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Magnetic Properties of UFe_{2+x} Prepared by Splat Cooling

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A series od UFe_{2+x} materials was prepared using splat cooling. The Laves phase structure can accommodate up to 0.3 Fe excess, while T_C is enhanced from 172 K to approximately 240 K. Higher Fe concentration leads to the segregation of α-Fe. ^{57}Fe Mössbauer spectroscopy indicates higher Fe magnetic hyperfine fields on Fe nuclei occupying the U sublattice than for the regular Fe sites.

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1. General information

Magnetic properties of uranium-based compounds are related to the character of the $5f$ electronic states, ranging between a localized character and itinerancy (see e.g. [1]).

UFe_2 is the first uranium compound reported to exhibit ferromagnetism [2]. The Curie temperature T_C given by various authors differs in some extent, but remains in the vicinity of 160 K in most of cases (see [1] and references therein).

The itinerant character of magnetism was deduced on the basis of suppression of T_C and spontaneous magnetization μ_s by pressure [3]. Neutron-diffraction study [4] revealed that UFe_2 involves both $3d$ and $5f$ magnetism. The main contribution to the spontaneous magnetization comes from Fe ($0.60\mu_B$/Fe), while U spin and orbital moments practically compensate.

The U–Fe phase diagram contains two intermediate phases, namely U_6Fe (formed non-congruently, weakly paramagnetic, superconducting [1]), and UFe_2. Amorphous U–Fe systems have been successfully prepared in the vicinity of the two deep eutectic points, occurring at $U_{17}Fe_{83}$ and $U_{66}Fe_{34}$, respectively. Amorphous systems with the composition close to U_6Fe are superconducting, in analogy to the crystalline phase [5]. Approaching the latter eutectic point, weakly ferromagnetic clusters start to arise below about 120 K [6]. Amorphous material in the vicinity of the former eutectic point ($U_{27}U_{73}$) was found to exhibit magnetic ordering with spin glass features below $T = 32$ K [7].

In this work we concentrated on a more Fe-rich part of the phase diagram. The cubic Laves phase UFe_2 was reported to exist over a certain concentration range. Fe deficient samples exhibit a reduction of T_C from 162 K for UFe_2 down to 112 K for $UFe_{1.7}$ [8]. The Fe deficiency manifests in an increase in the lattice parameter a from 705.7 pm for UFe_2 to 708.7 pm for $UFe_{1.7}$. An attempt to prepare UFe_2 with Fe excess by ball milling [9] has led to an increase in T_C for the amorphous phase up to 207 K. One should note that the high melting point of UFe_2 (1228°C) prevents to prepare amorphous UFe_2 by common fast cooling methods. Here we describe results of splat-cooling synthesis of the materials with the nominal stoichiometry from UFe_2 to UFe_6.

2. Experimental results and discussion

Our magnetization studies readily indicated that the ordering temperatures, which increase marginally for splat-cooled (SpC) UFe_2 [10], can increase markedly by the Fe excess, reaching 220–240 K for $UFe_{2.3}$ (Figs. 1, 2), while the respective anomaly becomes somewhat smeared out. A further increase in the Fe content led to a segregation of α-Fe. This was directly revealed by scanning electron micrography. The grains of the U-containing material are small (< 1 μm). Only for SpC UFe_2 their size reaches ≈ 5 μm [11]. X-ray diffraction indicates that the cubic Laves phase accommodates the excess of 0.3 Fe by the Fe occupation of U sites, shrinking the lattice parameter a. The observed tendency of the relation of T_C and a extrapolates the original dependence known for U excess which can be related to a larger lattice parameter induced by atomic disorder for the SpC material. As seen from Fig. 1, such shift in $T_C(a)$ could be expected from the pressure dependence of UFe_2 [3].

The increase in T_C is accompanied by a dramatic increase in spontaneous magnetization at 4.2 K (not shown here) from $1.0\mu_B$/f.u. in UFe_2 (difference of bulk and SpC is small) to $1.9\mu_B$/f.u. in $UFe_{2.3}$. The reason for such increase is well seen from the ^{57}Fe Mössbauer spectroscopy, performed both at room temperature (displaying a doublet spectrum plus the α-Fe sextet for higher Fe content) and at $T = 50$ K. The refinement reveals, besides the two sextets belonging to the two magnetically inequivalent sites known for UFe_2, a new component belonging undoubtedly to Fe placed into the U sublattice.

Such Fe atoms have the isomer shift by 0.11 mm/s higher comparing to the Fe sites in the Fe sublattice (pointing to a lower hybridisation with U states for the new type of sites). The magnetic hyperfine field B_{hf} is nearly doubled for such Fe

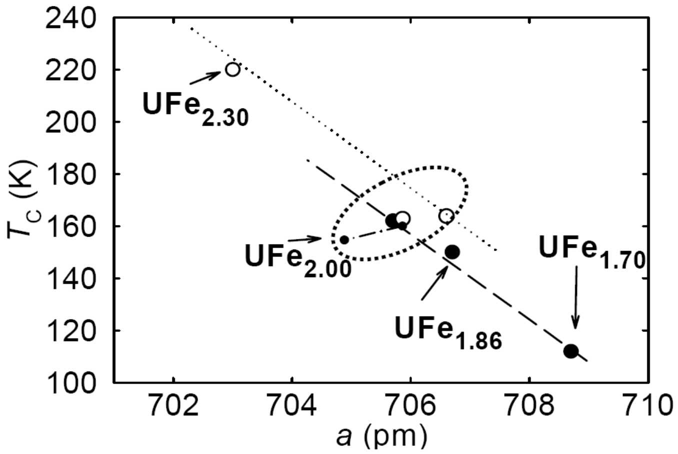

Fig. 1. Relation of T_C and the lattice parameter a for various UFe_x systems. Full symbols mark values on bulk UFe_{2-y} samples [2], empty symbols are our data on bulk UFe_2 and the splats. The short dash-dotted line shows the pressure dependence of T_C for bulk UFe_2 [5] using the experimental bulk modulus $B_0 = 239$ GPa.

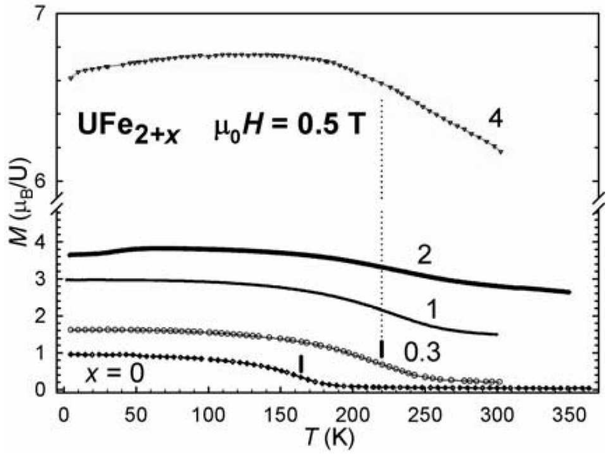

Fig. 2. Temperature dependence of magnetization in $\mu_0 H = 0.5$ T (zero-field-cooled mode) for UFe_{2+x} splats. Vertical bars and the dashed line indicate the respective T_C values.

antistructure atoms, reaching 8.4 T, while the Fe sublattice has the B_{hf} values still enhanced (to 4.4 and 4.1 T, respectively) comparing to the Fe sublattice in UFe_2 ($B_{hf} = 3.5$ and 2.9 T, respectively). Assuming the zero total magnetic moment on U atoms is preserved, the antistructure Fe atoms should contribute by a sizeable moment (1.7 μ_B each) to explain the total magnetization, if the magnetic moments of the Fe sublattice are assumed to be proportional to the B_{hf} values. With the

Fe concentration increasing over $UFe_{2.3}$, the spectra exhibit only the increase in the α-Fe component.

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