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Magnetic properties of Heusler alloys $Ru_{2-x}Fe_xCrSi$

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Abstract

We report on the structural and magnetic properties of newly synthesized Heusler alloys, $Ru_{2-x}Fe_xCrSi$, which have quite recently been shown to be candidates for ferromagnetic metals with high spin polarization from band structure calculations. Polycrystalline samples of Heusler alloys $Ru_{2-x}Fe_xCrSi$ were prepared for $0.5 \leqslant x \leqslant 1.8$. They were found to have $L2_1$ structures for $0.5 \leqslant x \leqslant 1.5$ and B2 for $x = 1.8$. Magnetic measurements showed that they are ferromagnets. The Curie temperature for $x = 1.0$ was found to be 370 K. The Curie temperature tends to increase with increasing Fe concentration x. The saturation magnetic moment increases almost linearly as x increases. For higher Fe concentration the saturation magnetic moment is close to 2 μ_B per formula unit, which is theoretically expected.

1. Introduction

Heusler alloys have been studied for a long time because of their unique magnetism. Recently, Heusler alloys have attracted renewed interest because they have been revealed to be promising materials for future applications. Among them are half metals and ferromagnetic shape memory alloys. A half metal(-lic ferromagnet) is a ferromagnetic metal where the minority band is semiconducting with a gap at the Fermi level. This means that the conduction electrons are 100% spin polarized. Half metals are expected to play an important role in devices in future magneto electronics, so-called spintronics. The half metal was predicted for the first time in NiMnSb [1], which belongs to a class of materials referred to as half-Heusler alloys, the structure of which relates intimately to that of usual (full-)Heusler alloys. Heusler alloys are of the type X_2YZ, where X, Y are transition metals and Z is an sp element. They crystallize in the cubic $L2_1$ type structure which consists of four penetrating fcc sublattices. In Heusler alloys, Ishida *et al* predicted from first principles calculations that Co_2MnZ, where Z is Si or Ge, is a half metal [2]. In a recent upsurge of interest in spintronics in particular, many works aiming at finding promising materials in Heusler alloys have been performed. Experimentally, however, the realization of half metallicity has not been satisfactory. It has been revealed both theoretically and experimentally that disorders, defects, and surfaces harm

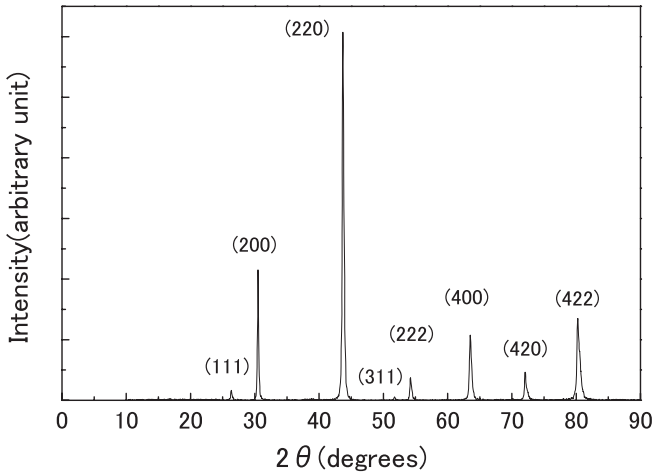

Figure 1. X-ray diffraction pattern with Cu Kα of $Ru_{1.5}Fe_{0.5}CrSi$.

the half metallicity [3–6]. From the standpoint of applications, ferromagnetic metals with high spin polarization which are insensitive to such crystalline disorders, even if they are not complete half metals, are highly valuable. Recently the Heusler-type alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ exhibited considerably large magnetoresistance in bulk form [7] and relatively large tunnelling magnetoresistance [8]. In the reports the structure is suggested to be disordered B2 structures. These results indicate that the spin polarization of $Co_2Cr_{1-y}Fe_yAl$ might not be easily affected by disorder. Atomic disorder effects on $Co_2Cr_{1-y}Fe_yAl$ have also been analysed by theoretical calculations [9].

Quite recently, from first principles band structure calculations the $Ru_{2-x}Fe_xCrSi$ series has been predicted as such materials in Heusler alloys [10]. It is shown that they are complete or nearly half metals and have a large density of states in the majority spin states at the Fermi level, which will lead to robustness against disorder. To our knowledge the Heusler alloys $Ru_{2-x}Fe_xCrSi$ have not been synthesized and their properties have not been reported so far. Motivated by the theoretical calculations we tried to synthesize these materials. In this paper we report the structural and magnetic properties of $Ru_{2-x}Fe_xCrSi$.

2. Experimental details

Polycrystalline samples were prepared by arc-melting high-purity constituent elements under high-purity argon atmosphere. In order to obtain homogeneity, the samples were remelted several times. Although the effects of the annealing of the sample were examined, no noticeable effects were observed so far. Therefore, we report here on arc-melted samples without annealing. The crystal structures were investigated by x-ray powder diffraction measurements. The magnetization was measured by a commercial superconducting quantum interference device (SQUID) magnetometer.

3. Results

We prepared polycrystalline samples of $Ru_{2-x}Fe_xCrSi$, with Fe concentration x being 0.5, 1, 1.5, 1.7, and 1.8. Figure 1 shows the results of x-ray diffraction measurements of $Ru_{1.5}Fe_{0.5}CrSi$. As shown in the figure, the sample was found to be a single phase with

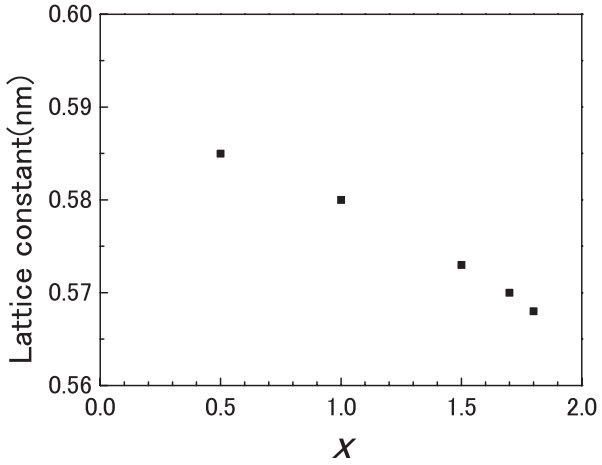

Figure 2. Lattice constant a of $Ru_{2-x}Fe_xCrSi$ as a function of x.

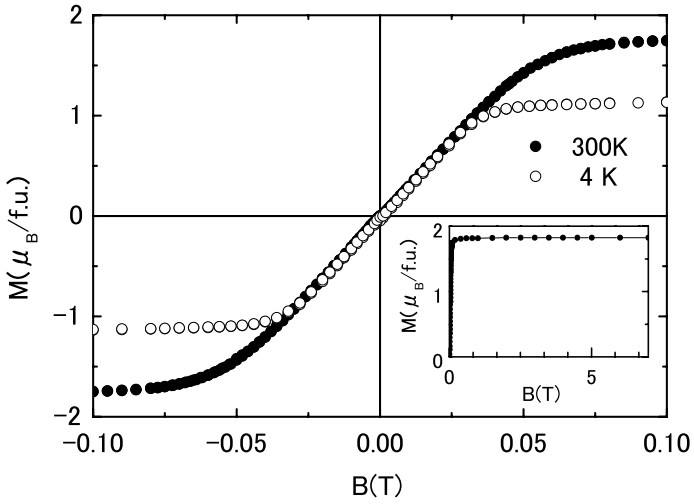

Figure 3. Magnetization curves of $Ru_{0.5}Fe_{1.5}CrSi$ at 4 and 300 K. The data taken in both increasing and decreasing field are shown. At both temperatures hysteresis is not visible. The inset shows the magnetization up to 7 T at 4 K.

the cubic $L2_1$ type structure. For $0.5 \leqslant x \leqslant 1.5$, single phase samples with the $L2_1$ type structure were obtained. The x-ray diffraction intensity ratio I_{111}/I_{220}, where I_{111} reflects the order of Y and Z atoms, is nearly constant for $x \leqslant 1.5$ and decreases fairly abruptly above $x \sim 1.5$. In the sample with $x = 1.8$, the 111 peak is no longer found and the sample is shown to crystallize in B2 structure. So far we did not succeed in synthesizing single phase samples of the series end materials with $x = 0$ and 2. For the sample with $x = 0$ the majority part was found to be the $L2_1$ phase; however, slight traces of impurity phases were detected in the diffraction pattern. The lattice constants a of $Ru_{2-x}Fe_xCrSi$ as a function of x are shown in figure 2. The lattice constant of the sample with $x = 0.5$ is 0.585 nm. It varies almost linearly with x. This monotonic tendency suggests that Fe is homogeneously mixed in the samples.

Figure 3 shows magnetization curves of $Ru_{0.5}Fe_{1.5}CrSi$ at 300 and 4 K without the correction for demagnetization. It exhibits ferromagnetism. The magnetization curves with

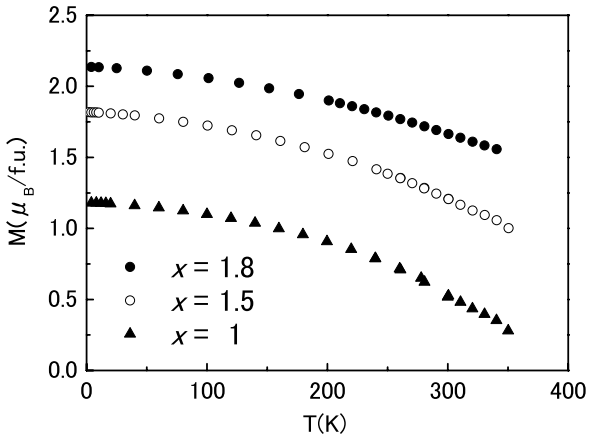

Figure 4. Temperature dependence of magnetization at 1 T of $Ru_{2-x}Fe_xCrSi$ with $x = 1$, 1.5, and 1.8.

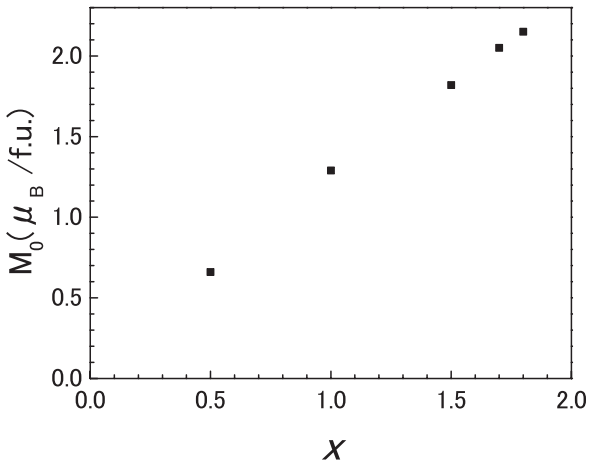

Figure 5. Saturation magnetic moment at 0 K, M_0, of $Ru_{2-x}Fe_xCrSi$ as a function of x.

increasing field and decreasing field coincide with each other. The coercive force is less than 1 mT in experimental resolution. Hysteresis is not observed, demonstrating that it is a soft ferromagnetic material. The magnetization is almost constant above ∼0.09 T at 4 K. By considering the demagnetizing effect, though precise correction is difficult, the saturation of the magnetization should be reached above ∼0.04 T. As shown in the inset of figure 3, above that field the magnetization is almost constant up to 7 T.

Figure 4 shows the temperature dependence of magnetization of $Ru_{2-x}Fe_xCrSi$ ($x = 1$, 1.5, 1.8) in a magnetic field of 1 T below 350 K. The curves are regarded as showing the temperature dependence of the saturation magnetization. As seen in figures 3 and 4, the saturation magnetization extrapolated to 0 K, M_0, for $x = 1.5$ is found to be 1.83 μ_B per formula unit (equivalent to 1.02×10^4 emu per mol formula unit). In figure 5 the values of M_0 per formula unit are plotted as a function of x. They increase almost linearly with increasing x up to $x = 1.8$. For $x = 1.8$ the structure changes to B2 type, whereas it seems that the tendency of the increase in the saturation moment with x is not affected. Comparisons should be made with the reported properties of similar Heusler alloys with Fe and Cr, $Fe_2T_{1-z}Cr_zSi$, where T is

V or Mn [11]. For Cr-rich samples it was found that they exhibit ferromagnetism. Although a sample of Fe_2CrSi was not obtained, extrapolation of the saturation magnetic moment results in 2.2 μ_B per formula unit, which agrees well with the present result. From the extrapolation of the magnetization data, the Curie temperature, T_C, for $x = 1$ is properly determined to be 370 K, and for $x = 0.5$ T_C it is 190 K. Although there is substantial uncertainty due to the lack of the data in high temperature region, T_C is roughly estimated from the extrapolation to be ∼460 K for $x = 1.5$ and ∼580 K for $x = 1.8$, respectively. The estimated T_C also increases with x as M_0 increases. For $x = 0.5$ in the limited range though, the temperature dependence of the magnetization obeys the Curie–Weiss law above T_C. The paramagnetic moment M_p is evaluated from the effective magnetic moment M_{eff} determined from the Curie constant, where $M_{eff} = \sqrt{M_p(M_p + 2)}$. M_p for $x = 0.5$ is estimated to be 1.9 μ_B per formula unit. The ratio M_p/M_0 is markedly larger than 1, which indicates that the ferromagnetism may have somewhat itinerant character, in particular for Ru-rich samples. In the sample with $x = 0$, which contains a small quantity of impurity phases, no indication of ferromagnetism was found down to 2 K.

4. Discussion

Motivated by the theoretical calculations, we have succeeded in preparing Heusler alloys $Ru_{2-x}Fe_xCrSi$ and have found that they are ferromagnets. However, we do not have any direct evidence of the spin polarization. At present we can compare with the prediction of the theory. The saturation magnetic moment has been calculated theoretically. The theory predicts that it is 2 μ_B, which is an integral multiple of μ_B as usual in half metals, and that it is almost independent of Fe concentration [10]. The present results, however, show that the saturation moment depends on Fe concentration x and varies almost linearly with x. Although for lower Fe concentration the saturation moment is rather smaller than 2 μ_B, for $1.5 \leqslant x \leqslant 1.8$ the saturation moment becomes close to the predicted value 2 μ_B. First, one possible reason for this discrepancy is that the exchange of the site occupation may influence the magnetic moment. While the theory claims that the spin polarization of $Ru_{2-x}Fe_xCrSi$ is insensitive to disorder, a more detailed calculation reveals the stability of spin polarization against several kinds of atomic site disorders [10]. It has been found that some types of disorder, if realized, degrade the spin polarization, but these kinds of disorder are often energetically unstable, as is demonstrated in $Co_2Cr_{1-y}Fe_yAl$ [9]. Since the structure for $x = 1.8$ is B2 type, the magnetic properties do not appear to be so much influenced by the disorder between Y and Z sites. It should be noticed, however, that from the x-ray diffraction measurements, we did not find any evidence that the disorder in $L2_1$ structure is enhanced for lower Fe concentration. At present the exact site occupancy for each element is not clear, and further comparison is left for the future. Considering that disorder effects are more or less inevitable in solid solutions like these materials, for higher x the magnitude of the saturation magnetic moment agrees well with the theoretical prediction. Secondly, the theoretical calculation shows that in Ru_2CrSi an antiferromagnetic state is preferable to the ferromagnetic state [10]. Actually the present results show that with decreasing x towards Ru-rich concentration, T_C decreases, and for $x = 0$ the ferromagnetic order disappears. This suggests that the ferromagnetic interaction becomes weaker as the concentration of Ru increases. It may be inferred that in $Ru_{2-x}Fe_xCrSi$, particularly when rich in Ru, complex magnetic interactions may exist, and these would cause the reduction of the saturation magnetic moment. A detailed investigation of the magnetic properties in the Ru-rich region is now in progress.

In conclusion, we have succeeded in preparing Heusler alloys $Ru_{2-x}Fe_xCrSi$. For higher Fe concentration ferromagnetism with relatively high Curie temperature is found, and the

saturation moment is close to the value predicted by the band structure calculations. For $x = 1.8$ the structure becomes more disordered B2 where Y and Z atoms are randomly distributed, while the magnetic properties do not seem to be changed so much. This suggests that this kind of disorder does not affect the magnetism significantly as in $Co_2Cr_{1-y}Fe_yAl$. Although the reason for the disagreement of the saturation magnetic moment for smaller x with the theory is not conclusive, for larger x the obtained material appears to have properties very close to those theoretically predicted. Namely, $Ru_{2-x}Fe_xCrSi$ rich in Fe is a very promising candidate for a ferromagnet which has high spin polarization with large density of states for the majority spins at the Fermi level. In order to clarify the spin polarization of these materials, more detailed comparisons between experimental studies and theories are required. Direct studies detecting the spin polarization are desirable and are under consideration.

Acknowledgments

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