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Change of anisotropy type in $UFe_{10-x}Al_xSi_2$

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Abstract

The homogeneity range in the $UFe_{10-x}Al_xSi_2$ solid solutions is found to be up to $x=3.0$. The magnetic moment and Curie temperature decrease linearly with increasing aluminium content whereas the uniaxial magnetocrystalline anisotropy is weakened, and the compounds with $x>2$ have a basal plane type of anisotropy. The total anisotropy of the compounds has moderate magnitude and is formed by large uniaxial contributions from the uranium and the Fe 8(i) sublattices and by a large basal plane contribution from the Fe 8(f) sublattice.

1. Introduction

The $UFe_{10}Si_2$ intermetallic compound is an actinide representative of a new class of magnetic materials $RFe_{12-x}M_x$ (R ≡ rare earth metal, Y; M ≡ Si, Ti, V, Cr, Mo, W, Re) with high iron content. The compound has uniaxial magnetocrystalline anisotropy, sufficiently high values of the spontaneous magnetization and the Curie temperature, and can be considered as a possible material for permanent magnets [1–4]. The Curie temperature, being equal to 653 K, is the largest for the known uranium-containing magnetics, and can even be increased to 750 K by partial substitution of cobalt for iron [5]. The tetragonal crystal structure of the compound belongs to the $ThMn_{12}$ type (space group *I4/mmm*). The lattice cell contains two formula units: the uranium atoms occupy one position of the 2(a) type; the iron and silicon atoms are distributed over the 8(f), 8(i) and 8(j) positions. It is known that the silicon atoms occupy 8(f) sites (3 atoms per unit cell) and 8(j) sites (1 atom per unit cell) [6, 7]. Thus, the 8(i) positions in $UFe_{10}Si_2$ are occupied practically by the iron atoms only. The compound has homogeneity range $1 \leqslant x \leqslant 3$ when using the $UFe_{12-x}Si_x$ notation [3].

Furthermore, the related ternary compound UFe_4Al_8 with moderate iron content forms the same crystal structure. It also has a homogeneity range, $6 \leqslant x \leqslant 9$ when using the $UFe_{12-x}Al_x$ notation, which does not reach as high an iron content as in the case of silicon. These solid solutions display a complicated magnetic behaviour caused by a competition between exchange interactions in different iron sublattices (see ref. 8 and references cited therein, as well as refs. 9 and 10).

The study of the change in magnetic properties following the transition between these two regions of stability of the $ThMn_{12}$ type of structure could give information on individual contributions of different iron sublattices to magnetic moment and magnetocrystalline anisotropy. The possibility of substituting aluminium for iron in the silicon-containing compounds was demonstrated in ref. 5 where preliminary results on UFe_9AlSi_2 have been reported. In the present work, we prepared the $UFe_{10-x}Al_xSi_2$ solid solutions, determined their homogeneity range and studied the magnetic properties.

2. Experimental details

The $UFe_{10-x}Al_xSi_2$ alloys were obtained by melting components (uranium 99.9% purity; iron, aluminium and silicon, 99.99%) in an arc furnace under an argon protective atmosphere and annealing at 900 °C for two weeks.

The powders with particle size less than 45 μm were mixed with 15% of silicon oil in the sample holders and aligned at room temperature in an electromagnet at a field of 1.2 T. The prepared samples were immediately cooled down to keep up the high degree of the alignment. The magnetization measurements were carried out at 4.2 K in magnetic fields up to 4 T parallel and perpendicular to the axis of alignment.

The Curie temperatures were determined by d.c. susceptibility measurements.

3. Results and discussion

The X-ray analysis revealed a single-phase state with the $ThMn_{12}$ type of structure for the compounds with $x<3$. The alloys with $x>3$ contain large amounts of at least two phases. The boundary compound, $UFe_7Al_3Si_2$, has traces (up to 5%) of a second phase and is considered here to belong to homogeneity range of aluminium solid solution in $UFe_{10}Si_2$. In Fig. 1, the concentration dependences of lattice parameters are presented. The increase in both parameters with increasing aluminium content reflects the larger size of aluminium atoms than iron atoms. Considerable non-linearity of the $a(x)$, $c(x)$ curves could be explained as a result of a preference in occupation of different positions by iron, aluminium and silicon atoms, which was observed by ^{57}Fe Mössbauer effect measurements (to be published in a separate paper). The lattice expansion is isotropic; the c/a ratio is nearly constant (Fig. 1).

Figure 2 shows the concentration dependences of the molecular magnetic moment M measured along the alignment axis at 4.2 K in a 4 T field and of the Curie temperature T_C. As could be expected, the substitution of iron by non-magnetic aluminium leads to monotonic, nearly linear decreases in

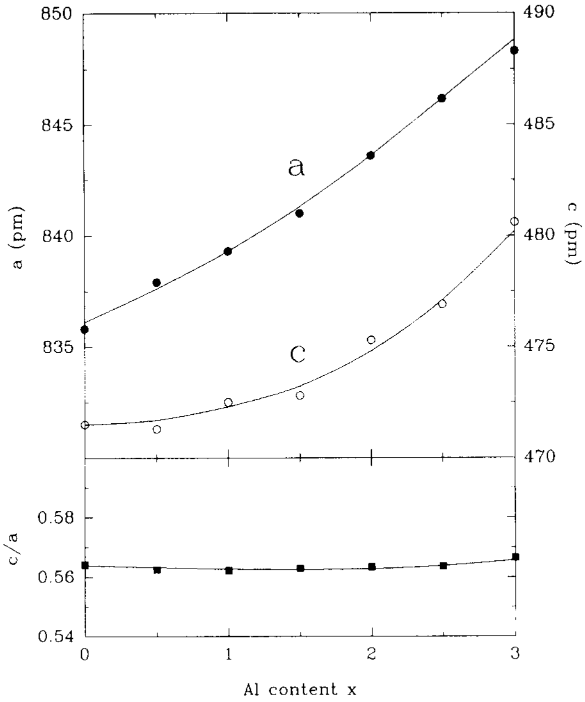

Fig. 1. Concentration dependences of the lattice parameters a, c and the c/a ratio.

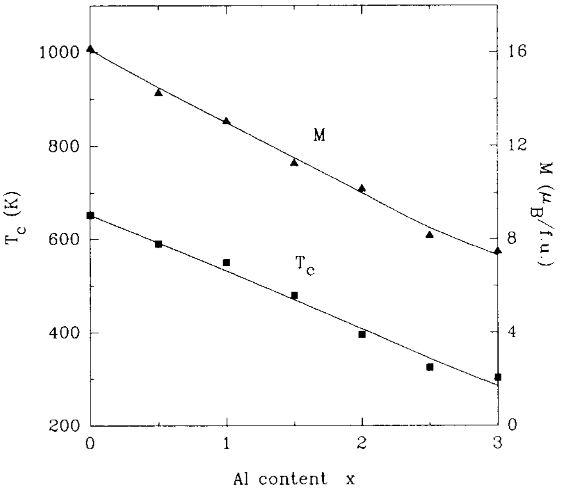

Fig. 2. Concentration dependences of the molecular magnetic moment M, measured along the alignment axis at 4.2 K in a field of 4 T, and of the Curie temperature T_C.

M and T_C; the values of both characteristics decreased by a factor of 2 for the aluminium-rich border of the homogeneity range in comparison with the initial compound.

The most interesting results concern the magnetocrystalline anisotropy. The field dependences of the molecular magnetic moment along and perpendicular to the axis of alignment at 4.2 K are presented in Fig. 3. The notation "e" in this figure means "easy" (parallel to the axis of alignment) and "h" means "hard" (perpendicular to the axis of alignment). One can see the large uniaxial anisotropy of $UFe_{10}Si_2$. The hard direction curve has a field-induced phase transition of the first-order magnetization process (FOMP) type beginning at 3.5 T. In ref. 4, where the magnetocrystalline anisotropy of $UFe_{10}Si_2$ was studied up to 6 T in comparison with $YFe_{10}Si_2$, the FOMP-type curve is attributed to the uranium sublattice and considered as evidence of uranium magnetic ordering. A saturation up to the easy direction moment was observed at 4.5 T.

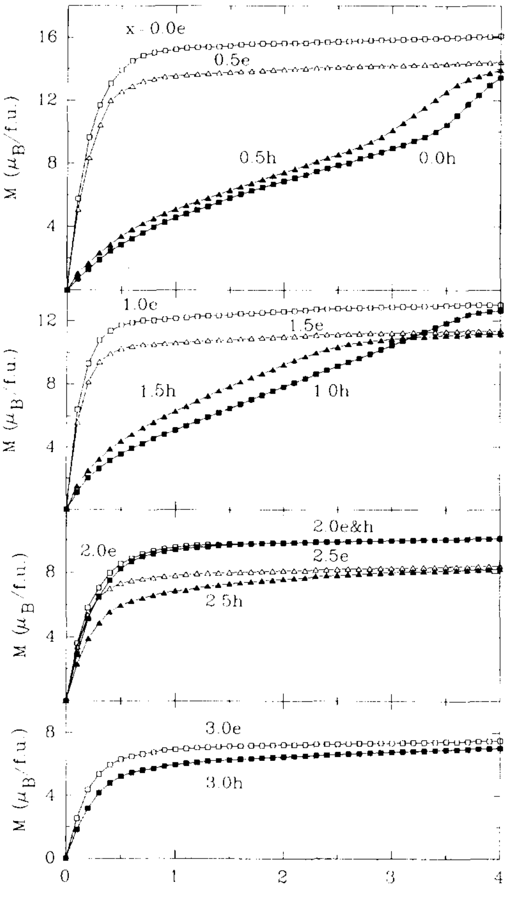

Fig. 3. Field dependences of the molecular magnetic moment of aligned powders parallel ("e", easy direction) and perpendicular ("h", hard direction) to the axis of alignment at 4.2 K for different aluminium contents x.

The character of the magnetization process for the compound with $x=0.5$ is the same. The energy E_a of magnetocrystalline anisotropy, which can be estimated as the area between easy and hard direction magnetization curves, becomes lower with increasing aluminium content. For $x \geqslant 1.0$, the FOMP anomaly disappears. The compound $UFe_8Al_2Si_2$ is rather isotropic, and the anisotropy occurs again for $x \geqslant 2.5$. The compound with $x=2.5$ is practically saturated in the hard direction at 3 T, whereas for $x=3.0$ the anisotropy field is considerably higher. However, the hard direction magnetization displays a high projection on zero field for $x \geqslant 2.5$, which indicates multiaxial anisotropy for these compounds. A qualitatively similar behaviour of magnetization was observed at room temperature, too, where a comparison of the X-ray spectra of isotropic and aligned powders has been carried out. This confirmed the change in anisotropy type with aluminium content increasing and revealed basal plane anisotropy for $x \geqslant 2.5$.

The different types of contributions to the magnetocrystalline anisotropy from the iron atoms located on different non-equivalent positions in the iron-containing compounds with the $ThMn_{12}$ structure were discussed in ref. 10 on the base of high field magnetization in UFe_xAl_{12-x} ($4 \leqslant x \leqslant 4.8$). It was concluded that the iron atoms on the 8(f) sites give a basal plane contribution to total anisotropy whereas 8(i) and 8(j) sublattices give uniaxial contributions. A possibility of change in the anisotropy type was predicted in a system where consequent substitution of iron atoms on different sites takes place in the concentration range from $x=5$ to $x=10$. In the investigated system, the homogeneity range is narrower; however, such a concentration spin reorientation has been observed.

Preliminary results of an ^{57}Fe Mössbauer effect study show that aluminium substitutes iron mainly in 8(i) sites [11]. In the initial compound these positions are completely filled by iron, and replacement of half of them ($x=2$) leads to loss of the uniaxial anisotropy. The basal plane contribution from the Fe 8(f) sublattice to the total magnetocrystalline anisotropy is large enough to compensate the uniaxial contribution not only from the semifilled Fe 8(i) sites but also from the uranium sublattice. The energy E_a of magnetocrystalline anisotropy can be estimated from the area between easy and hard direction magnetization curves. For these moderately anisotropic compounds, the error of such an estimation is less than 20% in comparison with the single-crystal measurements. In $UFe_{10}Si_2$, $E_a = 2.2$ MJ m^{-3} [4]. The FOMP-type transition indicates a negative contribution to E_a from the high-order anisotropy constants. The extrapolation of the hard direction curve from the 1–3 T field interval to fields higher than the transition gives the first anisotropy constant K_1 to be equal to 2.7 MJ m^{-3}. Let us write expressions for K_1 of the compound as follows:

$$K_1 = K_{1U} + K_{1i} + K_{1j} + K_{1f} = 2.7 \text{ MJ m}^{-3}, \quad \text{for } x=0 \tag{1}$$

$$K_1 = K_{1U} + 0.5K_{1i} + K_{1j} + K_{1f} = 0, \quad \text{for } x=2 \tag{2}$$

where K_{1U} is the uranium sublattice contribution, and other terms are the contributions from 8(i), 8(j) and 8(f) iron sublattices of $UFe_{10}Si_2$ ($x=0$).

The first term, K_{1U}, can be estimated as the difference between K_1 values for $UFe_{10}Si_2$ and $YFe_{10}Si_2$ (1.4 MJ m^{-3} [4]), being equal to 1.3 MJ m^{-3}. The solution of eqns. (1) and (2) gives $K_{1i}=5.4$ MJ m^{-3} and $K_{1j}+K_{1f}=-4.0$ MJ m^{-3}. We cannot separate the K_{1j} and K_{1f} contributions; it will be done after careful analysis of the Mössbauer effect results. However, as a first approximation, we can neglect the contribution of the Fe 8(j) sublattice, because the Fe 8(f) sublattice is concluded to be responsible for basal plane anisotropy and, consequently, for a negative contribution to K_1 [9, 10]. If the Fe 8(j) sublattice has some uniaxial anisotropy, the absolute value of (negative) K_{1f} would be even larger than the value 4 MJ m^{-3} obtained above.

Of course, such a speculation is very approximate. We did not take into account at least two factors which could be important: first, a possible change in the ground state of uranium, and, second, the size factor reflected in Fig. 1. Anyway, the conclusion about the moderate size of the magnetocrystalline anisotropy of the whole iron sublattice as a result of the cancellation of large positive and negative contributions to K_1 seems to be right. It might hold not only for the compounds studied in the present paper, but also for all iron-containing compounds with the $ThMn_{12}$ type of structure. For example, our preliminary results of magnetization measurements of $LuFe_{11}Si_2$ give a surprisingly low $K_1 \leqslant 0.7$ MJ m^{-3} in comparison with 1.4 MJ m^{-3} for $YFe_{10}Si_2$ [4] and 1.9 MJ m^{-3} for $LuFe_{11}Ti$ [12]. Usually, in known R–3d intermetallic compounds with the same structure, the same 3d metal content and non-magnetic R, the values of K_1 vary slightly. In the considered compounds, the slight variation in distribution of the iron atoms over non-equivalent positions can slightly influence the uniaxial and basal plane contributions; however, their difference could be changed relatively strongly. Therefore, in the $ThMn_{12}$-type compounds with magnetic 3d metals one should not use the total 3d sublattice description but always consider three individual sublattices.

4. Conclusion

The homogeneity range in the $UFe_{10-x}Al_xSi_2$ solid solutions is found to extend up to $x=3.0$, and the magnetic properties within this range were studied. The magnetic moment and Curie temperature decrease linearly with increasing aluminium content. The uniaxial magnetocrystalline anisotropy, characterized by the first constant $K_1=2.7$ MJ m^{-3} in $UFe_{10}Si_2$, is sharply weakened with the Fe–Al substitution, the compound $UFe_8Al_2Si_2$ is magnetically isotropic, and the compounds with $x>2$ have a basal plane type of anisotropy. The total anisotropy of the compounds has moderate magnitude and is formed by large uniaxial contributions from the uranium and the Fe 8(i) sublattices (for $x=0$, they are approximately 1.3 MJ m^{-3} and 5.4 MJ m^{-3} respectively) and by a large basal plane contribution from the Fe 8(f) sublattice (-4 MJ

m^{-3}). The contribution from the Fe 8(j) sublattice is still open to discussion. The results obtained for the system investigated here can be useful for consideration of all 3d-metal-containing compounds with the $ThMn_{12}$ type of structure.

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