Magnetic Properties of $\text{UFe}_{2+\delta}$ Prepared by Splat Cooling

L. Havela\textsuperscript{a}, K. Miliyanchuk\textsuperscript{a}, J. Pešička\textsuperscript{a}, A.P. Gonçalves\textsuperscript{b}, J.C. Waerenborgh\textsuperscript{b}, L.C.J. Pereira\textsuperscript{b}, E. Šantavá\textsuperscript{c} and J. Šebek\textsuperscript{c}

\textsuperscript{a}Charles University, Faculty of Mathematics and Physics, Department of Condensed Matter Physics, Ke Karlovu 5, 121 16 Prague 2, Czech Republic
\textsuperscript{b}Dep. Química, Instituto Tecnológico e Nuclear, 2686-953 Sacavém, Portugal
\textsuperscript{c}Institute of Physics, Academy of Sciences of the Czech Republic, Prague, Czech Republic

A series of $\text{UFe}_{2+\delta}$ 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 $\alpha$-Fe. $^{57}$Fe Mössbauer spectroscopy indicates higher Fe magnetic hyperfine fields on Fe nuclei occupying the U sublattice than for the regular Fe sites.

PACS numbers: 71.28.+d, 75.50.Bb

1. General information

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

$\text{UFe}_2$ 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 $\mu_s$ by pressure [3]. Neutron-diffraction study [4] revealed that $\text{UFe}_2$ involves both 3\textit{d} and 5\textit{f} 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$_6$Fe (formed non-congruently, weakly paramagnetic, superconducting [1]), and $\text{UFe}_2$. 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 $U_{6}Fe$ 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 $UFe_2$ ($1228^\circ$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 $UFe_0$.

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 $\alpha$-Fe. This was directly revealed by scanning electron micrography. The grains of the U-containing material are small ($< 1 \mu m$). Only for SpC $UFe_2$ their size reaches $\approx 5 \mu m$ [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 $\alpha$-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
Magnetic Properties of UFe$_{2+x}$ Prepared by Splat Cooling

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 ($1.7 \mu_B$ 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 $\alpha$-Fe component.

**Acknowledgments**

This work was supported by the Program GRICES/ASCR 2007, by the Grant Agency of the Czech Republic under the grant No. 202/07/0418, and by the Grant Agency of the Academy of Sciences of the Czech Republic under the grant No. A100100530. It was a part of the research plan MSM 0021620834 financed by the Ministry of Education of the Czech Republic.

**References**


