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Magnetic anisotropy and magnetostriction of $UFe_{10}Si_2$

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Abstract

The magnetic anisotropy and magnetostriction of $UFe_{10}Si_2$ were studied on aligned polycrystals. At 4.2 K the magnetic anisotropy is described by $K_1 = 3.0$ MJ m^{-3} and $K_2 = -0.9$ MJ m^{-3}. The magnetostriction constants obtained from the magnetostriction and thermal expansion measurements are $\lambda_1^{\alpha,0} = 2.14 \times 10^{-3}$, $\lambda_2^{\alpha,0} = 6.07 \times 10^{-3}$, $\lambda_1^{\alpha,2} = -0.4 \times 10^{-4}$, $\lambda_2^{\alpha,2} = 0.9 \times 10^{-4}$ and $\lambda^{\gamma,2} = 1.8 \times 10^{-4}$.

Keywords: Uranium intermetallics; Hard magnetic materials; Magnetic anisotropy; Magnetostriction

1. Introduction

$UFe_{10}Si_2$ is an actinide representative of the $RFe_{12-x}M_x$ class of magnetic materials. It has a b.c.t. lattice of the $ThMn_{12}$ type and the space group is *I4/mmm*. The unit cell contains two formula units (Fig. 1). The U atoms occupy 2*a* sites. The 8*i* sites are occupied by Fe atoms only, while the 8*j* and 8*f* sites are occupied by both Fe and Si atoms (7Fe + 1Si on 8*j*, 5Fe + 3Si on 8*f*) [1–3]. The $UFe_{12-x}Si_x$ system has a homogeneity range from $x = 1$ to 3, which is much wider than that of rare earth analogues ($1.8 < x < 2.2$ in the case of R ≡ Y). The molecular magnetic moment μ_m and the Curie temperature T_c depend on x non-monotonically, with maxima at $x = 2$ [4].

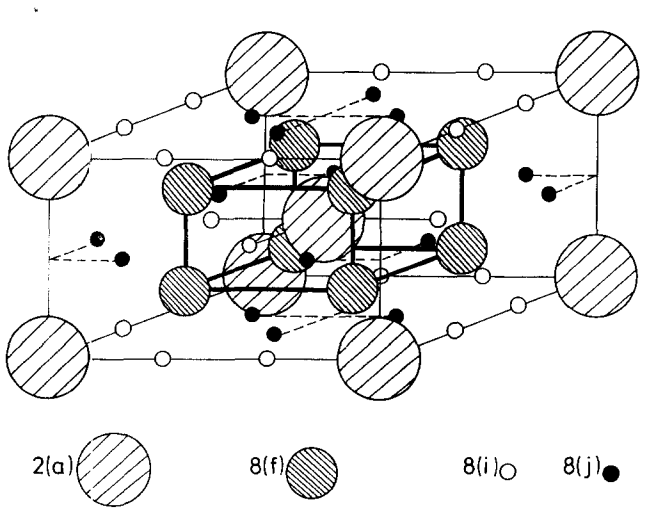

Fig. 1. Unit cell of $ThMn_{12}$-type structure.

In a few known U compounds with a high content of 3d metal ($UCo_{5.3}$ and $R_{1-x}U_xCo_5$) the U atoms do not carry a magnetic moment. The magnetic moment μ_d of the 3d atom, T_c and the magnetic anisotropy are considerably smaller than for RCo_5 with non-magnetic R (La, Y) owing to the filling of the 3d band by additional electrons from U [5–7]. One might also expect $UFe_{10}Si_2$ to be a weak analogue of the related compounds $YFe_{10}Si_2$ and $LuFe_{10}Si_2$. Indeed, μ_{Fe} is reduced to 1.6 μ_B in $UFe_{10}Si_2$ from 1.8 μ_B in $YFe_{10}Si_2$ [3]. However, the much higher T_c of $UFe_{10}Si_2$ in comparison with $YFe_{10}Si_2$ and even with $GdFe_{10}Si_2$, the large U contribution to the magnetic anisotropy and the field-induced phase transition in the hard direction all indicate a magnetic state of U in $UFe_{10}Si_2$.

The magnetic anisotropy of the compound has so far been studied only at 4.2 K. Here the temperature dependence of the magnetic anisotropy and preliminary results of magnetostriction measurements are presented.

2. Experimental details

The $UFe_{10}Si_2$ alloy was prepared by melting the components (uranium purity, 99.9%; iron and silicon, 99.99%) in an arc furnace on a water-cooled copper bottom under a protective argon atmosphere. The ingots were turned several times to avoid inhomogeneities and were subsequently annealed at 900 °C for 1 week.

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Standard X-ray, metallographic and thermomagnetic analyses showed the single-phase state of the alloy. The ingot was crushed into a powder with a particle size of about 50 μm. The powder was mixed with stycast and the mixture solidified in a steady field of 2.4 T at room temperature. Since the compound exhibits uniaxial magnetic anisotropy, the c axes of the sample particles are well aligned along the applied magnetic field.

Magnetization data were obtained by an induction method in pulsed fields up to 15 T with a rise time of about 5 ms at temperatures from 4.2 to 320 K. The magnetostriction was measured by a capacitor method at 4.2 K in the same pulsed field installation.

3. Results and discussion

3.1. Magnetic anisotropy

Fig. 2 shows the magnetization curves perpendicular to the axis of alignment (i.e. along the basal plane) at various temperatures. At 4.2 K the curve is in a good agreement with previous results [8], but the transition at about 3.2 T is more pronounced in the present work owing to a better alignment of the powder particles by an alignment field twice the size of that used in Ref. [8]. The easy direction magnetization curves saturate in a field range of 0.3–1.0 T and coincide with the corresponding hard direction curves at 5–6 T. The temperature dependences of the molecular spontaneous magnetic moment M_s, the anisotropy field B_a, the first anisotropy constant K_1 and the transition field B_{tr} are presented in Fig. 3. M_s was determined by extrapolation of the easy direction magnetization curves to zero field. B_a was found by extrapolation of the initial part (in the field interval $1<B<2.5$ T) of the hard direction magnetization curve up to its crossing with the easy direction curve. B_{tr} was determined as the field of maximum differential susceptibility (see inset of Fig. 2). K_1 at 4.2 K is found to be 10% larger than in Ref. [8] owing to a better alignment of the sample. It should be mentioned that a possible contribution to the measured K_1 from a field-induced deformation of the magnetic structure can be neglected because of the absence of any volume effect during the magnetization process in the hard magnetic direction (see Section 3.2).

The field-induced transition in $UFe_{10}Si_2$ has several unusual features. First, it exists up to unexpectedly high temperatures of about $0.5T_c$. Second, the transition field is practically temperature independent. Third, it has no noticeable hysteresis (however, this is not too rare [9]). The transition observed belongs to the so-called FOMP-1 type (first-order magnetization process) [9,10] where the saturation magnetization is reached after transition. In an uniaxial ferromagnet with an FOMP-1 transition the hard direction magnetization curve can be described by two anisotropy constants, positive K_1 and negative K_2:

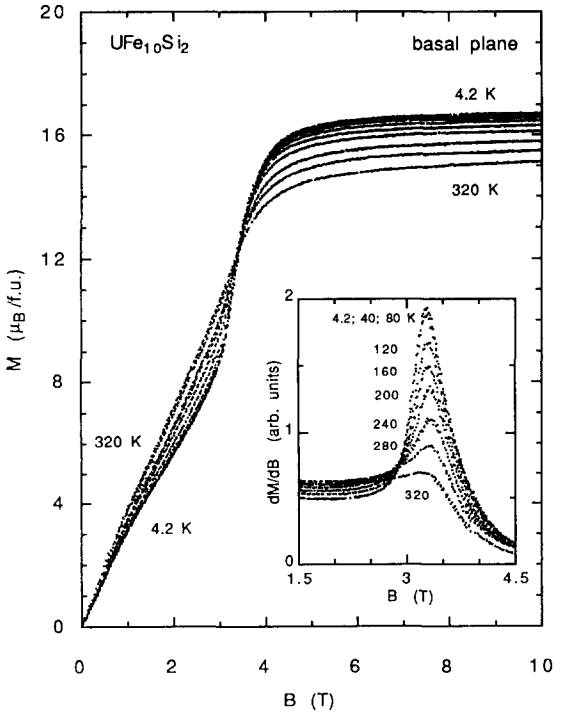

Fig. 2. Hard direction magnetization curves at various temperatures. Inset: dM/dB in range of field-induced transition.

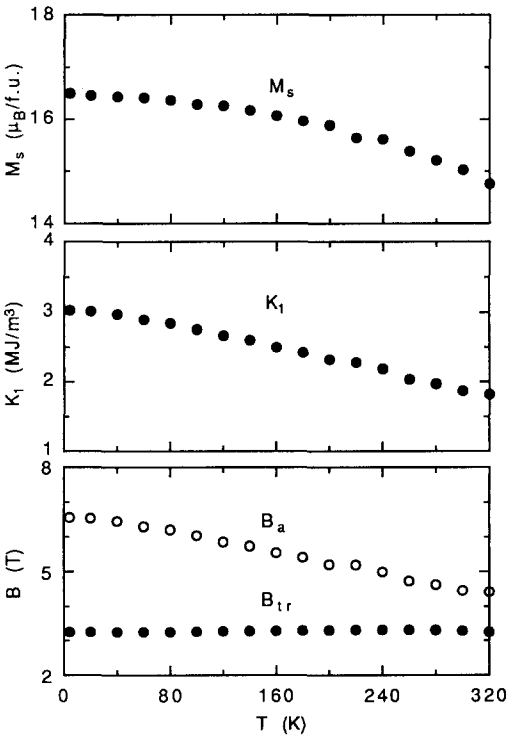

Fig. 3. Temperature dependences of spontaneous magnetic moment M_s, first anisotropy constant K_1, anisotropy field B_a and transition field B_{tr}.

$$\frac{B}{j} = \frac{2K_1}{M_s} + \frac{4j^2K_2}{M_s} + NM_s \quad (1)$$

where B is the external field, M_s is the spontaneous magnetization, j is the reduced magnetization M/M_s and N is the demagnetizing factor of the sample ($4\pi/3$ in the present case for a cube). In Fig. 4 examples of these fits are given for several temperatures. The higher initial slope of the experimental curves reflects an imperfect alignment of the powder. The K_1 values obtained from these fits are practically the same as presented in Fig. 3. The results for K_2 are shown in Fig. 5. M_s, B_a and K_1 decrease monotonically with increasing temperature as well as absolute value of K_2.

It is not possible to analyse the $K_1(T)$ dependence by a comparison with $M_s(T)$, because it is known that U nearly does not contribute to the total magnetic moment (about 3%) but gives more than half of K_1, as determined in Refs. [2,8,11] by a comparison of K_1 of $UFe_{10}Si_2$ with that of $RFe_{10}Si_2$ with non-magnetic R ≡ Y, Lu. However, the decrease in K_1 with increasing temperature is at least not sharper than in the isostructural compounds $YFe_{11}Ti$ and $LuFe_{11}Ti$ with non-magnetic rare earth atoms, for which single-crystal data on the magnetic anisotropy are available [12,13]. The ratio $K_1(0.5T_c)/K_1(0)$ is equal to 0.6 in $UFe_{10}Si_2$ but only 0.5 in $YFe_{11}Ti$ and $LuFe_{11}Ti$. In the case of R–3d metal intermetallics with magnetic R having the same

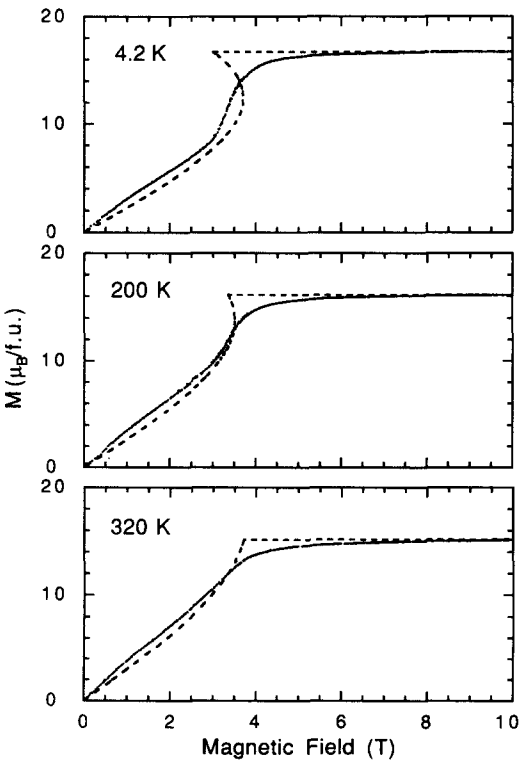

Fig. 4. Hard direction magnetization curves and their fits (dashed lines) by formula (1). The fits correspond to the case of a single crystal.

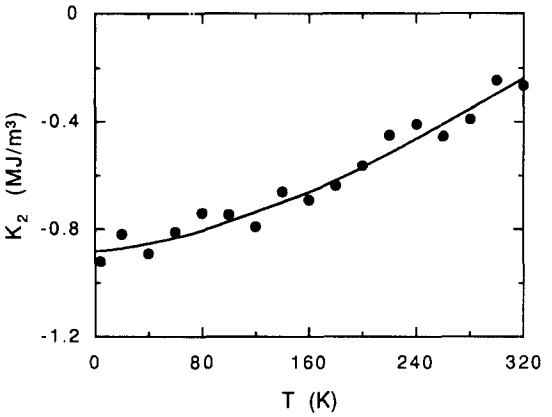

Fig. 5. Temperature dependence of second anisotropy constant K_2. The line is merely a guide for the eye.

sign of contribution to the magnetic anisotropy as the 3d metal, this ratio is always lower than for Y or Lu, because the magnetic moment of R decreases with increasing temperature much faster than that of the 3d metal. For example, $K_1(0.5T_c)/K_1(0) = 0.4$ in $SmFe_{11}Ti$ [14] and less than 0.1 in $HoFe_{11}Ti$ [12]. This shows that the U contribution to the magnetic anisotropy, and consequently the U magnetic moment, decreases with increasing temperature surprisingly slowly.

We can roughly estimate the temperature dependence of the U magnetic moment from the temperature dependence of K_2, because the Fe sublattice does not give a noticeable contribution to this constant. In such a case, within the localized single-ion model, $K_2(T)$ is proportional to the tenth power of the f-metal magnetic moment $\mu_f(T)$ [15]. This leads to $\mu_U(320\ K)/\mu_U(4.2\ K) = 0.88$, the same ratio as for the molecular magnetic moment which is almost completely determined by the Fe sublattice. This speculation is not correct, since the U magnetism in $UFe_{10}Si_2$ cannot be described within the localized model, because neither the sign nor the magnitude of the U contribution to the magnetic anisotropy corresponds to a prediction of this model [11,16]. However, it shows qualitatively that the $\mu_U(T)$ decrease is not drastic, in agreement with the slow temperature dependence of K_1.

3.2. Magnetostriction

The field dependences of the longitudinal (λ_{aa}) and the transverse (λ_{ca} and λ_{ba}) magnetostriction at 4.2 K are shown in Fig. 6. The magnetic field is applied perpendicular to the c axis for all curves. The transverse magnetostriction is measured along (λ_{ca}) and perpendicular to (λ_{ba}) the c axis (the first index denotes the strain measurement direction, the second corresponds to the field direction). The curves display a pronounced correlation with the magnetization curves. Using the formula for the magnetostriction of uniaxial crystals from Ref. [17], the values after saturation (at 5 T) can

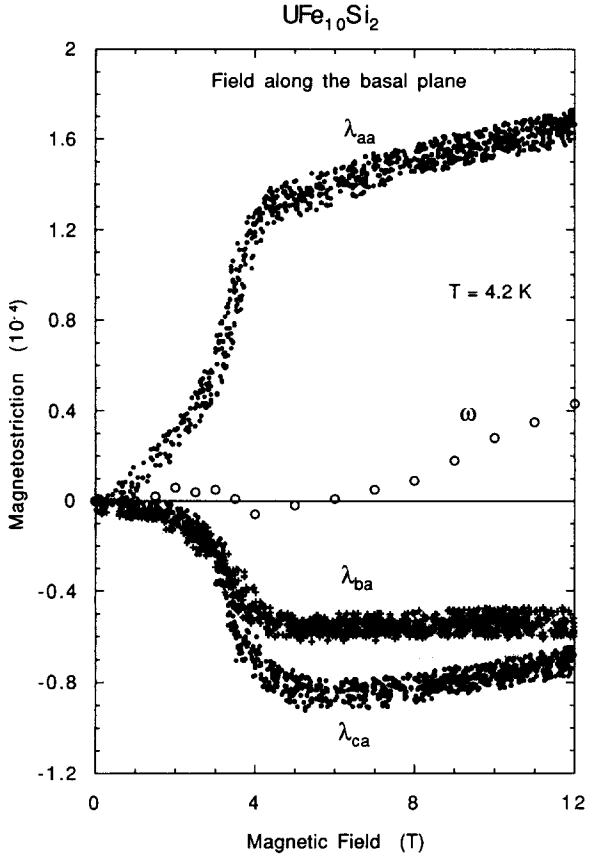

Fig. 6. Field dependences of longitudinal (λ_{aa}) and transverse (λ_{ca} and λ_{ba}) magnetostriction at 4.2 K. The magnetic field is applied perpendicular to the c axis for all curves. The transverse magnetostriction is measured along (λ_{ca}) and perpendicular to (λ_{ba}) the c axis. $\omega = \lambda_{aa} + \lambda_{ba} + \lambda_{ca}$ is the volume magnetostriction.

be presented as

$$\lambda_{aa} = -\lambda_1^{\alpha,2} + \tfrac{1}{2}\lambda^{\gamma,2}$$

$$\lambda_{ba} = -\lambda_1^{\alpha,2} - \tfrac{1}{2}\lambda^{\gamma,2}$$

$$\lambda_{ca} = \lambda_2^{\alpha,2} \qquad (2)$$

The anisotropic magnetostriction constants at 4.2 K are found from (2) to be $\lambda_1^{\alpha,2} = -0.4\times10^{-4}$ (if the uniaxial crystal structure is approximated by a cylinder this constant describes a change of the cylinder radius upon rotation of the magnetic moment from the basal plane to the c axis), $\lambda_2^{\alpha,2} = 0.9\times10^{-4}$ (this constant denotes the change in the cylinder height upon the same rotation of the magnetic moment) and $\lambda^{\gamma,2} = 1.8\times10^{-4}$ (this constant describes an orthorhombic distortion of the lattice when the magnetic moment lies in the basal plane).

The spontaneous magnetostriction of $UFe_{10}Si_2$ was studied in Ref. [4] by X-ray dilatometry. The magnetic ordering is accompanied, as in other high Fe content intermetallics, by a large volume effect ω_s which is equal to 10.2×10^{-3} at 5 K; ω_s is distributed anisotropically over the main axes. The linear strains in the basal plane (λ_a) and along the c axis (λ_c) are equal to 2.1×10^{-3} and 6.1×10^{-3} respectively. In a uniaxial ferromagnet these strains can be described by the magnetostriction constants as [17]

$$\lambda_a = \lambda_1^{\alpha,0} + \tfrac{2}{3}\lambda_1^{\alpha,2}$$

$$\lambda_c = \lambda_2^{\alpha,0} + \tfrac{2}{3}\lambda_2^{\alpha,2} \qquad (3)$$

The zero-order magnetostriction constants $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ describe the changes in the cylinder radius and height respectively in the magnetically ordered state compared with the paramagnetic state regardless of the direction of the magnetic moment. Therefore they are of exchange origin. From the thermal expansion data only the combination of the exchange and anisotropic magnetostriction constants, which are expressed by formulae (3), can be found.

Now, using the above results on the magnetostriction, the zero-order magnetostriction constants $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ are derived to be 2.14×10^{-3} and 6.07×10^{-3} respectively at 5 K. One can see that the anisotropic magnetostriction of $UFe_{10}Si_2$ is much weaker than the exchange magnetostriction. Ratios between constants which can be directly compared ($\lambda_1^{\alpha,2}/\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,2}/\lambda_2^{\alpha,0}$) do not reach 2% and the spontaneous strains λ_a and λ_c are practically equal to $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ respectively.

The volume effect in R–Fe intermetallics can be described, neglecting the weakest R–R interaction, by

$$\omega_s = n_{FeFe}\mu_{Fe}^2 + n_{RFe}\mu_R\mu_{Fe} \qquad (4)$$

where n_{FeFe} is the intrasublattice magnetoelastic coupling coefficient in the Fe sublattice and n_{RFe} is the intersublattice magnetoelastic coupling coefficient (in the present case R ≡ U). An attempt to separate the Fe–Fe and U–Fe exchange interaction contributions to ω_s is shown in Fig. 7. The second curve in this figure represents the Fe–Fe contribution calculated using $n_{FeFe} = 2.8\times10^{-3}\ \mu_B^{-2}$ from the results on the spontaneous magnetostriction of the isostructural $YFe_{11}Ti$ compound [18], $\mu_{Fe}(4.2\ K) = 1.59\ \mu_B$ in $UFe_{10}Si_2$ found from ^{57}Fe Mössbauer effect measurements of $U_{1-x}Y_xFe_{10}Si_2$ solid solutions [2] and assuming that μ_{Fe} depends on T/T_c in the same way as in $YFe_{11}Ti$, where $\mu_{Fe}(T/T_c)$ is known from single-crystal measurements [12]. The U–Fe interaction contribution is estimated to be very high, 30% of ω_s, which is much larger than in the case of rare earth metals in high Fe content intermetallics, where the analogous R–Fe contribution does not exceed about 10% of ω_s. This correlates generally with the higher Curie temperature of $UFe_{10}Si_2$.

As seen in Fig. 6, the field-induced transition is not accompanied by a volume effect due to a compensation of the linear strains. This shows that the transition is a simple rotation of the magnetic moment without a change in magnitude of the sublattice magnetic moments and their coupling. Indeed, any such change would influence the exchange interaction and consequently

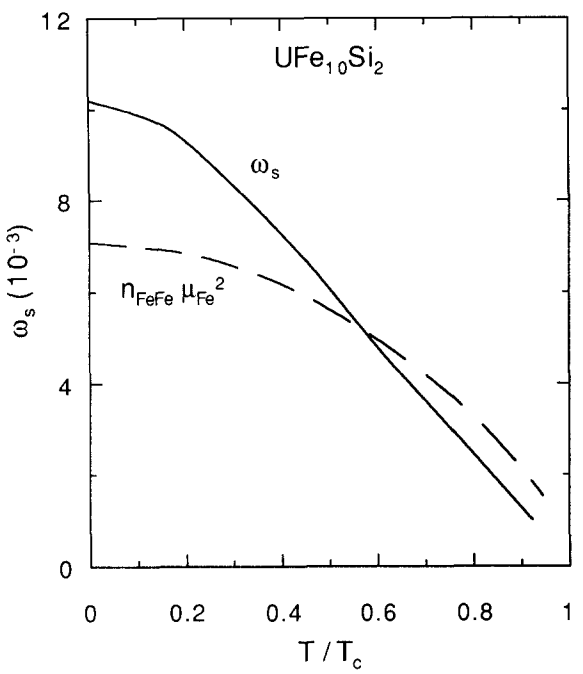

Fig. 7. Temperature dependences of spontaneous volume magnetostriction ω_s and calculated Fe–Fe interaction contribution to ω_s (see text).

the $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ magnetostriction constants. Since they are much larger than the anisotropic magnetostriction constants, even a small change in them will be comparable with $\lambda_1^{\alpha,2}$ and $\lambda_2^{\alpha,2}$ and produce a considerable (on the scale of Fig. 6) volume effect. The conclusion is valid for both Fe and U sublattices because of comparable contributions to ω_s from the Fe–Fe and U–Fe interactions.

4. Conclusions

The magnetic anisotropy and magnetostriction of $UFe_{10}Si_2$, the only uranium compound with a large 3d metal content where U contributes considerably to the magnetism up to high temperature, were studied on aligned polycrystals. At 4.2 K the magnetic anisotropy is described by $K_1 = 3.0$ MJ m^{-3} and $K_2 = -0.9$ MJ m^{-3}. The field-induced transition in the hard direction, which belongs to the FOMP-1 type, exists up to temperatures as high as $0.5T_c$ and the transition field is practically temperature independent.

The magnetostriction constants obtained from the magnetostriction and thermal expansion measurements are $\lambda_1^{\alpha,0} = 2.14 \times 10^{-3}$, $\lambda_2^{\alpha,0} = 6.07 \times 10^{-3}$, $\lambda_1^{\alpha,2} = -0.4 \times 10^{-4}$, $\lambda_2^{\alpha,2} = 0.9 \times 10^{-4}$ and $\lambda^{\gamma,2} = 1.8 \times 10^{-4}$.

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Note added in proof

When the paper was ready to be submitted, preliminary data on a single crystal of $UFe_{10}Si_2$ became available [19]. The hard direction magnetization curve at 4.2 K is in very good agreement with Figs. 4 and 5, but without the curvature in low field attributed to imperfect alignment. The fit in Fig. 5 with $K_1 = 3.0$ MJ m^{-3} and $K_2 = -0.9$ MJ m^{-3} actually describes the magnetization curve of a single crystal at 4.2 K very well.

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