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High field magnetization of $UAl_{12-x}Fe_x$ ($4.0 \leqslant x \leqslant 4.8$) intermetallic compounds

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Abstract

The magnetization of $UAl_{12-x}Fe_x$ intermetallic compounds ($4.0 \leqslant x \leqslant 4.8$) at 4.2 K in magnetic fields up to 35 T has been studied on powder particles free to be oriented by the applied field and on particles fixed by frozen alcohol. The magnetic anisotropy and possible magnetic structures are discussed.

1. Introduction

The uranium compound UAl_8Fe_4 has the tetragonal crystal structure of the $ThMn_{12}$ type which belongs to the *I4/mmm* space group. The thorium site 2a is occupied by uranium and the three manganese sites 8f, 8i and 8j, four per formula unit, can be occupied by iron as well as by aluminium with preference for the f site by iron (Fig. 1). UAl_8Fe_4 and solid solutions based on this compound have been investigated in recent years by means of magnetization measurements, Mössbauer spectroscopy and neutron diffraction techniques (see ref. 1 and references cited therein). However, serious discrepancies exist in the data. In particular, for UAl_8Fe_4, there are three contradictory results of neutron diffraction studies.

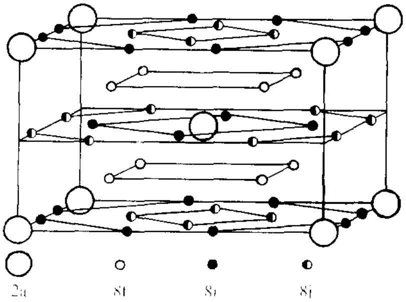

Fig. 1. Unit cell of the $ThMn_{12}$-type of structure.

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(1) The atomic arrangement is considered on the basis of perfect occupation of the 8f sites by iron (in this case, the structure is equivalent to the $CeMn_4Al_8$-type ordered modification of the $ThMn_{12}$ structure). Uranium is found to have no magnetic moment, whereas the iron magnetic moments on the 8f sites are equal to 1.6 μ_B and oriented along the *c* axis in a complex antiferromagnetic arrangement [2].

(2) The atomic structure is of the $CeMn_4Al_8$ type. The uranium magnetic moments ($\mu_U = 1.0\ \mu_B$) are ordered ferromagnetically and oriented along the *c* axis. The iron magnetic moments ($\mu_{Fe} = 1.0\ \mu_B$) are oriented along the *c* axis and form a simple antiferromagnetic structure [1, 3].

(3) There is spin-glass behaviour of both the uranium and the iron sublattices. The iron spins are randomly frozen within the basal plane perpendicular to the tetragonal *c* direction. No indication exists of long-range order of the uranium moments, but application of an external field causes alignment of the randomly oriented 5f moments parallel to the direction of the field [4]. The possibility that iron atoms are present on the 8j sites is assumed to be one of the reasons for the observed spin-glass behaviour.

We have prepared a series of samples in the concentration range ($4.0 \leqslant x \leqslant 4.8$) in order to check the main characteristics and to study the high field behaviour to obtain information about the magnetocrystalline anisotropy.

2. Experimental details

The samples have been prepared in an arc furnace as 5 g ingots from the pure constituents uranium (99.8% purity), aluminium (99.99% purity) and iron (99.99% purity). They were annealed for 1 week at 800 °C. By X-ray analysis the compounds were checked to be single phase. The lattice parameters are in good agreement with data in the literature (Fig. 2 and Table 1). A small amount of an undetermined second phase is observed only for $x = 4.8$. This could be the reason that the value of the lattice parameter

TABLE 1.

Lattice parameters *a* and *c*, unit-cell volumes *V*, spontaneous magnetic moments μ_m per formula unit at 4.2 K and the ratios of the zero-field extrapolated magnetizations of free and fixed powder for $UAl_{12-x}Fe_x$ intermetallic compounds.

x	a (pm)	c (pm)	V (nm^3)	μ_m (μ_B (formula unit)$^{-1}$)	μ_{fixed}/μ_{free}
4.0	874.5	504.1	0.3855	1.4	0.79
4.2	873.0	503.9	0.3840	3.7	0.76
4.4	872.6	503.1	0.3831	6.0	0.86
4.6	870.3	502.1	0.3803	6.7	0.90
4.8	867.0	501.7	0.3770	7.2	0.88

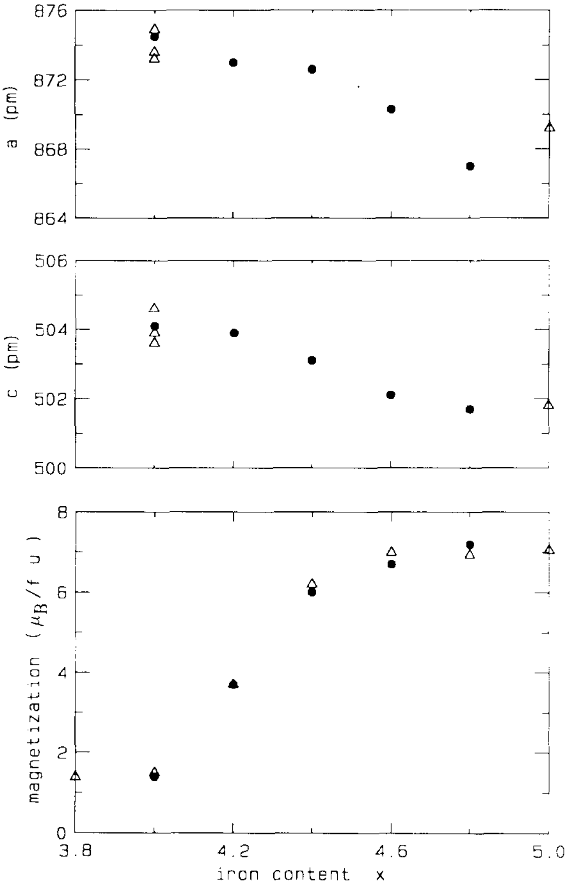


Fig. 2. Concentration dependences of the lattice parameters and the spontaneous magnetic moment (per formula unit): (●, present work; △, from refs. 1–3 and 6).

a is slightly low (Fig. 2). Magnetization measurements were carried out at 4.2 K in fields up to 35 T in the Amsterdam High Field Installation. The samples were measured as powder free to rotate in the applied field and as powder with random orientation fixed by frozen alcohol.

3. Results and discussion

Figure 3 shows the magnetization curves measured on free-powder samples. The spontaneous magnetic moments of the free powders, determined by extrapolation of the magnetization to zero field, are in good agreement with the data in ref. 5 (Fig. 2). To ensure that domain-wall motion processes are completed, extrapolations were made from the field interval 3–10 T. The

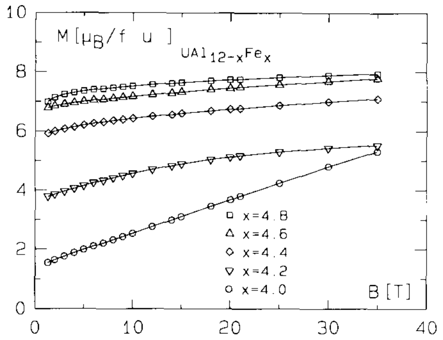
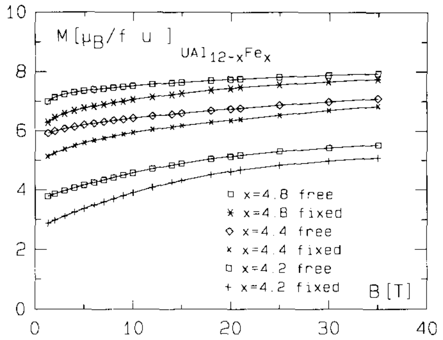


Fig. 3. Magnetization curves of powders free to rotate in the applied field (fu, formula unit).



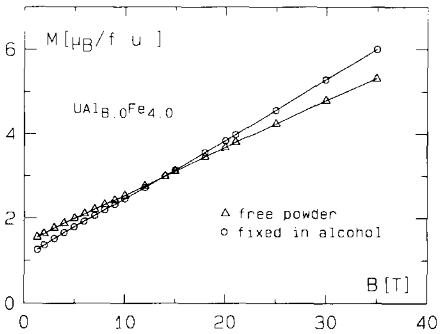


Fig. 4. Comparison between the magnetization curves of free and fixed powders for $x=4.2$, 4.4 and 4.8 (fu, formula unit).

Fig. 5. Comparison between the magnetization curves of free and fixed powders for $x=4.0$ (fu, formula unit).

difference between the preparation of our samples and those used in ref. 5, where induction melting and an annealing temperature of 700 °C were used, appears to have no influence on the structural and magnetic properties.

In Figs. 4 and 5, the magnetization curves of the fixed powders are compared with those of the free powders. In all samples a high magnetocrystalline anisotropy is observed. The ratios $\mu_{\text{fixed}}/\mu_{\text{free}}$ of the spontaneous magnetizations of the fixed powder to the free powder are presented in Table 1. Theoretically, for a highly anisotropic magnet this ratio is equal to 0.5 in the case of uniaxial anisotropy and to 0.785 for basal plane type of anisotropy. Our result for $x=4.0$ corresponds to the basal plane character of the anisotropy, in contradiction with the uniaxial arrangement of the magnetic moments proposed for this composition in refs. 2 and 3. For the compounds with higher iron contents, in which iron atoms occupy not only the 8f sites but also the 8j and 8i sites, the ratio shows a tendency to increase, indicating a more isotropic character of the anisotropy.

The observed results may be explained as follows. At $x=4.0$, the magnetic order of the iron moments on the 8f sublattice is antiferromagnetic, but the occupation of the 8f sites may be not perfect, as proposed in refs. 3 and 6. If some iron atoms are absent on 8f sites, as has been assumed in ref. 4, the magnetic moments on the 8f sites are not completely compensated and possess spontaneous magnetic moments. According to ref. 4, these moments are directed perpendicular to the *c* axis and therefore may be responsible for the basal-plane type of magnetocrystalline anisotropy. The iron atoms on the 8j sites have slight uniaxial anisotropy and for x close to 4 the basal plane anisotropy contribution from the 8f sublattice dominates. With increasing x and, consequently, with increasing number of iron atoms on the 8j sites, the uniaxial contribution to the anisotropy becomes larger. This leads to a decrease in the total anisotropy, which is reflected in an increase in the $\mu_{\text{fixed}}/\mu_{\text{free}}$ ratio. As is known from measurements on $UFe_{10}Si_2$ [1, 7] and also on compounds without uranium, having the same structure and having a high iron concentration (*e.g.* $LuFe_{11}Ti$ [8]), the iron sublattice has uniaxial anisotropy. Therefore we can suppose a concentration-dependent spin reorientation to occur in the iron sublattice, proceeding from basal plane to uniaxial via an almost isotropic competitive mechanism. The latter arises because, with increasing iron content, filling of not only 8f but also of 8j and 8i sites takes place. Apparently, in particular iron atoms on 8j and 8i sites are responsible for the relatively strong uniaxial anisotropy in the compounds with high iron content.

In a magnetic field, the antiferromagnetic spin structure of the iron atoms on the 8f sublattice undergoes a spin-flip transition, which is reflected in the large slope of the $\mu(B)$ curve for $x=4.0$. A higher iron content leads to stronger ferromagnetic exchange owing to an increase in the number of iron atoms on the 8j and 8i sites and the antiferromagnetic coupling of the Fe atoms becomes suppressed. This is reflected in the large increase in the spontaneous moment between $x=4.0$ and $x=4.4$ and the decrease in the differential high field susceptibility. At $x \geqslant 4.6$, only ferromagnetic coupling remains.

In this explanation of the observed results, a non-magnetic ground state of uranium is likely, because the arrangement of the uranium atoms along the *c* axis, as proposed in ref. 3, should lead to strong uniaxial anisotropy. However, the spin-glass arrangement of the uranium magnetic moments reported in ref. 4 also does not contradict the above picture, if we assume that the alignment of the uranium atoms in low fields is not very strong and that the value of the low field magnetization is mainly determined by the iron sublattice. The spin-glass arrangement of the iron atoms in the basal plane [4] is in agreement with our explanation, if a small ferromagnetic component is assumed to be present in the spin glass.

The crossing of the magnetization curves of the free and fixed powders for UAl_8Fe_4 (Fig. 5) indicates the complex character of the magnetic arrangement and should be the subject of a separate investigation, preferably on a single crystal.

Acknowledgments

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References

1 W. Suski, *J. Magn. Magn. Mater.*, *90–91* (1990) 99.

2 H. Ptasiewicz-Bak, A. Baran, W. Suski and J. Leciejewicz, *J. Magn. Magn. Mater.*, *76–77* (1988) 439.

3 W. Schäfer, G. Will, J. Gal and W. Suski, *J. Less-Common Met.*, *149* (1989) 237.

4 J. Gal, I. Yaar, D. Regev, S. Fredo, G. Shani, E. Arbaboff, W. Potzel, K. Aggarwal, J. A. Pereda, G. M. Kalvius, F. J. Litterst, W. Schäfer and G. Will, *Phys. Rev. B*, *42* (1990) 8507.

5 A. Baran, W. Suski, O. J. Zogal and T. Mydlarz, *J. Less-Common Met.*, *121* (1986) 175.

6 J. Stepien-Damm, A. Baran and W. Suski, *J. Less-Common Met.*, *102* (1984) L5.

7 A. V. Andreev, S. V. Andreev and E. N. Tarasov, *J. Less-Common Met.*, *167* (1991) 255.

8 A. V. Andreev, V. Sechovsky, N. V. Kudrevatykh, S. S. Sigaev and E. N. Tarasov, *J. Less-Common Met.*, *144* (1988) L21.