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High field magnetic investigations of the $UFe_{10-x}Ni_xSi_2$ system*

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Abstract

Magnetic measurements of $UFe_{10-x}Ni_xSi_2$ were performed in magnetic fields up to 14 T at temperatures of 4.2–300 K. The values of the saturation moment amount to about 15 μ_B/f.u. for $UFe_8Ni_2Si_2$. The remanence increases with an increase in the Ni concentration, however, the saturation magnetization and the Curie point decrease, reaching $p_s = 2$ μ_B/f.u. and $T_c = 45$ K, respectively, for $UFeNi_9Si_2$. The shape of the magnetization curves for $x = 9$, 8 and 6 suggests the possibility of the first order magnetization process (FOMP). The comparison of the results of the present investigation and that performed using the ^{57}Fe Mössbauer effect provides evidence of a substantial contribution of the uranium, and possibly the nickel, sublattices to magnetic ordering.

1. Introduction

Our previous investigations have shown that $UNi_{10}Si_2$ is a paramagnet with an inverse magnetic susceptibility following a modified Curie–Weiss law in the temperature range 4.2–300 K [1,2]. The high temperature deviation from this law probably indicates a strong influence of the crystal field interactions as observed in analogous rare earth compounds [3]. The ^{29}Si NMR Knight shift is temperature independent in the 4.2–294 K range [2]. The enhanced γ value [1] and the absence of long range magnetic ordering suggest that $UNi_{10}Si_2$ is near the crossover from an itinerant non-magnetic 5f-system to a magnetically ordered, localized 5f-system. On the other hand, a closely related compound, $UFe_{10}Si_2$, is strongly ferromagnetic below 650 K with considerably high magnetization, but with negligible remanence [4].

The preliminary magnetic examination of $UFe_{10-x}Ni_xSi_2$ has shown that this system exists in exactly single-phase form for $x > 8$ [2]. Because the single-phase samples with small concentration of Fe exhibit relatively high remanence at low temperatures, it seemed interesting to extend preliminary research [2] to a broader composition range ($x \leqslant 8$), in spite of a very small admixture (3 at.%) of an unidentified phase.

The ^{57}Fe Mössbauer effect in these alloys was also investigated to determine the distribution of iron atoms in available crystallographic positions and the magnetic moment of the iron sublattice. A summary of this research is presented in Fig. 1 [5] in the form of a magnetic (T, x) phase diagram. There are two curves corresponding to the boundaries of three magnetic phases. The lower curve is the high temperature limit of a ferromagnetic phase, as determined by Mössbauer measurements, and for $x = 9$ and 9.5, *via* Mössbauer and magnetic methods. the upper curve, which is the low temperature limit of the paramagnetic phase, is determined exclusively by magnetometric measurements, since Mössbauer spectra do not exhibit any

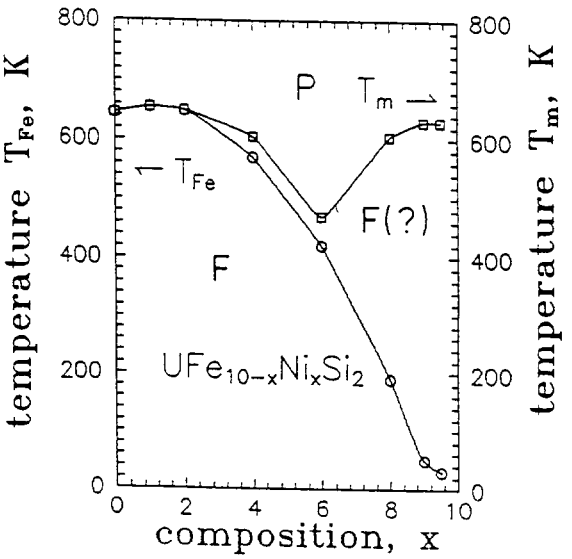

Fig. 1. Magnetic (T, x) phase diagram for the $UFe_{10-x}Ni_xSi_2$ alloys [5].

*This paper is devoted to the memory of the late Bohdan Staliński.

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splitting at temperatures above the lower curve. The magnetic character of the phase existing between both curves is a puzzle at present. The examination of the samples with low Fe concentration [2] seemed to suggest that the high temperature phase transition, T_m, results from the presence of micrograins of free Ni. This idea is confirmed by the lack of evidence of magnetic ordering in the Fe sublattice from the Mössbauer experiment above T_{Fc} and by observation of the same anomaly at the same temperature in $UCo_{0.5}Ni_{9.5}Si_2$ [6]. However, high resolution electron microscopy investigations of these materials [6] failed to confirm this conclusion. Also a decrease in T_m with increasing Fe concentration contradicts the possibility of the presence of the free Ni as a reason for T_m. The phase between the curves exhibits ferromagnetic character, which can be attributed to concentrations of free Ni of about 3 and 7 at.% for $x=9.5$ and 9.0, respectively [2].

In the present paper, we report on high field magnetization investigations up to 14 T and in the temperature range 4.2–300 K.

2. Experimental details

All the samples were obtained by alloying the components in stoichiometric amounts, and the preparation of the samples as well as the crystallographic examinations have been described previously [2,5]. The lattice parameters follow roughly the Vegard rule. Magnetic measurements were carried out using standard magnetometry at the International Laboratory of High Magnetic Fields and Low Temperatures.

3. Results and discussion

Fig. 2 presents the magnetization of the $UFe_{10-x}Ni_xSi_2$ alloys versus temperature measured under a magnetic field of 5 T. One can see that for all x, $M(T)$ slightly decreases with temperature with approximately the same slope. At low temperatures for smaller x ($x \leqslant 6$), there are diffuse maxima observed, whereas for higher concentration of Ni ($x=8$), this maximum becomes more pronounced as can be seen from Fig. 3. The temperature of these maxima roughly corresponds to those observed previously [2], although their source is not clear to us at present. They could result from a domain effect, however; preliminary investigations of domain structure of the samples with higher concentration of Fe [7] do not reveal any temperature dependent anomalies. The possibility of a spin-glass (SG) state seems to be very tempting; what is more, this type of behavior has been detected in other $ThMn_{12}$-type uranium intermetallics [8]. For the investigated alloys, the presence of three

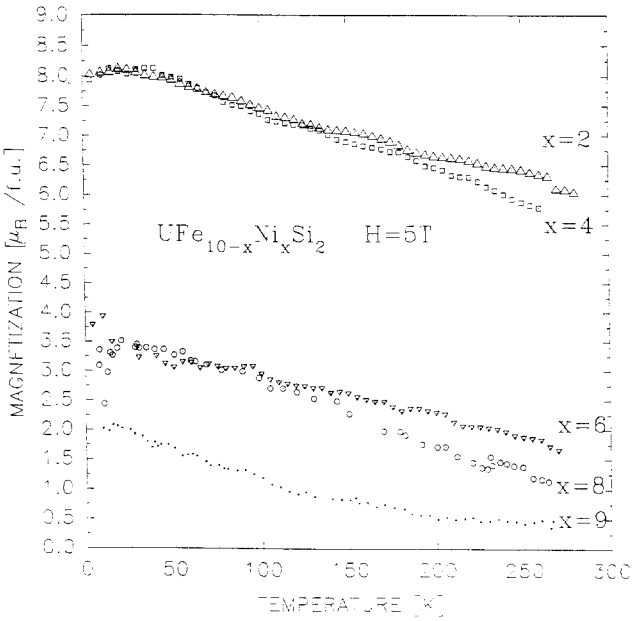

Fig. 2. Magnetization of the $UFe_{10-x}Ni_xSi_2$ alloys vs. temperature measured in a magnetic field of 5 T.

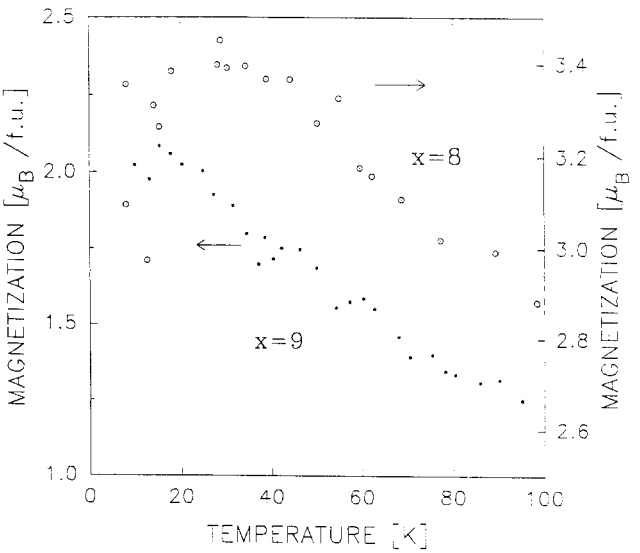

Fig. 3. Low temperature details (below 100 K) of the temperature dependence of magnetization of the $UFe_{10-x}Ni_xSi_2$ alloys with $x=8$ and 9. Note the different magnetization scales for both materials.

non-equivalent crystallographic positions available for the Fe atom makes the SG state highly probable. However, this problem needs further examination.

Figure 4 presents the magnetization of the $UFe_{10-x}Ni_xSi_2$ alloys versus magnetic field strength measured at 4.2 K. One can see that the maximal value of the magnetic moment amounts to about 15 μ_B/f.u. for $x=2$. It is seen that with an increase in the Ni concentration, x, the remanence increases, but unfortunately, at the same time the saturation magnetization and Curie points become lower, reaching for $x=9$ about

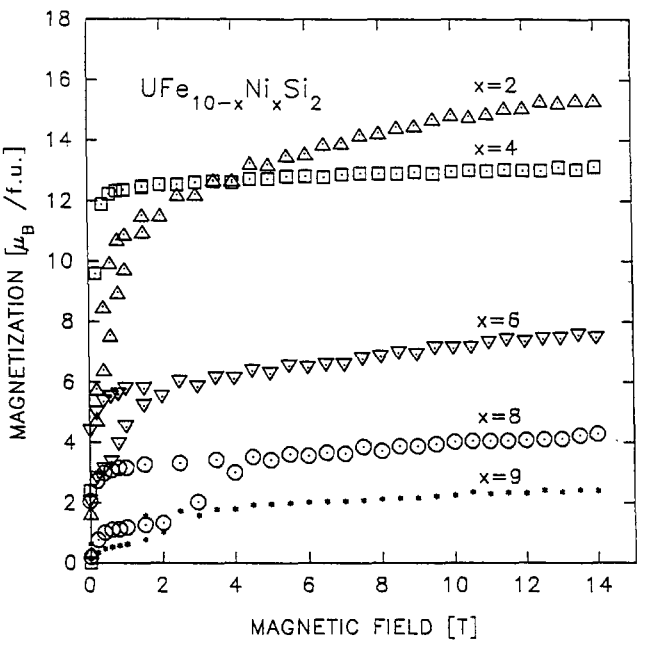

Fig. 4. Magnetization of the $UFe_{10-x}Ni_xSi_2$ alloys vs. magnetic field strength at 4.2 K.

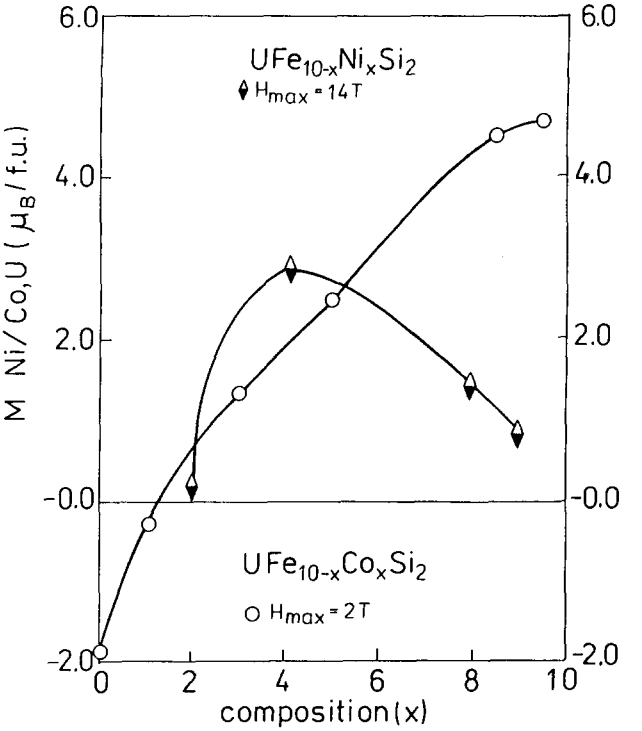

Fig. 5. The total magnetic moment of the U + Ni/Co sublattices vs. Ni/Co concentration, x. Results for $UFe_{10-x}Co_xSi_2$ are evaluated using data from ref. 9.

2 μ_B/f.u. and $T_c = 45$ K, respectively. For the samples with $x = 9$, 8 and 6, the clear deviation from typical behavior for normal ferromagnetic magnetization curves is observed. This shape of magnetization curve for the $ThMn_{12}$-type compounds is usually considered as a potential result of the first order magnetization process (FOMP) and an indication of the f-electron element contribution to the magnetism of the compound. Other indications result from Fig. 5. This figure shows the total moment of the U + Ni/Co sublattices versus concentration of Ni/Co. The total moment of these sublattices has been obtained subtracting the value of the magnetic moment of the Fe sublattice determined for $UFe_{10-x}Ni_xSi_2$ [5] and $UFe_{10-x}Co_xSi_2$ alloys in ^{57}Fe Mössbauer experiments from the value of the magnetic moment of the alloy obtained from magnetometric measurements. As far as the Co alloys are concerned, the (U, Co) magnetic moment increases non-linearly with the cobalt concentration. Such behavior is quite clear because $UCo_{10}Si_2$ is also ferromagnetic below 550 K [10] and an increase in the (U, Co) moment corresponds mainly to the Co moment, although a contribution of the U moment is also possible. The non-linear plot can result from complex substitution of the Fe by the Co atoms and also from the contribution of uranium. The negative value for $x \leqslant 2$ suggests both some contribution of the uranium atom moment (estimated for $U_{1-x}Y_xFe_{10}Si_2$ at $p_U = 0.5$ μ_B [11]) and/or the antiparallel contribution of the Co magnetic moment at 8(f) and 8(j) sites.

For the Ni system, this plot is much more complicated. The value of the (U, Ni) sublattices moment exhibits a pronounced maximum for $x = 4$. Since $UNi_{10}Si_2$ is non-magnetic, one could expect that in the $UFe_{10-x}Ni_xSi_2$ alloys, the magnetic moment of the Ni sublattice is absent and the magnetic moment of the (U, Ni) subsystem is determined by the uranium sublattice exclusively. The present results show that it is not the case, and the presence of the Fe atoms induces a magnetic moment also in the Ni sublattice. The strongest indication of this follows from the value of the magnetic moment for $x = 4$ amounting to about 3 μ_B, which cannot be ascribed exclusively to the uranium sublattice. The magnetic moment for $x = 2$ is close to 0.3 μ_B and is in fair agreement with the previously determined value of 0.5 μ_B for the U sublattice [11]; for this alloy the contribution of the Ni sublattice seems to be non-existing. These results, together with earlier crystallographic and Mössbauer data, suggest a change of band structure for alloys with $x = 2$–4. The value of magnetic moment extrapolated to $x = 10$ did not find any confirmation either in magnetic [1] or NMR experiments [2]; however, the value is very small and can be attributed to experimental uncertainty.

Unfortunately, we cannot separate the contribution of either the U or the Ni sublattices from the present experiments; this can be done only in neutron diffraction experiments. Nevertheless, the present experiment provides a new indication of the U sublattice contribution to the magnetism of $ThMn_{12}$-type uranium intermetallics.

4. Conclusions

The results presented confirm the ferromagnetic character of the alloys investigated below 300 K. Magnetization curves at 4.2 K measured in magnetic fields up to 14 T yield values of saturation moment which in turn, after subtracting the value of the magnetic moment of the Fe sublattice determined from the ^{57}Fe Mössbauer effects, provide a determination of the magnetic moment of the U and Ni sublattices. These results suggest a modification of the band structure of the alloys investigated and an apparent magnetic contribution of the uranium sublattice.

Acknowledgment

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