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Structure, magnetic and electrical properties of the $UMn_{4-x}Fe_xAl_8$ system

W. Suski[a,b,*], K. Wochowski[a], D. Badurski[a], A. Gilewski[b], T. Mydlarz[b]

[a]*Polish Academy of Sciences, W. Trzebiatowski Institute of Low Temperature and Structure Research, P.O. Box 937, 50-950 Wrocław 2, Poland*
[b]*International Laboratory of High Magnetic Fields and Low Temperatures, ul. Gajowicka 95, 53-421 Wrocław, Poland*

Abstract

UMn_4Al_8 is paramagnetic, however, it seems to be on the verge of magnetic ordering. Therefore, it is interesting to check if an admixture of Fe can induce a magnetic ordering in the U and Mn sublattices. The $UMn_{4-x}Fe_xAl_8$ system exists in the whole concentration range $0 \leq x \leq 4$. The lattice parameters follow roughly Vegard's law. Magnetic susceptibility, which is measured at $T=1.8$–300 K in a magnetic field of 0.5 T, fulfils the Curie–Weiss law above 100 K for $x=1$ and 2, and above 150 K for $x=3$; however, the values of the effective magnetic moment and the paramagnetic Curie temperature have only the meaning of the material constants. Below 100 K, the temperature dependence of magnetic susceptibility exhibits anomalies which can be related to magnetic ordering of different sublattices (Fe,U,Mn?). Magnetization measurements at 4.2 K in magnetic fields up to 40 T (pulsed field) show a hysteresis and saturation that indicate a ferro(ferri)magnetic character of the ordering. The electric resistivity curves versus temperature show a shape which is far from a typical metallic behavior. Magnetoresistivity curves are similar to those observed for UFe_4Al_8.

Keywords: Magnetic properties; Electrical properties; Uranium intermetallics

1. Introduction

The $UMn_{4-x}Fe_xAl_8$ system belongs to the extensively investigated $ThMn_{12}$-type compounds, reviewed recently in Ref. [1]. In this type of structure there are four non-equivalent crystallographic positions which frequently make the magnetic and related properties of these materials quite complicated. UMn_4Al_8 is paramagnetic in all the investigated temperature range [2]; however, below 200 K the $\chi(T)$ plot has a diffuse maximum which is absent for $ThMn_4Al_8$. This indication of a possible magnetic ordering in the U-sublattice was not confirmed by a neutron diffraction (ND) experiment [3]. In turn, the magnetic structure of UFe_4Al_8 can be considered ferrimagnetic, as is shown by the ND experiment [4]. The Fe atoms located in the 8f positions of the $ThMn_{12}$ structure are ordered antiferromagnetically in a $+-+-$ sequence propagating along $\langle 110 \rangle$ directions. In zero field, the Fe moments are tilted ~16° towards the direction of the U moment which is ferromagnetically ordered. Both sublattices order at $T_N \sim$ 155 K.

Electrical resistivity, ρ, of UMn_4Al_8 is weakly temperature dependent, however, at $T=200$ K there is a diffuse minimum [5]. For UFe_4Al_8, $\rho(T)$ exhibits a diffuse maximum at about 150 K, with an apparent additional peak at about 160 K, the temperature roughly corresponding to the Néel point [6].

Because UMn_4Al_8 seems to be on the verge of magnetic ordering we have decided to check if an admixture of Fe will induce a magnetic ordering in the $UMn_{4-x}Fe_xAl_8$ alloys.

2. Experimental details

The $UMn_{4-x}Fe_xAl_8$ alloys were prepared by melting the elements in stoichiometric quantities in an arc furnace under a protective argon atmosphere. This procedure was followed by prolonged annealing at about 800°C for 2 weeks. The measurements were performed on a bulk piece of polycrystalline material.

The magnetic susceptibility was investigated in the temperature range 1.8–300 K in a magnetic field of 0.5 T, whereas electrical resistivity was investigated at $T=4.2$–300 K. Magnetization at 4.2 K was measured up to 14 T in a steady field, and up to 38 T in a pulsed field. The magnetoresistivity was determined as the difference be-

*Corresponding author. Tel.: +48 71 3435021; fax: +48 71 441029; e-mail: suski@highscreen.int.pan.wroc.pl

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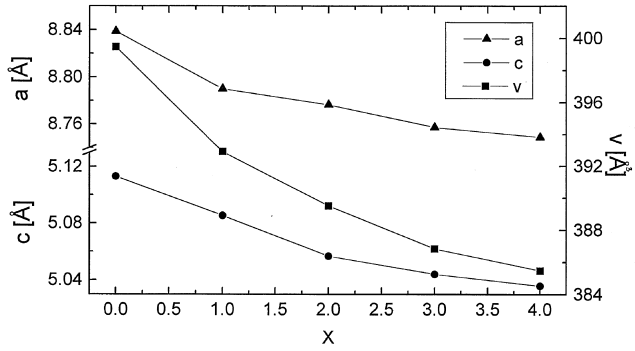

Fig. 1. Lattice parameters, a (triangles), c (circles), and cell volume, V (squares), versus Fe concentration, x, at room temperature.

tween the 3 T and 0 T resistivity at T=4.2–110 K and at T=4.2 K up to 8 T. All measurements were carried out using the standard equipment in our laboratories.

3. Results and discussion

The $UMn_{4-x}Fe_xAl_8$ system exists as a single phase in the whole concentration range. As is seen from Fig. 1, both lattice parameters follow roughly Vegard's law. For comparison, earlier data for UMn_4Al_8 and UFe_4Al_8 (see Ref. [1]) are also included. Such a smooth change of lattice parameters and volume seems to suggest that the mutual substitution of Mn by Fe occurs in the frame of the same crystallographic position. However, this is not obvious because in UFe_4Al_8 [4] the Fe atoms are located in the 8f position, whereas in UMn_4Al_8 [3,7] the Mn atoms are distributed in the 8f and 8j sites, sharing these locations with the Al atoms. Thus, a final conclusion can only be drawn as soon as the ^{57}Fe Mössbauer experiment, which is now in progress in our Laboratory, has been finished.

Results of magnetic measurements are presented in Figs. 2–4. The reciprocal magnetic susceptibility presented in Fig. 2 follows the Curie–Weiss law for $UMn_{4-x}Fe_xAl_8$

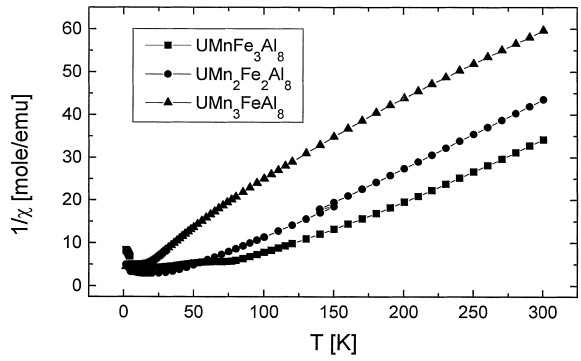

Fig. 2. Reciprocal magnetic susceptibility, χ^{-1}, versus temperature measured in magnetic field of 0.5 T for $UMnFe_3Al_8$ (squares), $UMn_2Fe_2Al_8$ (circles) and UMn_3FeAl_8 (triangles).

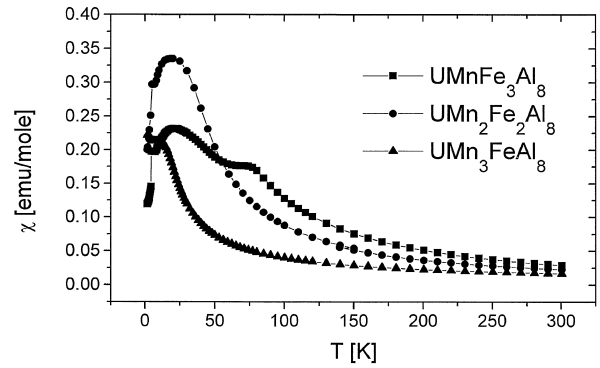

Fig. 3. Magnetic susceptibility, χ, versus temperature measured in magnetic field of 0.5 T for $UMnFe_3Al_8$ (squares), $UMn_2Fe_2Al_8$ (circles) and UMn_3FeAl_8 (triangles).

alloys with x=1 and 2 at $T\geq$100 K and for x=3 at $T\geq$150 K. The values of magnetic moment p_{eff}=6.79, 7.01 and 7.53 μ_B/f.u. and the Weiss constants θ=−49, 32 and 60 K for x=1, 2 and 3, respectively, have only the meaning of material constants. The three components, U, Fe and Mn, can contribute to susceptibility and their separation is impossible at present. The $\chi(T)$ plots presented in Fig. 3 show some anomalies at low temperatures; however, none of the samples exhibit an anomaly at ~200 K, as was observed for UMn_4Al_8 [2]. The sample with x=2 is most similar to the polycrystalline sample of UFe_4Al_8 [8] which exhibits a maximum at 50 K and a small jump at ~30 K; however, both anomalies are shifted to slightly lower temperature, and that at lower temperature has a different shape. It should be noted that the Th compound does not show the lower temperature anomaly [8]. It is also worthwhile to note that the Néel point at ~155 K of UFe_4Al_8 is not seen in magnetization measurements of the latter compound or in the currently reported solid solu-

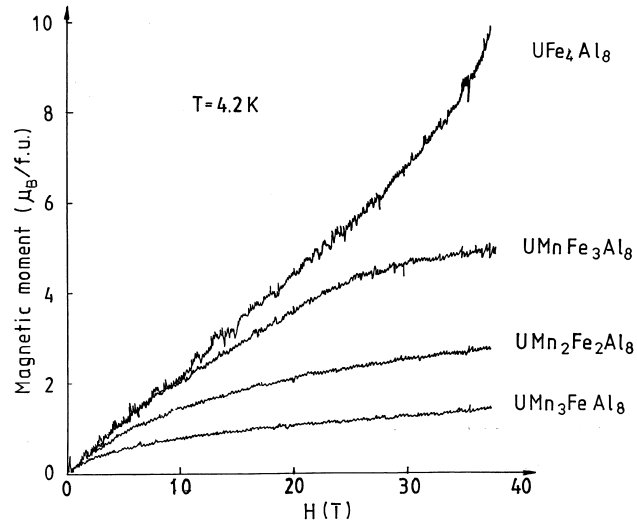

Fig. 4. Magnetization of $UMn_{4-x}Fe_xAl_8$ alloys and UFe_4Al_8 versus magnetic field at T=4.2 K.

tions. For the sample with $x=3$, the $\chi(T)$ plot exhibits a maximum corresponding to the one mentioned above, but there is a kind of local maximum at about 80–90 K and a small jump followed by temperature-independent susceptibility (TIS) below ~30 K. Finally, for the sample with $x=1$ there is also TIS at low temperature with a very diffuse maximum at about 30–35 K. It is clear that the presence of three 'magnetic' species in these alloys can be the reason for these anomalies. However, without additional experiments any speculation does not make sense. Fig. 4 presents magnetization curves for the $UMn_{4-x}Fe_xAl_8$ alloys and UFe_4Al_8. All solid solutions show a kind of saturation and small hysteresis (not shown for sake of brevity). 'Saturation' moments are relatively low in comparison with the effective moment. The most interesting result, however, is the increase of magnetization of UFe_4Al_8 in the magnetic field above 30 T. Andreev et al. [9] have measured the magnetization at 4.2 K of the UFe_xAl_{12-x} system in magnetic fields up to 35 T on powder particles free to be oriented by the applied field and on particles fixed by frozen alcohol. Their samples exhibit linear increase of magnetization with increasing field. We do not know the reason for this discrepancy. It is just on the edge of our experimental facilities, and an extension of these measurements to still higher fields would be extremely interesting. This seems to be an indication of a metamagnetic transition. Unfortunately, the polycrystalline form of the sample does not allow us to compare the present results with those presented in Ref. [9] as well as in Ref. [10].

The results of transport measurements are shown in Figs. 5–7. Electrical resistivity, ρ, versus temperature (Fig. 5) does not exhibit an anomaly for any alloy, corresponding to that observed at about 200 K for UMn_4Al_8 [5], and at about 120 K for UFe_4Al_8 [6]. The sample with $x=1$ is the most reminiscent of metallic samples, except for a very small minimum in $\rho(T)$ at ~20 K. The $\rho(T)$ for the sample with $x=3$ has a similar shape to semiconductors; however, recent works on UNiSn or CeNiSn show that such a shape can result from band effects in metallic material. Finally, for the $x=2$ sample, a clear minimum in $\rho(T)$ is seen at T~60–70 K, and the concept of the Kondo effect in this magnetically concentrated system seems to be excluded. The behavior of the $x=1$ sample is similar to that reported for UFeGe [11] and $U_{1-x}Np_xRu_2Si_2$ with $0.3 \leq x \leq 0.7$ [12]. In turn, the behavior of the $x=3$ sample corresponds to the temperature dependence of the electrical resistivity of $U_{0.75}Lu_{0.25}PdSn$ [13]. In all these cases, however, no explanation is given. Unfortunately, the polycrystalline form of the samples with cracks and imperfections prevents us from proposing any quantitative conclusion.

The temperature dependence of magnetoresistivity, and the field dependence of the magnetoresistivity, measured on polycrystalline samples are presented in Figs. 6 and 7. The strong scatter of the data makes it difficult to interpret and compare with the results reported on single-crystal UFe_4Al_8 [6,10]. There are some general similarities, i.e. magnetoresistance is negative, and decreases with an increase of magnetic field, at least at $T=4.2$ K. The absolute value is not very impressive and thus does not correspond to the enthusiastic title of [10].

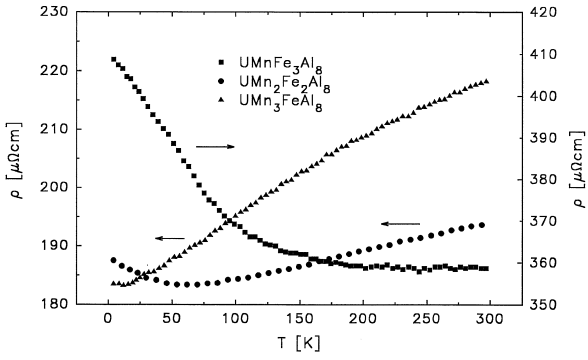

Fig. 5. Electrical resistivity, ρ, versus temperature for $UMnFe_3Al_8$ (squares), $UMn_2Fe_2Al_8$ (circles) and UMn_3FeAl_8 (triangles).

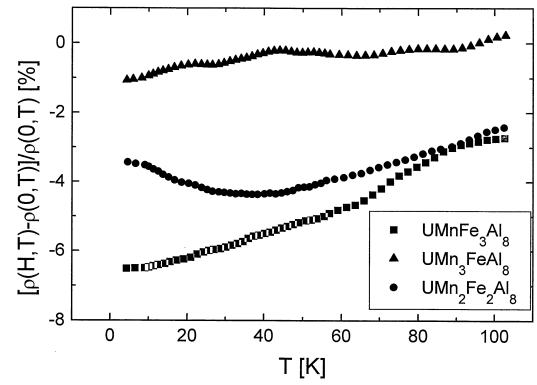

Fig. 6. Magnetoresistvity, $[\rho(B)-\rho(O)]/\rho(O)$, versus temperature measured in magnetic fields of 0 and 3 T for $UMnFe_3Al_8$ (squares), $UMn_2Fe_2Al_8$ (circles) and UMn_3FeAl_8 (triangles).

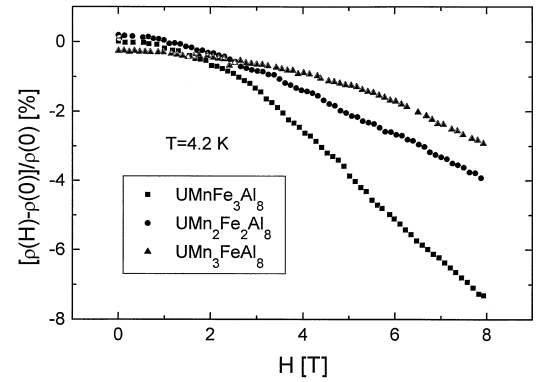

Fig. 7. Magnetoresistivity, $[\rho(B)-\rho(O)]/\rho(O)$, versus magnetic field, at $T=4.2$ K, for $UMnFe_3Al_8$ (squares), $UMn_2Fe_2Al_8$ (circles) and UMn_3FeAl_8 (triangles).

4. Conclusions

In the present paper we report on the structure, and magnetic and electrical properties of the $UMn_{4-x}Fe_xAl_8$ alloys. These alloys form single phases with lattice parameters that follow Vegard's law. At low temperature, these materials exhibit some anomalies in the temperature dependence of magnetic susceptibility. There is some indication of a ferro(ferri)magnetic character of these compounds; however, it is difficult to give any more precise explanation of their magnetism. Similar problems follow from electrical and magnetoelectrical measurements which are still enhanced by the polycrystalline form of the samples.

Summarizing, one can state that an addition of one atom of Fe introduces magnetic ordering to these alloys; however, this quantity of Fe is quite large to draw any conclusion concerning Mn and U sublattices. For a better understanding of the present results, further experiments are necessary on more diluted Fe concentrations and on the single-crystal species, if possible.

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