(ferro)} = 0.328(9)$ μ_B and $\mu_{Fe(antiferro)} = 1.024(2)$ μ_B, respectively. This set of values gives excellent standard agreement factors for the nuclear and magnetic parts of the neutron diffraction pattern, being $R_N = 0.044$ and $R_M = 0.011$. The appropriate nuclear and magnetic R-factor is defined as the conventional $_{Bragg}RB$ factor in (5) and is used to estimate the quality of the agreement between observed and calculated nuclear and magnetic intensities, respectively

$$_{Bragg}RB = 100 \frac{\sum_k |I_{observed} - I_{calculated}|}{\sum_k |I_{observed}|}. \tag{5}$$

Obviously, the total magnetic moment of iron calculated from the quoted values, according to the formula $\sqrt{(\mu^2_{Fe(ferro)} + (\mu^2_{Fe(antiferro)})}$, must be 1.075(5) μ_B. Thus, from (4) we can expect the ferromagnetic component $\mu_{Fe(ferro)}$ to be 0.30(2) μ_B, in good agreement with the neutron value just quoted as well as the one given by Paixão *et al* (1997).

The neutron data (figure 2) indicate unambiguously the presence of uranium and iron ferromagnetic components at the lowest temperature. However, it is worth noting that in contrast to Paixão *et al* (1997) we do not see the magnetic moment on the uranium atom above 30 K, which is very close to the second transition temperature $\sim$35 K, found for the powdered UFe$_4$Al$_8$ sample by Dobrzyński *et al* (1996). It is also known from the older papers

(van der Kraan and Buschow (1977), Buschow and van der Kraan (1978)) that ordering of rare-earth atomic moments in isostructural $RFe_{4+x}Al_{8-x}$ compounds (R = rare earth) takes place in a similar temperature range. However, based on the magnetization measurements we could find no reason to expect different transition temperatures for uranium and iron. Therefore, either the magnetic moment of uranium gives such a small contribution to both magnetization and neutron data that it can easily be overlooked, or the ferromagnetic interactions between uranium and iron atoms in the predominantly antiferromagnetic system are too weak to align uranium moments at temperatures higher than ∼30 K.

It is fortunate that relatively high intensities measured in the experiment of Rećko *et al* (1996) allow one to make fair estimations of the magnetic structure based on unpolarized neutron powder diffraction data, providing that careful corrections for the $\lambda/2$ contribution and effects of thermal vibrations are made. The magnetic model, assuming the presence of ferromagnetic interactions in UFe_4Al_8 alloy, is clearly better than the one with purely antiferromagnetic ordering. The short description of the magnetic structure factors allow one to simply explain the origin of differences between the aforementioned magnetic models. The appropriate magnetic structure factors F_M are as follows:

$$F_{M(110)} \sim 2\mu_U - 8\mu_{Fe(antiferro)}; \ F_{M(310)} \sim 2\mu_U + 8\mu_{Fe(antiferro)}.$$

These two reflections appear as singlets in the diffraction pattern and give the value of the antiferromagnetic iron component which does not depend on the applied model. From the point of view of the magnetic model, the intensities of the unresolved double reflections occur to be much more important. We note that for three doublets of interest one has:

$$F_{M(200)} \sim 2\mu_U - 8\mu_{Fe(ferro)} \quad \text{and} \quad F_{M(101)} \sim 2\mu_U$$
$$F_{M(220)} \sim 2\mu_U + 8\mu_{Fe(ferro)} \quad \text{and} \quad F_{M(211)} \sim 2\mu_U$$
$$F_{M(002)} \sim 2\mu_U - 8\mu_{Fe(ferro)} \quad \text{and} \quad F_{M(301)} \sim 2\mu_U.$$

Thus we may conclude that if the canting of iron moments disappeared, the integrated intensities of all these six reflections should become temperature independent, especially above 30 K, where no ordered magnetic moment was observed on uranium. As we shall see in figures 3(a) and (b), which illustrate the temperature behaviour of the integrated intensity of the (200/101) and (220/211) reflections, respectively, this is not the case. These reflections contain the magnetic contribution up to $T_{C,N} = 155$ K, i.e. up to the phase transition temperature determined in the magnetization studies.

Our results thus fully confirm the existence of weak ferromagnetism in powdered UFe_4Al_8.

6. Discussion

Using the neutron results we calculate that the iron contribution to the magnetization is $4\mu_z = 4 \times 0.33(1) = 1.32(4) \ \mu_B$ per molecule. However, the magnetic moment measured by us for powdered UFe_4Al_8 at 77 K for the magnetic field extrapolated to zero temperature turned out to be 1.93 μ_B per molecule (Rećko *et al* 1996). From the magnetization measured in the field of 1 T it follows that this moment raises to 2.25(6) μ_B (Rećko *et al* 1997). The excess moment in field of 1 T should thus belong to uranium and be 0.93(8) μ_B. This agrees very well with the value of 0.96(12) μ_B obtained for μ_U in UFe_5Al_7 in the neutron diffraction experiment (Rećko *et al* 1996). However, it differs from the moment of uranium of 0.5 μ_B obtained by (Paixão *et al* 1997) for the single crystal sample, and 0.35(2) μ_B in the present paper for our powder sample. Although the saturation magnetization of our sample is higher than the one in the experiment of Paixão *et al* (1997), the ferromagnetically oriented moments of U and Fe atoms seem smaller.
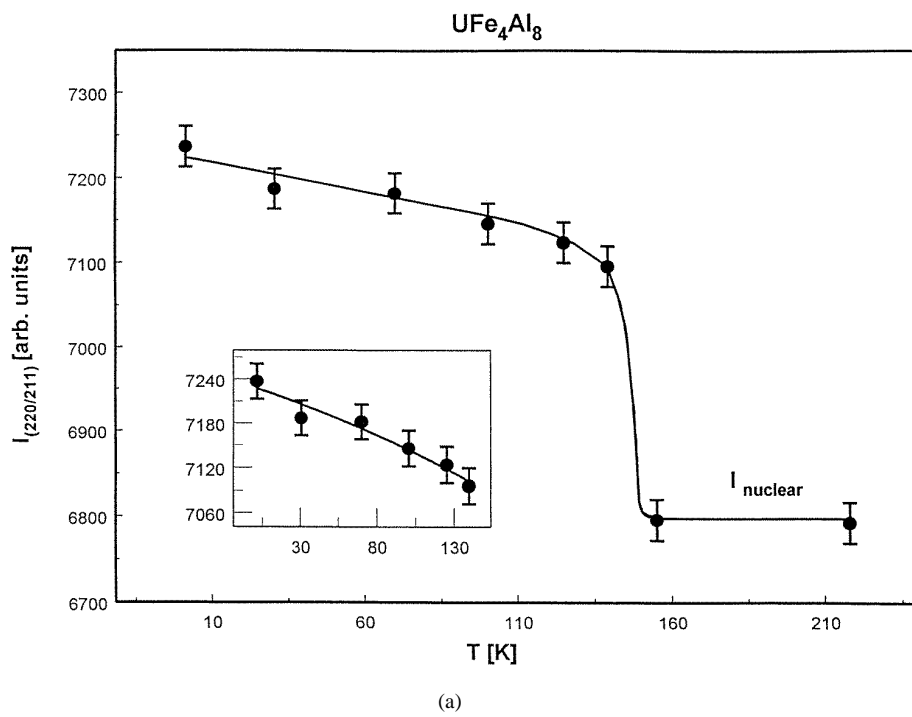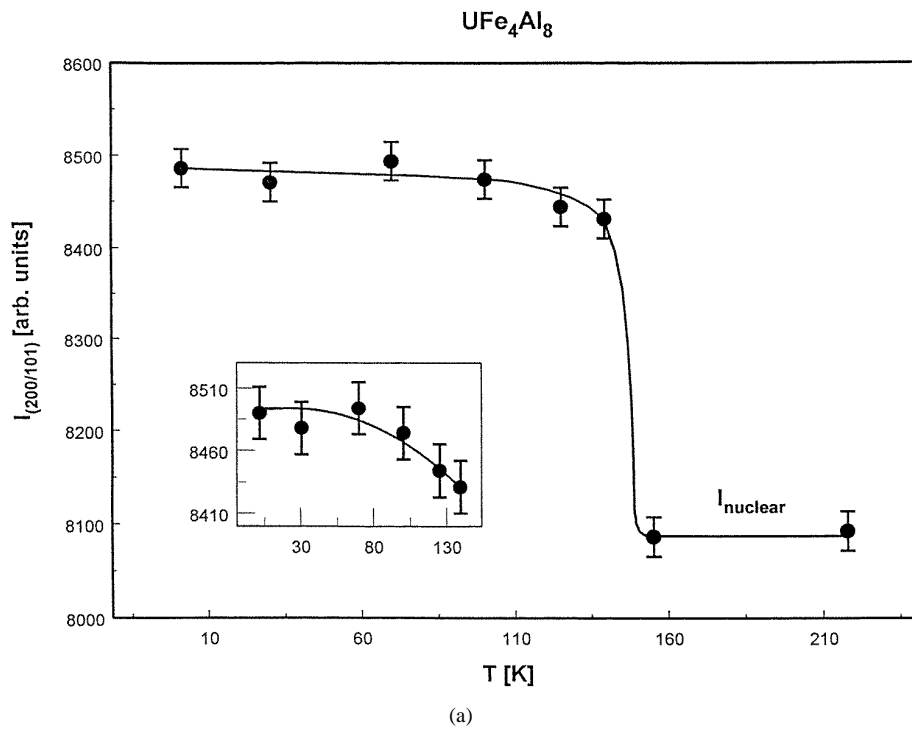
Figure 3. Temperature dependence of the magnetic reflections (a) (200)/(101); (b) (220)/(211). The inset shows the behaviour of magnetic intensities in temperature region 1.5–140 K. The full curves are as guides to the eye only.

The different saturation magnetizations of the different samples can arise from possible differences in the stoichiometries. Indeed, the magnetization measurements of UFe$_x$Al$_{12-x}$ compounds (Andreev *et al* 1992) clearly revealed that the magnetization is very sensitive to the alloy composition. For x slightly larger than 4.0 it follows from figure 4 of Andreev *et al* (1992) that dμ/dx in the field of 1.2 T at 4.2 K is not smaller than 10 μ_B. An increase of x by 0.1 thus corresponds to the change of magnetic moment per molecule of UFe$_4$Al$_8$ by one Bohr magneton! At elevated temperature this value must be even higher because the magnetic transition temperature increases with x. On the other hand, the magnetization does not change with lowering x below the level of $x = 4.0$. One can thus expect a high sensitivity of magnetic properties on the sample stoichiometry and homogeneity.

The only way to allow for changes of the composition is to let the occupational numbers be free parameters, unconstrained by the sample stoichiometry. If this is done, the neutron and x-ray data can be analysed with the isotropic Debye–Waller factor as in Rećko *et al* (1996) or using the site values obtained by Paixão *et al* (1997). It turns out that the choice of the Debye–Waller factors is not very important and the occupation numbers obtained are the same (within the experimental errors) for both. However, while the neutron results would allow for $x = 4.06(3)$, the x-ray data show an opposite trend. Because neutrons deliver information from all the sample volume we are inclined to believe more in the neutron results than the x-ray ones. Nevertheless, one cannot say that we have any clear-cut evidence that the sample stoichiometry is different from the nominal one.

According to the results of Paixão *et al* (1997) the polarized neutron experiment in a magnetic field 4.6 T ∥ [010] established that the ferromagnetic uranium moment is 0.47(2) μ_B per atom at 4.2 K. In the field of 1.5 T at 4.2 K the uranium moment was determined to be 0.45 μ_B. One can expect from the linear approximation of μ_U against applied magnetic field, that in the absence of the magnetic field the moment of the uranium atom should be equal to 0.44 μ_B, which is larger than 0.35 μ_B obtained from our unpolarized neutron experiment.

Comparing the neutron results with the magnetization measurements it can be seen that both values of uranium moment are too small to explain the total moment per UFe$_4$Al$_8$ molecule observed by us. This leads us to the conclusion that the ordering of the uranium sublattice observed by us in zero field must be incomplete. The easiest explanation of such a behaviour is that the uranium moments at the a-site are partly frustrated by the antiferromagnetically coupled eight iron atoms surrounding uranium. This, together with a weak ferromagnetic U–Fe coupling may be responsible for the relatively quick disordering of uranium moments with increasing temperature.

One has to understand why the neutron results obtained by Paixão *et al* (1997) and by us are different when the value of the uranium moment is concerned. We recall that the ordered moment of uranium was observed by Paixão *et al* (1997) at temperatures up to the Curie point. We may suspect that due to a certain disorder observed in our sample the phenomena observed might arise from the presence of ferromagnetic clusters, which are weakly bound to the basically antiferromagnetic system. In the applied field (this is the case of magnetization measurements and these were experimental conditions of Paixão *et al* (1997)) the clusters order, which in turn may cause an ordering of the uranium moments. It follows from the single crystal measurements of Paixão *et al* (1997) that the canting depends on an applied magnetic field. One can expect a similar behaviour in our powdered sample. The applied magnetic field induces a larger ferromagnetic component within the iron sublattice and this in turn may cause a ferromagnetic ordering of the uranium moments. This would qualitatively explain the difference between the magnetic moment per molecule obtained in neutron and magnetization measurements.

7. Summary

Re-analysis of the powder neutron diffraction data collected in zero external field (Rećko *et al* 1996) confirms the result of Paixăo *et al* (1997), that the uranium sublattice is ferromagnetically ordered, while iron sublattice exhibits weak ferromagnetism in the (*ab*) plane. The ordered uranium magnetic moment was determined to be 0.35 μ_B at 1.5 K. This moment vanishes at temperatures greater than 30 K.

The problem of the magnetic properties of UFe_4Al_8 discussed in this paper may be summarized as follows.

(1) The different values of the magnetic saturation moments measured by us and Bonfait *et al* (1996) could most easily be explained by a possible difference in the sample stoichiometries. Slightly higher iron content (about 2.5%) or deficiency of aluminium would explain the difference. Our attempts to find such deviations were unsuccessful. However, we noted that the neutron diffraction pattern could be well described if the required change of the stoichiometry was assumed.

(2) The magnetic phase transition temperature, 153 K, is close to the values reported for UFe_4Al_8. We note, however, that a change of x from $x = 4$ to $x = 4.1$ would cause a shift of the Curie temperature not larger than about 10 K, which is comparable with the accuracy of the determination of the phase transition temperature.

(3) Application of the circularly polarized Mössbauer source brought direct experimental evidence that approximately one-fourth of the total iron magnetic moment contributes effectively to the magnetization in the direction of the applied field of 1 T. Our present Mössbauer data and the results based on the Rietveld type refinement of the neutron data are consistent with the model of canted iron spins with the ferromagnetic component of iron moments of about 0.3 μ_B, as given by (Paixăo *et al* 1997).

(4) In order to reconcile the values of the total magnetic moments with the ferromagnetic moments found on the iron atoms one has to assume that the uranium atoms exhibit a moment of 0.93(8) μ_B. This value agrees with the one found in ferromagnetic UFe_5Al_7 (Rećko *et al* 1996). In contrast, a much lower moment (0.35 μ_B at 1.5 K) is visible in the neutron powder diffraction pattern of UFe_4Al_8. We conclude that a certain disorder of magnetic moments of uranium must be present in zero field.

(5) The discrepancy described above could be understood if ferromagnetic clusters were present in the sample. Such clusters would better order their moments under influence of the magnetic field. This ordering could trigger the ferromagnetic ordering of uranium moments.

Acknowledgments

References

Andreev A V, Nakotte H and Boer F R 1992 *J. Alloys Compounds* **182** 56
Baran A, Suski W and Mydlarz T 1984 *J. Less-Common Met.* **96** 269
Blaes N, Fisher H and Gonser U 1985 *Nucl. Instrum. Methods* B **9** 201
Bonfait G, Godinho M, Estrela P, Gonçalves A P, Almeida M and Spirlet J C 1996 *Phys. Rev.* B **53** R480
Buschow K H J and van der Kraan A M 1978 *J. Phys. F: Met. Phys.* **8** 921

Dobrzyński L, Satula D, Fjellvag H, Hauback B C, Baran A, Suski W, Wochowski W and Lebech B 1996 *J. Alloys Compounds* **236** 121

Frauenfelder H, Nagle D E, Taylor R D, Cochran D R F and Visscher W M 1962 *Phys. Rev.* **126** 1065

Gal J, Yaar I, Regev D, Arbaboff E, Etedgi H, Litterst F J, Aggarwal K, Pereda J A, Kalvius G M, Schäfer W and Will G 1990 *Phys. Rev.* B **42** 8507

Godinho M, Bonfait G, Gonçalves A P, Almeida M and Spirlet J C 1995 *J. Magn. Magn. Mater.* **140–144** 1417

van der Kraan A M and Buschow K H J 1977 *Physica* B **86–8** 93

Paixǎo J A, Lebech B, Gonçalves A P, Brown P J, Lander G H, Burlet P, Delapalme A and Spirlet J C 1997 *Phys. Rev.* B **55** 14 370

Pfannes H-D and Fisher H, 1977 *Appl. Phys.* **13** 317

Ptasiewicz-Bąk B, Baran A, Suski W and Leciejewicz J 1988 *J. Magn. Magn. Mater.* **76–77** 439

Rećko K, Biernacka M, Dobrzyński L, Perzynska K, Satula D, Waliszewski J, Suski W, Wochowski K, André G and Bourée F 1996 *26iémes Journées des Actinides, April 10–14, 1996 (Szklarska Poręba, Poland)* p 86

——1997a *Physica* B **234–236** 696

——1997b *J. Phys.: Condens. Matter* **9** 9541

Rodriguez-Carvajal J 1993 *FULLPROF* version 2.4.2 Dec 93–ILLJRC

Sayetat F, Fertey P and Kessler M 1998 *J. Appl. Cryst.* **31** 121

Steiner W, Reissner M, Bogner J, Sassik H and Sepiol B 1996 *Hyp. Int.* **1** 372

Suski W 1990 *J. Magn. Magn. Mater.* **90–91** 99

Szymański K 1998a *Nucl. Instrum. Methods* B **134** 405

——1998b *Nucl. Instrum. Methods* B **143** 601

Szymański K, Dobrzyński L, Prus B and Cooper M J 1996 *Nucl. Instrum. Methods* B **119** 438

Thacher A C J 1960 *Chem. Phys.* **32** 638