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Magnetic anisotropy of $UCo_{10}Si_2$

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Abstract

Magnetic properties of $UCo_{10}Si_2$ have been studied on aligned powders. Results are compared with properties of the isostructural compound $YCo_{10}Si_2$. The Curie temperature ($T_C = 550$ K) and the molecular magnetic moment ($\mu_m = 8.6\ \mu_B$) were found to be considerably lower in the U compound compared with $YCo_{10}Si_2$ (750 K and 11.0 μ_B respectively). However, $UCo_{10}Si_2$ has a strong uniaxial magnetic anisotropy (the first anisotropy constant $K_1 = 3.3$ MJ m^{-3} at 4.2 K), whereas $YCo_{10}Si_2$ has anisotropy of the cone type. If the difference in anisotropy is attributed to the U sublattice, this might point to a magnetic state of U in $UCo_{10}Si_2$ like in the Fe isostructural analogue $UFe_{10}Si_2$.

Keywords: Uranium intermetallics; Magnetic properties; Magnetocrystalline anisotropy

1. Introduction

$UCo_{10}Si_2$ belongs to a very large group of rare-earth and actinide $RT_{12-x}M_x$ ternary intermetallics with a high 3d-metal content that have been extensively studied in the last decade. The element T here is a late 3d transition metal. In these ternaries the third component M, which stabilizes the binary tetragonal crystal structure of the $ThMn_{12}$-type (*I4/mmm* space group, two formula units per unit cell), can be either an early 3d metal (Ti, V, Cr) or a nonmagnetic element of different groups (Al, Si, Mo, W, Re). The structure is presented in Fig. 1. The U atoms occupy the 2a positions, all of them are equivalent. The concentration x of element M does not exceed 3 per formula unit (with the exception of Al) which is not enough to completely fill one type of nonequivalent Mn position; the T and M atoms are distributed over all Mn sites with different partial preference for different M.

The compounds of this class with T = Fe (including the Fe analogue of $UCo_{10}Si_2$, $UFe_{10}Si_2$), which are promising materials for permanent magnets, have been investigated in detail, in some cases on single crystals (see review papers in Refs. [1,2]). The compounds with T = Co have not attracted much attention so far. In the present paper, we report on the magnetic properties of $UCo_{10}Si_2$, studied on aligned powder, including the first data on the magnetic anisotropy of the compound. The properties are compared with those of $YCo_{10}Si_2$ with nonmagnetic Y.

2. Experimental details

The $UCo_{10}Si_2$ alloy was prepared by melting the components (uranium purity 99.8%, cobalt and silicon 99.99%) in an arc furnace on a water-cooled copper bottom under a protective argon atmosphere. The ingot was turned several times to avoid inhomogeneities, and afterwards was annealed at 900°C for 1 week. Standard X-ray, metallographic and thermomagnetic analyses showed the single-phase state of the alloy (with lattice parameters $a = 822.5$ pm and $c = 464$ pm). The ingot was crushed into a powder of particle size approximately 50 μm. The powder was mixed with epoxy and aligned in a steady field of 0.5 T.

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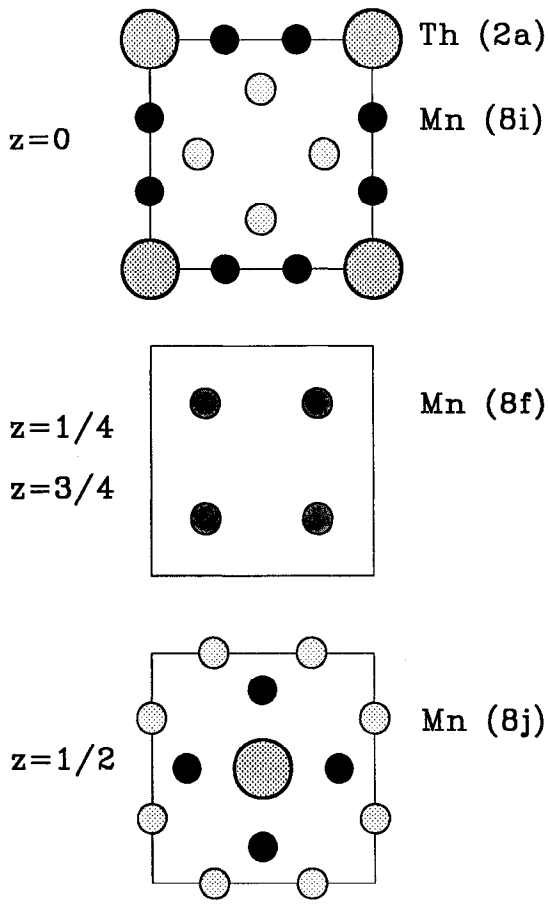

Fig. 1. $ThMn_{12}$-type crystal structure.

Magnetization was measured by an induction method in pulsed fields up to 10 T in the temperature range 4.2–300 K. Along the hard-magnetization direction at 4.2 K, it was also measured up to 40 T. For determination of the absolute value of magnetic moment, a room-temperature vibrating sample magnetometer was used with steady field up to 2 T. The Curie temperature was determined by d.c. susceptibility temperature scan.

3. Results and discussion

As usual for the U intermetallics, especially with 3d metals, the main problem is to determine the magnetic state of U. Among the intermetallics containing both U and Co there are only a few magnetically ordered compounds known, and a coexistence of U and Co magnetism has never been found. For example, only Co carries a magnetic moment in $UCo_{5.3}$ and $Y_{1-x}U_xCo_5$ [3,4]. On the contrary, in the hydride $UCoH_{2.7}$ [5] and in the compounds of the UTX group (UCoGa, UCoSn) [6] only U is magnetic. In the compound under consideration, $UCo_{10}Si_2$, and in another high Co content ternary intermetallic, $U_2Co_{15}Ge_2$, a large magnetic moment and Curie temperature originate from the Co sublattice, whereas the magnetic state of uranium is unknown [7–10].

$UCo_{10}Si_2$ is found to be a rather strong ferromagnet with both high Curie temperature ($T_C = 550$ K) and molecular magnetic moment at 4.2 K ($\mu_m = 8.6\ \mu_B$ as determined by extrapolation of the high-field magnetization curve along the axis of alignment to zero field). These relatively high T_C and μ_m values certainly originate from the Co sublattice. Nevertheless, they are considerably lower than in the Y analogue. In $YCo_{10}Si_2$, $T_C = 750$ K and $\mu_m = 11.0\ \mu_B$ [11]. This is similar to the relationship between binaries $UCo_{5.3}$ and YCo_5. The U atoms do not carry a magnetic moment in $UCo_{5.3}$; the additional electrons from U are transferred to the 3d band of Co, filling it and decreasing both T_C and μ_m. Consequently, the compound is a weakened analogue of YCo_5 [3].

However, we found $UCo_{10}Si_2$ to exhibit a strong uniaxial magnetocrystalline anisotropy, whereas $YCo_{10}Si_2$ has multiaxial anisotropy of the cone type [11]. Fig. 2 shows the magnetization curves along the axis of alignment at 4.2 and 300 K and perpendicular to it. They are characteristic of the case of uniaxial anisotropy of a compound with imperfect alignment; this reflects in an initial rise of magnetization in the hard direction. X-ray diffraction patterns of randomly oriented and magnetically aligned powders of $UCo_{10}Si_2$, presented in Fig. 3, also show clearly the uniaxial type of anisotropy. In Fig. 3, the parallel and perpendicular alignments are with respect to the surface of the sample for standard powder diffractometry.

High-field magnetization curves along the axis of alignment at 4.2 K and perpendicular to it are shown in Fig. 4. The hard-direction magnetization curve has no field-induced transition, which was found in $UFe_{10}Si_2$. The anisotropy field B_a reaches 13 T at 4.2

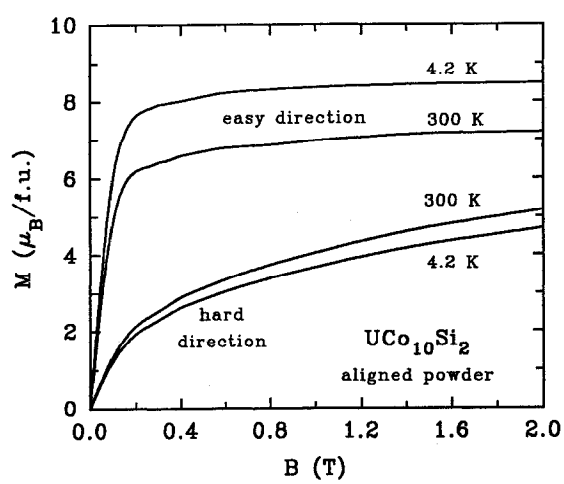

Fig. 2. Low-field part of the magnetization curves at 4.2 K and 300 K.

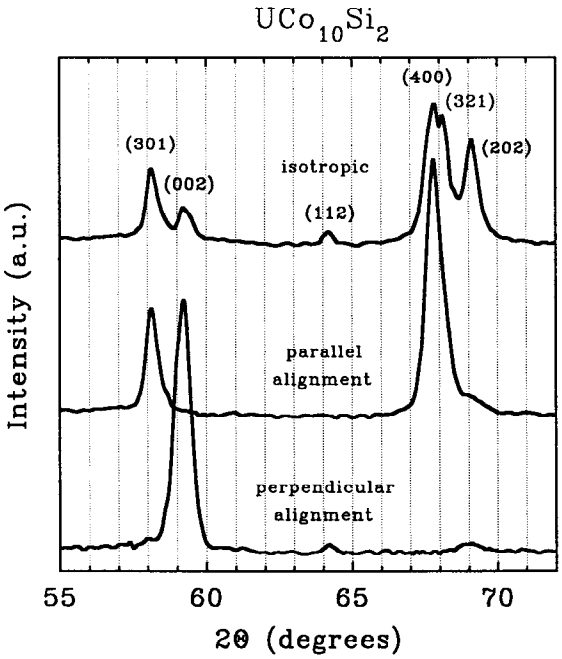

Fig. 3. X-ray patterns of randomly oriented and magnetically aligned powders of $UCo_{10}Si_2$.

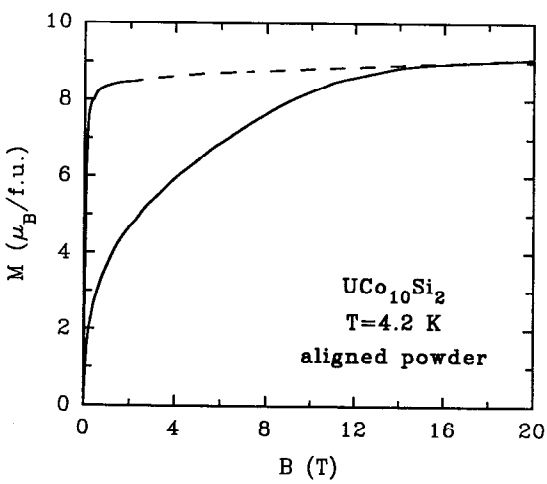

Fig. 4. High-field magnetization curves at 4.2 K.

K. The value of B_a was determined by the single-point detection method as a field where the second derivative d^2M/dB^2 has a minimum. This corresponds to the first anisotropy constant $K_1 = \mu_m B_a/2 = 3.3$ MJ m^{-3}. At room temperature, $\mu_m = 7.2\mu_B$, $B_a = 8$ T and $K_1 = 1.7$ MJ m^{-3}.

If the difference in anisotropy between $UCo_{10}Si_2$ and $YCo_{10}Si_2$ is attributed to the U sublattice, this might point to a magnetic state of U in $UCo_{10}Si_2$, as found in the Fe isostructural analogue $UFe_{10}Si_2$. In such a case, $UCo_{10}Si_2$ would be the first compound where both the U and Co atoms carry an ordered magnetic moment. It is rather difficult to study the U magnetic moment in the presence of strong ferromagnetism of other sublattices because the expected value of μ_U is at about the level of experimental error of the total moment. Low μ_U is frequently a result of cancellation of spin and orbital moments, whereas each of them can be rather large. Therefore, the U magnetism can manifest itself mainly in the magnetic anisotropy and/or magnetostriction connected with orbital moment (as was found in UFe_2 [12]), but not in the contribution to the total magnetic moment of the compound. In $UFe_{10}Si_2$, μ_U is estimated to be 0.5 μ_B compared with $\mu_m = 17$ μ_B [13]. In both UFe_2 and $UFe_{10}Si_2$, the magnetic moment of the 3d metal is reduced considerably compared with the Y analogues, as is found for $UCo_{10}Si_2$. In $UFe_{10}Si_2$, there are two kinds of evidence for U magnetism. The first is a much larger T_C, than in $YFe_{10}Si_2$, which is attributed to the U–Fe exchange-interaction contribution. In $UCo_{10}Si_2$, the U–Co contribution, if it exists, is certainly lower than the reduction of the Co–Co contribution because of a much lower T_C compared with $YCo_{10}Si_2$. The second is the field-induced transition in the hard direction which has never been seen in the Y–T intermetallics, but is frequently observed in the R–T compounds with magnetic R. In $UCo_{10}Si_2$ there is no such transition.

The difference in anisotropy type between $UCo_{10}Si_2$ and $YCo_{10}Si_2$ is thus the only argument found for U contribution to the magnetism. The uniaxial anisotropy of $UCo_{10}Si_2$ is relatively large; K_1 exceeds its available values for $YT_{12-x}M_x$ from single-crystal magnetization curves or single-point detection method on polycrystals by a factor of 2–3. Unfortunately, there are no quantitative data on K_1 for $YCo_{10}Si_2$. However, the cone-type anisotropy found in this compound points to a low absolute K_1 value, because in the cone range a ratio

$$|K_1| \leq 2K_2 \quad (1)$$

should be fulfilled, and the second anisotropy constant K_2 has never been found to be noticeable in Y–T intermetallics. Moreover, K_1 is negative in the multiaxial case. In the isostructural analogue $YCo_{11}Ti$, a basal-plane anisotropy with $K_1 \approx -0.9$ MJ m^{-3} was found [14]. Owing to a lower Co content in $YCo_{10}Si_2$, the absolute value of K_1 is expected to be smaller, in agreement with observed cone-type anisotropy. Therefore, the large positive value of $K_1 = 3.3$ MJ m^{-3} found in $UCo_{10}Si_2$ might be attributed completely to the U sublattice. Practically the same $K_1 = 3.0$ MJ m^{-3} was observed in $UFe_{10}Si_2$ [15], but this was due to a considerable uniaxial contribution of the Fe sublattice; the part attributed to U sublattice was estimated as 1.6 MJ m^{-3}.

Nevertheless, another explanation of the observed results without the U contribution is also possible. In the case of Fe, all the $YFe_{12-x}M_x$ compounds have a uniaxial anisotropy magnitude slightly dependent on

the nonmagnetic component M. Studies of solid solutions $YFe_{12-x-y}Co_yM_x$ showed that uniaxial anisotropy decreases with increasing Co content; that means that Co actually has a negative contribution to the anisotropy [1,11]. However, contrary to the results of Ref. [14], the uniaxial anisotropy was observed in $YCo_{11}Ti$ [16]. The X-ray patterns of aligned powders presented in Ref. [16], similar to those shown in Fig. 3, show the uniaxial anisotropy without any doubt. For an explanation of such a contradiction, one can propose that $YCo_{11}Ti$ has a certain homogeneity range and K_1 changes its sign within this range, as was observed in the related compound Y_2Co_{17} [17,18]. This may be possible because T_C values reported for $YCo_{11}Ti$ in Refs. [14] and [16] differ considerably (943 K and 1050 K respectively). Recently, this homogeneity range was actually found to be rather large in $YCo_{11}Ti$ [19] and the uniaxial anisotropy was confirmed [20]. The sensitivity of magnetic properties and, in particular, magnetic anisotropy to the Co content might be understood by taking account of the three nonequivalent positions for T atoms in the $ThMn_{12}$-type lattice (Fig. 1) with different local properties [1]. In the case of competitive anisotropy, a slight change in distribution of Co atoms over the nonequivalent sites can lead to a considerable change in total anisotropy. If the anisotropy of Co compounds is rather sensitive to this factor, the difference in anisotropy type between $UCo_{10}Si_2$ and $YCo_{10}Si_2$ could also be attributed to different Co–Si occupancy of the positions, but not to the U contribution.

The anisotropy of uniaxial $YCo_{11}Ti$ was determined in Ref. [16] only at room temperature as $K_1 = 0.75$ MJ m^{-3}. Since the compound has a very high Curie temperature, $K_1(T)$ dependence should be rather smooth between 4.2 K and room temperature and K_1(4.2 K) should not exceed 1.5 MJ m^{-3}; in the basal-plane $YCo_{11}Ti$ [14], $K_1 = -0.5$ MJ m^{-3} at room temperature compared with -0.8 MJ m^{-3} at 77 K. K_1 of $UCo_{10}Si_2$ is considerably larger than this estimated value for possible positive Co contribution, and $\Delta K_{1U} \approx 1.8$ MJ m^{-3} still remains for the U contribution. This is practically the same as that found for ΔK_{1U} in $UFe_{10}Si_2$ [13,15]. The anisotropy of other high Co content compounds containing U was measured on single crystals of $UCo_{5.3}$ [3] and $U_2Co_{15}Ge_2$ [10]. K_1 was found to be 2.0 MJ m^{-3} in uniaxial $UCo_{5.3}$ and -1.7 MJ m^{-3} in $U_2Co_{15}Ge_2$ with basal-plane anisotropy. In $UCo_{5.3}$ it is attributed to the Co sublattice; U is found to be nonmagnetic. For $U_2Co_{15}Ge_2$, a possible U contribution cannot be determined without comparison with the Y analogue. The magnetic anisotropy in $UCo_{10}Si_2$ is noticeably larger than that of $UCo_{5.3}$ (and also larger than the absolute value of K_1 in $U_2Co_{15}Ge_2$, being attributed completely to the Co sublattice). Therefore, even taking into account possible uniaxial Co contribution to the anisotropy of $UCo_{10}Si_2$, we can still insist on the magnetic state of U.

The weak point of the above arguments is the fact, that K_1 in $UCo_{10}Si_2$ considerably exceeds the anisotropy of the Co sublattices only in isostructural or closely related compounds. They exhibit anisotropy as strong as in pure Co ($K_1 \approx 1$ MJ m^{-3}). There would be no doubt concerning the U magnetism in $UCo_{10}Si_2$, if K_1 exceeds any possible Co anisotropy. However, in principle, in compounds with other structures and stoichiometries, Co can provide higher anisotropy, up to $K_1 \approx 7$ MJ m^{-3} in YCo_5 (e.g. see Ref. [4]). For this reason, any additional evidence is desirable.

4. Conclusion

The strong uniaxial magnetic anisotropy found in $UCo_{10}Si_2$, instead of the multiaxial anisotropy found in $YCo_{10}Si_2$ (the isostructural analogue with nonmagnetic Y), might point to a magnetic state of uranium. In this case, $UCo_{10}Si_2$ would be the first compound, where ordered magnetism of U and Co coexists.

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References

[1] H.S. Li and J.M.D. Coey, in K.H.J. Buschow (ed.), *Handbook of Magnetic Materials*, Vol. 6, North-Holland, Amsterdam, 1991, p. 1.

[2] W. Suski, in K.A. Gschneidner, Jr. and L. Eyring (eds.), *Handbook on the Physics and Chemistry of Rare Earths*, North-Holland, Amsterdam, in press.

[3] A.V. Deryagin and A.V. Andreev, *Sov. Phys. JETP, 44* (1976) 610.

[4]. A.V. Deryagin, A.V. Andreev and V.A. Reimer, *Sov. Phys. JETP, 47* (1978) 933.

[5] A.V. Andreev, M.I. Bartashevich, A.V. Deryagin, L. Havela and V. Sechovsky, *Phys. Status Solidi A, 98* (1986) K47.

[6] V. Sechovsky and L. Havela, in E.P. Wohlfarth and K.H.J. Buschow (eds.), *Ferromagnetic Materials*, Vol. 4, North-Holland, Amsterdam, 1988, p. 309.

[7] W. Suski, A. Baran, K. Wochowski and T. Mydlarz, *J. Magn. Magn. Mater., 95* (1991) L123.

[8] T. Berlureau, B. Chevalier, P. Gravereau, L. Fournes and J. Etourneau, *J. Magn. Magn. Mater., 102* (1992) 166.

[9] T. Berlureau, B. Chevalier, L. Fournes and J. Etourneau, *Mater. Lett., 9* (1989) 21.

[10] Y. Shiokawa, M. Hara, Y. Haga, R. Amano and M Takahashi, *J. Alloys Comp., 213–214* (1994) 513.

[11] C. Lin, G.Z. Li, Z.X. Liu, H.W. Jiang, Z. Wan, J.L. Yang, B.S. Zhang and Y.F. Ding, *J. Appl. Phys., 70* (1991) 6543.

[12] Yu.F. Popov, R.Z. Levitin, A.V. Deryagin, M. Zeleny and A.V. Andreev, *Sov. Phys. JETP, 51* (1980) 1223.
[13] A.V. Andreev, F.G. Vagizov, W. Suski and H. Drulis, *J. Alloys Comp., 187* (1992) 401.
[14] M. Solzi, L. Pareti, O. Moze and E. David, *J. Appl. Phys., 64* (1988) 5084.
[15] A.V. Andreev, M.I. Bartashevich, H. Aruga Katori and T. Goto, *J. Alloys Comp., 216* (1994) 221.
[16] K. Odashi, H. Ido, K. Konno and Y. Yoneda, *J. Appl. Phys., 70* (1991) 5986.
[17] S. Yajima, M. Hamano and H. Umebayashi, *J. Phys. Soc. Jpn., 32* (1972) 861.
[18] M. Hamano and S. Yajima, *Trans. Jpn. Inst. Met., 20* (1979) 57.
[19] J.H.V.J. Brabers, D.C. Zeng, N. Tang, F.R. de Boer and K.H.J. Buschow, *J. Magn. Magn. Mater., 140–144* (1995) 1087.
[20] J.H.V.J. Brabers, D.C. Zeng, N. Tang, F.R. de Boer and K.H.J. Buschow, in press.