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Evidence of uranium magnetic ordering in $UFe_{10}Si_2$

A. V. Andreev
Permanent Magnets Laboratory, Institute of Physics and Applied Mathematics, Ural State University, Ekaterinburg 620083 (Russian Federation)

W. Suski
Institute of Low Temperatures and Structural Research, P.O. Box 937, 50-950 Wrocław (Poland)

N. V. Baranov
Permanent Magnets Laboratory, Institute of Physics and Applied Mathematics, Ural State University, Ekaterinburg 620083 (Russian Federation)

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Abstract

The magnetocrystalline anisotropy of $UFe_{10}Si_2$ has been studied on aligned powder samples at 4.2 K. The enhancement of uniaxial anisotropy in comparison with that of $YFe_{10}Si_2$ and the field-induced phase transition in hard direction have been observed. The conclusion about a strong uranium contribution to the anisotropy energy has been deduced from comparison with the isostructural compounds $YFe_{10}Si_2$, $HoFe_{10}Si_2$ and $SmFe_{11}Ti$.

1. Introduction

The rare earth intermetallic compounds $RFe_{12-x}M_x$ (M ≡ Si, Ti, V, Cr, Mo, W, Re; $x = 1$–3) having a tetragonal crystal structure of the $ThMn_{12}$ type form a new class of magnetic materials with a very high iron content [1–5]. The iron subsystem in these compounds has a uniaxial magnetocrystalline anisotropy. This very rare property (the second case, the first being the $R_2Fe_{14}B$ compounds) immediately attracted great interest since some of the $RFe_{12-x}M_x$ compounds can be considered as possible materials for permanent magnets (namely $SmFe_{11}Ti$). On the contrary, these compounds all display the interesting features of the magnetic behaviour of the high iron content, rare earth intermetallics (in particular, a spontaneous spin reorientation and field-induced first-order phase transitions), having, at the same time, a relatively simple crystal structure with only one high symmetry position for R and three for iron and M. By comparison, the related compounds $R_2Fe_{14}B$ have two non-equivalent positions for R and six for iron. Therefore the $ThMn_{12}$-type compounds are favourable model subjects for the investigation of the intrinsic properties of iron-rich hard magnetic materials.

Two actinide representatives of this class, $UFe_{10}Si_2$ and $UFe_{10}Mo_2$, were found in 1988 [6]. The high Curie temperature and high magnetic moment

of $UFe_{10}Si_2$ made this compound more interesting, and several papers about its intrinsic magnetic properties, spontaneous magnetostriction, domain structure, coercivity and solid solutions based on it have recently appeared [6–8]. However, from these results it could not be concluded whether the uranium atoms have magnetic moments.

In this paper, the results of the magnetic anisotropy measurements on $UFe_{10}Si_2$ in comparison with those in $YFe_{10}Si_2$ are presented as a proof of the magnetic state of uranium.

2. Experimental details

The $UFe_{10}Si_2$ and $YFe_{10}Si_2$ alloys were obtained by melting the components (uranium and yttrium of 99.9% purity, and iron and silicon of 99.99% purity) in an arc furnace under a protective argon atmosphere. The X-ray analysis showed a single-phase state for the uranium compound. $YFe_{10}Si_2$ contained about 5–10% of extraneous α-Fe phase, which did not disappear during homogenization. This is in agreement with the findings in ref. 9, where it was concluded to be impossible to prepare single-phase samples of $YFe_{10}Si_2$ as well as of these silicides with rare earth metals. The values of the lattice parameters and Curie temperatures of both compounds are also in good agreement with previous results [6–9].

Powders with a particle size less than 4.5 μm were mixed with 15% silicone oil in the sample holders and aligned at room temperature in an electromagnet at a magnetic flux density of 1.2 T. The prepared samples were immediately cooled to maintain the high degree of the alignment. The magnetization measurements were carried out at 4.2 K in magnetic flux densities of up to 6 T parallel and perpendicular to the axis of alignment.

3. Results and discussion

The field dependences of the molecular magnetic moment along and perpendicular to the axis of alignment at 4.2 K for $UFe_{10}Si_2$ and $YFe_{10}Si_2$ are presented in Fig. 1. One can see that the easy-direction curves are saturated at approximately 4 T and the values of the molecular magnetic moment μ_m are equal to 16.4 μ_B and 18.0 μ_B respectively. This difference can be explained by at least three alternative reasons. Firstly, it could be due to a decrease in the iron magnetic moment due to the filling of 3d band by additional (in comparison with yttrium) electrons of uranium. In the rigid band model, this could lead to decrease in μ_m by 3 μ_B. Secondly, it could be caused by antiferromagnetic arrangement of the iron and uranium sublattices. The difference arising for only this reason could reach 3.3 μ_B as a maximum (U^{3+} free-ion configuration). Thirdly, extraneous iron could be present in the yttrium-containing sample. The difference could be up to 2.2 μ_B in the case when there is 10% α-Fe. We should also not forget about

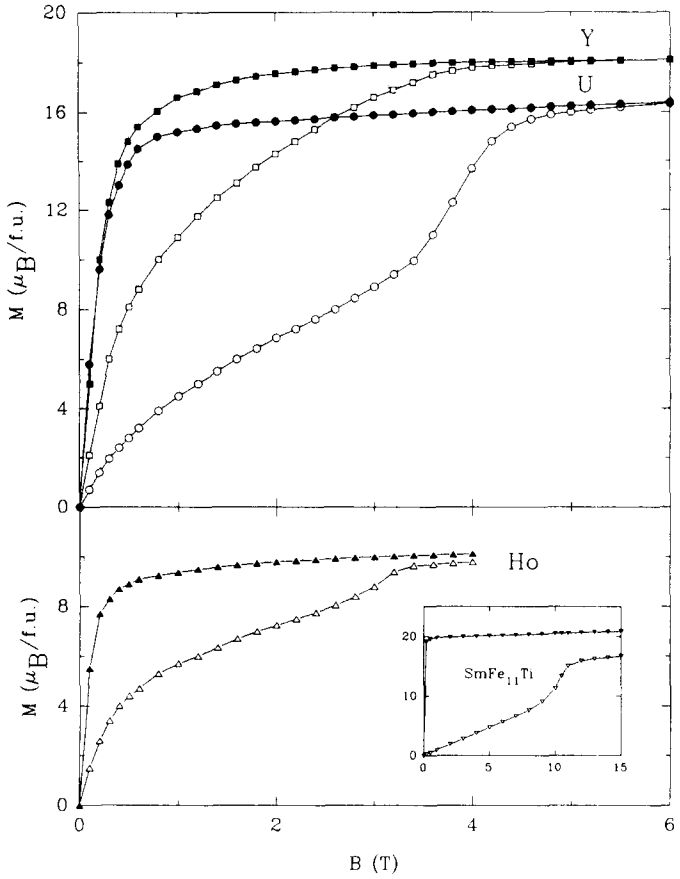

Fig. 1. Magnetization curves parallel (●, ■, ▲) and perpendicular (○, □, △) to the axis of alignment for aligned powder samples of $UFe_{10}Si_2$, $YFe_{10}Si_2$ (this work) and $HoFe_{10}Si_2$ [10]. The inset shows the magnetization curves along the c axis (▼) and in the basal plane (▽) for a single crystal of $SmFe_{11}Ti$ [13]. All curves were obtained at 4.2 K.

the difference in the lattice parameters (a and c for $YFe_{10}Si_2$ are larger than a and c for $UFe_{10}Si_2$ by 0.5%) and that there was some scatter in the data in the literature (18.3 μ_B for $YFe_{10}Si_2$ [9], 17.5 μ_B [7] and even 19 μ_B [6] for $UFe_{10}Si_2$; the last value refers to an α-Fe-containing sample). Thus this difference should not be considered as a base for some speculations about the state of uranium in $UFe_{10}Si_2$.

The hard-direction magnetization curves of uranium and yttrium compounds exhibit uniaxial magnetic anisotropy; however, they differ quantitatively and qualitatively. The magnetocrystalline anisotropy of $UFe_{10}Si_2$ is considerably larger, and there is a field-induced phase transition at around 4 T. The energy E_a of magnetocrystalline anisotropy can be estimated as the area between the 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 $YFe_{10}Si_2$, $E_a = 1.4$ MJ m^{-3} (1.3 MJ m^{-3} from the values of μ_m and anisotropy field H_a, presented in ref. 9). In $UFe_{10}Si_2$, $E_a = 2.2$ MJ m^{-3}. It is already larger by a factor of 1.5 than the value for $YFe_{10}Si_2$, but this value is only a minimum estimation, because the area is considerably decreased owing to the transition. The extrapolation of the hard-direction curve from the 1–3 T interval to higher magnetic flux densities than the transition value gives $E_a = 2.7$ MJ m^{-3}. The only reason for such a considerable difference in the anisotropy energy could be the uranium sublattice contribution and, consequently, the magnetic state of uranium.

The second argument for magnetic ordering of the uranium sublattice is the above-mentioned transition. Such a transition is rather usual for hard-direction magnetization curves in high iron content, rare earth intermetallics $RFe_{12-x}M_x$, $R_2Fe_{14}B$ and R_2Fe_{17} composed of magnetic R ions (the so-called first-order magnetization process (FOMP)). It is explained by taking into account the phenomenological anisotropy constants, K_1, K_2 and K_3 [10, 11]. In Fig. 1, the magnetization curves of an aligned powder of $HoFe_{10}Si_2$ [9] and of a single crystal of isostructural $SmFe_{11}Ti$ [12] are presented as examples of such a transition in the hard direction of uniaxial $RFe_{12-x}M_x$ compounds (strictly speaking, there is a slight canting in $HoFe_{10}Si_2$ [9]). The similarity between the behaviours of samarium, holmium and uranium compounds is evident as well as the difference from $YFe_{10}Si_2$ where yttrium is non-magnetic. In $UFe_{10}Si_2$, the transition is a FOMP 1 type. This means that magnetization after FOMP reaches saturation (as in $HoFe_{10}Si_2$) unlike FOMP 2 in $SmFe_{11}Ti$ without saturation after transition. The hysteresis of the transition is about 0.01 T.

The uranium sublattice contribution of about 1 MJ m^{-3} to the anisotropy energy and the presence of FOMP point to a non-zero orbital moment L of the uranium ion in $UFe_{10}Si_2$. On the contrary, the high Curie temperature $T_C = 653$ K, which is 100 K larger than in $YFe_{10}Si_2$ and even larger than the T_C of $GdFe_{10}Si_2$ (623 K [9]), indicates a strong U–Fe exchange interaction and, consequently, a noticeable spin moment S of uranium. The total moment $J = L - S$ can be rather small and invisible in the usual neutron diffraction measurements. The situation seems to be similar to that in UFe_2, where the compensation of noticeable L and S moments was proposed from indirect results (magnetization, magnetic anisotropy and magnetostriction [13, 14]) and proved by band structure calculation [15] and careful study of the uranium form factor by neutron diffraction [16].

The spin moments of 4f or 5f ions are always coupled antiferromagnetically with 3d metal spin moments. Thus, if S for uranium is larger than L, the U–Fe arrangement would be antiferromagnetic. In the case when $L > S$, a ferromagnetic U–Fe arrangement would occur. As L and S might be close to each other, we cannot predict the real arrangement. Ferromagnetism seems to be more probable, since light rare earth and actinide metals usually form ferromagnets with 3d metals, and the above-mentioned compound UFe_2 is ferromagnetic.

The additional indirect argument for magnetic state of uranium in $UFe_{10}Si_2$ is the temperature dependence of the spontaneous volume magnetostriction ω_s [7]. It does not scale with $\mu_m^2(T)$ as in the other R–Fe intermetallics with a non-magnetic R but decreases with increasing temperature much more sharply. The situation can be explained by the presence of not only the Fe–Fe but also the U–Fe exchange interaction.

4. Conclusion

The uranium sublattice contribution of about 1 MJ m^{-3} to the magnetocrystalline anisotropy energy and the presence of FOMP in the hard direction point to a considerable orbital moment L of uranium in $UFe_{10}Si_2$. On the contrary, the large enhancement of the Curie temperature in comparison with $YFe_{10}Si_2$ indicates a considerable spin moment S for uranium. Since the total moment J in the early transition elements is equal to L-S, the situation in $UFe_{10}Si_2$ can be similar to that in UFe_2, where the uranium ions due to cancellation of two large contributions have almost zero magnetic moment but strongly influence the intrinsic magnetic properties of the compound.

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