Giant magnetoelastic distortions of the crystal structure of the weakly anisotropic compound UFe₂

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We investigate the temperature dependences of the crystal-lattice parameters, of the anisotropy, and of the Young's modulus of the intermetallide compound UFe₂. The contribution of the uranium ions to the magnetic moment of this compound is negligible ($\mu_U = 0.06\mu_B$), owing to the almoft complete delocalization of the uranium $5f$ electrons. The measured magnetic anisotropy is relatively small ($K_1 \approx -10^6$ erg/cm³ at 0 K). However, the transition to the magnetically ordered state ($T_c = 170$ K) is accompanied by large rhombohedral distortions of the UFe₂ crystal structure and by a considerable anomaly of the Young's modulus, thus attesting to a large value of the magnetoelastic interaction in this compound ($\Delta K_1{}^{me} \approx -8 \times 10^6$ erg/cm³).

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The transition to the magnetically ordered state produces crystal-structure distortions in many actinide compounds.[1,2] These distortions are usually observed in magnets having a considerable magnetic anisotropy energy and are explained in the single-ion model as being the result of a magnetoelastic interaction between the localized $5f$ electrons of the actinides and the crystal lattice field.[1-4] At the same time it is assumed that if the $5f$ electrons are collectivized and the magnetic anisotropy and the magnetoelastic interaction are small.[2-5]

However, the nature of the magnetic anisotropy and of the magnetoelastic interaction of actinide compounds is not fully clear to this day. In particular, we do not know the degree of delocalization of the $5f$ electrons at which the decrease of the magnetic anisotropy and of the magnetoelastic interaction becomes noticeable. It is of interest therefore to investigate the magnetic anisotropy and the distortions of the crystal structure following the magnetic ordering of the intermetallide compound UFe₂. This compound has at room temperature a cubic structure of the MgCu₂ type (space group Fd3m). Below the Curie temperature ($T_c = 158 - 195$ K for various samples, see, e.g., Ref. 6, where earlier work is cited), UFe₂ becomes ferromagnetic state. The saturation magnetic moment is $(1.02 - 1.36)\mu_B$ (Ref. 6).[2] Neutron-diffraction investigations[7] have shown that the magnetic moment of UFe₂ is due mainly to the moment of the iron, and the localized moment of uranium is very small in this compound, $0.06\mu_B$ at 4.2 K, smaller by practically two orders of magnitude than the moment of the trivalent uranium ion. Thus, the $5f$ electrons of the uranium in UFe₂ are almost completely delocalized, its magnetic properties are described within the framework of the collectivized magnetism model,[3,4,5] and if the arguments advanced above are correct, one can expect this compound to have low anisotropy and magnetoelastic-interaction energies.

The UFe₂ samples were obtained by smelting the components in an arc furnace, followed by remelting in a resistance furnace in helium at slightly above the melting temperature, so that the ingot had a coarse grain structure. Metallographic, x-ray diffraction, and x-ray microspectral measurements have shown that the alloy is single-phase and corresponds to the stoichiometric composition of UFe₂. Individual grains of the ingot were used to produce spherical and cubic single-crystal samples 3–4 mm in size. To relieve the mechanical stresses, the samples were annealed for six hours at 1000 °C and cooled with the furnace. Laue patterns from different sides were obtained to check on the monocrystallinity, and the grain disorientation did not exceed 1.5 – 2°.

The magnetization of the UFe₂ was measured in the temperature interval 8–200 K with a vibration magnetometer. The magnetic anisotropy constants were determined from the torsion curves in the (110) and (111) planes of the crystal using an Akulov-type anisometer (at 77 – 150 K), and also an anisometer with strain sensors[9] at 4.2 – 150 K. The magnetic-anisotropy constants were calculated with a computer using a program that takes into account the deviation of the magnetization vector from the field direction. The crystal structure and the lattice parameters were investigated with a "Gegerflex" diffractometer in the temperature interval 4.2 – 300 K. The Young's modulus was measured with a compound vibrator for a polycrystalline UFe₂ sample at 4.2 – 250 K.

The magnetization measurements have shown that at low temperatures UFe₂ is a ferromagnet. The Curie temperature obtained in accord with the theory of second-order phase transitions is 170 ± 2 K, the magnetic moment per formal unit, extrapolated to 0 K, is $1.16 \pm 0.04\mu_B$. These values agree with data given by others for UFe₂ of stoichiometric composition.[6]

The temperature dependences of the first (K_1) and second (K_2) magnetic-anisotropy constants are shown in Fig. 1. K_1 and K_2 decrease monotonically in absolute value with increasing temperature, with $K_1 < 0$, $K_2 > 0$ and the directions of the easy magnetization, are axes of the type $\langle 111 \rangle$, in agreement with other studies.[6,7]

It follows from our data that the magnetic anisotropy of UFe₂ is small: when calculated per iron atom it is of the same order as the anisotropy of metallic iron[10] and of its intermetallic compounds with nonmagnetic
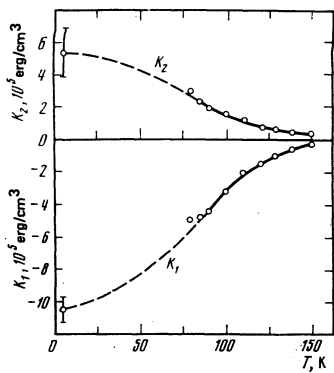
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FIG. 1. Temperature dependence of the anisotropy constants of UFe_2.

TABLE I.

	$\varepsilon_0 \cdot 10^3$	ΔK_1^{me}, 10^8 erg/cm^3	K_1, 10^8 erg/cm^3
UFe_2	2.9	−0.076	−0.011
$NpFe_2$	7.94 [3]	−0.573	10 [15] *
$TbFe_2$	4.5 [12, 13]	−0.49	−7 [16]
$TbCo_2$	4.4 [14]	−0.468	−2 [12] **

*Rough estimate from polycrystal mag-
netization curves.
**Estimate from single-crystal mag-
netization curves.

elements (for example the compound $LuFe_2$ which is isostructural with UFe_2, Ref. 11). Thus, investigations of the magnetic anisotropy of UFe_2 would seem to confirm results obtained by others, that uranium ions, owing to almost complete delocalization of the $5f$ electrons, make no contribution to this compound's magnetic anisotropy, which is determined mainly by the iron.

Since it is usually assumed that in uranium compounds the magnetic anisotropy and the magnetoelastic interaction are due to the same cause—interaction between the localized $5f$ electrons and the crystal lattice field—the magnetoelastic interaction in UFe_2 and the distortions of its crystal structure on going into the ferromagnetic state are expected to be small.

X-ray measurements have shown, however, that this is not the case. In the paramagnetic region the UFe_2 structure is face-centered cubic, and the unit-cell parameter is $a = 7.057$ Å at room temperature and decreases with decreasing temperature (Fig. 2). The transition into the ferromagnetic state is accompanied by the rhombohedral distortions of the crystal structure: the length of the cube body diagonal [111], which is parallel to the magnetization direction, increases (it follows from our data that the crystal breaks up into four domains in accord with the number of the possible easy-magnetization axes of type $\langle 111 \rangle$). As a result, the angle α between neighboring edges of the cube,
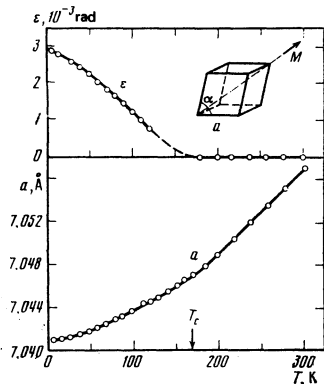

FIG. 2. Temperature dependence of the rhombohedral distortions of the UFe_2 crystal lattice; ε is the deviation of the angle α from $\pi/2$ and a is the lattice parameter.

which is equal to $\pi/2$ in the paramagnetic phase, decreases. The temperature dependences of the lattice parameter a and the deviation ε of the angle α from $\pi/2$ ($\varepsilon = \pi/2 - \alpha$) are shown in Fig. 2 (for a comparison with the fcc structure in the paramagnetic region, we consider in the ferromagnetic state not a primitive cell, as usual, but a face-centered rhombohedral cell). It is seen that the rhombohedral distortions of the UFe_2 structure increase with decreasing temperature and at 4.2 K they reach a value $\varepsilon_0 = 10'(2.9 \cdot 10^{-3}$ rad).

The rhombohedral distortion ε_0 of UFe_2 is compared in the table with the published data for other rare-earth and actinide intermetallic compounds with largest presently known rhombohedral distortions. It follows from the table that the value of ε_0 in UFe_2 is smaller by only a factor 1.5–2 than in these compounds. At the same time the magnetic anisotropy of UFe_2 is smaller by two or three orders of magnitude than in other intermetallic compounds with large magnetoelastic distortions of the crystal structure. We have thus in UFe_2 an unusual situation-tremendous magnetoelastic distortions of the crystal lattice with a small magnetic anisotropy.

The magnetoelastic interaction contributes to the measured anisotropy energy. Using the relations given in Ref. 17, we easily obtain for the magnetoelastic contribution made to the constant K_1 by the rhombohedral distortions[3]

$$\Delta K_1^{me} = -\tfrac{9}{2} c_{44} \varepsilon^2. \tag{1}$$

The temperature dependence of ΔK_1^{me} for UFe_2 is shown in Fig. 3 (we used in the calculations a value 2.02×10^{11} dyn/cm^2 for the elastic constant[4] c_{44}). The table lists the values of ΔK_1^{me} of various intermetallic compounds at 4.2 K (the elastic constant c_{44} of $NpFe_2$ was assumed to be the same as that of UFe_2, and for $TbFe_2$ and $TbCo_2$ we used the value 5.38×10 dyn/cm^2, Ref. 18). It is seen that whereas in compounds of terbium and neptunium ΔK_1^{me} is much less than K_1, in UFe_2 the magneto-elastic contribution to the anisotropy exceeds the measured value by an order of magnitude.

The measured anisotropy constant K_1 is the sum of the anisotropy constant K_1^0 of the undeformed lattice and the contribution made to the anisotropy constant by the magnetoelastic interaction ΔK_1^{me}:

$$K_1 = K_1^0 + \Delta K_1^{me}. \tag{2}$$

Knowing the measured anisotropy constant K_1 and the magnetoelastic contribution made to it by ΔK_1^{me}, we
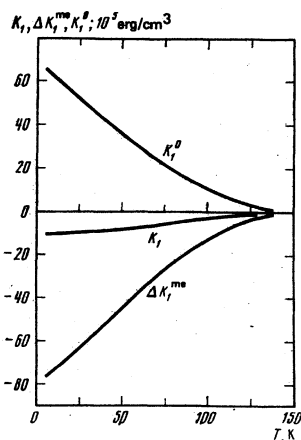
FIG. 3. Temperature dependence of K_1 and of the calculated values of the anisotropy constant K_1^0 of the undeformed lattice lattice and of the contribution ΔK_1^{me} made to the anisotropy by the magnetoelastic interactions.

FIG. 4. Temperature dependence of the Young's modulus of UFe_2.

have obtained the temperature dependence of the anisotropy constant K_1^0 of the undeformed UFe_2 lattice (Fig. 3). At low temperatures $|K_1| \ll |\Delta K_1^{me}|$, therefore $K_1^0 \approx -\Delta K_1^{me}$. Consequently the small value of the measured anisotropy of UFe_2 is the result of "accidental" mutual compensation of two large quantities: the anisotropy of the underformed lattice and the anisotropy due to the magnetoelastic interaction. The situation is similar also in some other magnets. For example, in terbium iron garnet $Tb_3Fe_5O_{12}$ at low temperatures $\Delta K_1^{me} > K_1^0$, and in mixed terbium-yttrium iron garnets $Tb_{3-x}Y_xFe_5O_{12}$ at $x \approx 2.2$ the anisotropy of the undeformed anisotropy is cancelled by the anisotropy due to the magnetoelastic interaction.[19]

It follows thus from our measurements that the anisotropy of undeformed UFe_2 is large (of the order of 10^7 erg/cm^3) and, naturally, cannot be due only to the contribution of the iron. If it is assumed, as is usually done,[2-5] that the delocalized $5f$ electrons of the uranium make no contribution to the anisotropy, then it must be proposed that the large magnetic anisotropy of UFe_2 is due to that small fraction of $5f$ electrons which remains localized. In other words, the localized part of the spin density of the $5f$ electrons remains quite anisotropic, and this leads to a high anisotropy energy and magnetoelastic energy, whereas the uranium magnetic moment due to the localized $5f$ electrons is small.

We note that a similar situation obtains in Sm^{3+}, for example in rare-earth iron garnets. In these compounds the magnetic moment of the Sm^{3+} ion is close to zero, because of the effect of the crystal field on the state of this ion. At the same time, the magnetic moment of Sm^{3+} in iron garnets is $L \neq 0$, and the one-ion magnetic anisotropy due to the interaction of the anisotropic cloud of the $4f$ electrons of the Sm^{3+} with the crystal lattice field, is large in iron garnets.[20]

Consequently, measurements of the magnetoelastic properties of UFe_2 show that this compound cannot be regarded, as is usually done, as a ferromagnet with fully delocalized $5f$ electrons of uranium—the presence of a small fraction of localized $5f$ electrons affects its
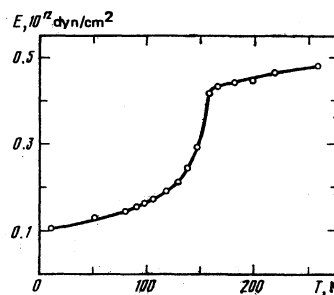
properties strongly.

In ferromagnetics below the Curie point, an anomaly of Young's modulus is observed and is due to the influence of external elastic stresses on the domain structure and on the orientation of the magnetic moments in the domains.[10] Our measurements have shown that the magnetoelastic anomaly of the Young's modulus of UFe_2 is large (Fig. 4): when cooled from the Curie temperature to 4.2 K, the Young's modulus decreases by an approximate factor of five. It follows from the theory[21] that the decrease of the Young's modulus as a result of rotation of the magnetic moments can be represented in the form

$$\Delta E/E = A(\Delta K_1^{me}/K_1), \tag{3}$$

where A is a coefficient of the order of unity. This relation is satisfied for UFe_2.

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