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Effect of Fe substitution on magnetic properties of antiferromagnetic Heusler alloy Ru_2MnGe

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ABSTRACT

The present study reveals that the correlation between the magnetic properties with composition x in the polycrystalline $Ru_2Mn_{1-x}Fe_xGe$ system. Hard ferromagnetic properties appeared at the intermediate composition between antiferromagnetic Ru_2MnGe with soft ferromagnetic Ru_2FeGe. $Ru_2Mn_{0.6}Fe_{0.4}Ge$ shows an anisotropic and negative magnetoresistance (MR) effect more than 8% although the both end materials are small MR effect less than 1%. These experimental results correlate the variation in MR ratio with that of coercive field as a function of x. Moreover, the correlation is shown at the boundary of magnetism between antiferromagnetic and ferromagnetic. Therefore, the present experimental results imply that the coexistence of antiferromagnetic and soft ferromagnetic leads to induce the MR effect in this system.

1. Introduction

Heusler alloy systems have attracted renewed attention because the half-metallic ferromagnetic Heusler alloy [1–3] can be used as a spin-filter material for TMR devices on the field of the development of tunnel magnetoresistance (TMR) device. Therefore, it is very important to found novel half metallic materials for the further development of spintronic devices. For example, a magnetic tunneling junction (MTJ) with a thin film electrode of full Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ shows large TMR at room temperature [4–5]. For $Co_2Cr_{0.6}Fe_{0.4}Al$ [6], it was reported to show a large magnetoresistive effect under a small magnetic field at room temperature [6–10]. The experimental and theoretical studies on the electronic structure of the $Co_2Cr_{1-x}Fe_xAl$ system [11,12] revealed that the full Heusler alloy $Co_2Cr_{0.6}Fe_{0.4}Al$ is a half metallic ferromagnet with high Curie temperature.

Many studies have been performed for the 3*d*-, 4*d*- or 5*d*-transition metal based systems to discover new functional full Heusler alloy systems. It is generally believed that the electron–electron Coulomb repulsion strength U among the d electrons decreases when base element changes from 3*d* to 4*d*/5*d* elements due to the difference in the spatial expanse of *d*-orbitals. Moreover, the difference between the 3*d* and 4*d*/5*d* electrons seems to affect on the interplay between the chemical composition and electron correlation. Therefore, drastic changes in the magnetic and transport properties are expected for 4*d*- or 5*d*-transition metal based Heusler alloys in comparison with the 3*d*-transition metal based alloys.

Among the 4*d*- or 5*d*-transition metal based Heusler alloys it was reported that Ru_2FeZ (Z = Ge, Sn) [13,14] shows ferromagnetism, while Ru_2FeSi [15–17] and Ru_2MnZ (Z = Sn, Si, Sb, Ge) [13] are antiferromagnetic. Moreover, the existence of a polarized conduction electron band has been reported for Ru_2FeGe [14]. Therefore, it is interesting to study the influence of the crossover from antiferromagnetism to ferromagnetism on the transport properties in the $Ru_2Mn_{1-x}Fe_xGe$ system. In this paper, we synthesized the Ru based Heusler alloy system $Ru_2Mn_{1-x}Fe_xGe$, and investigated the

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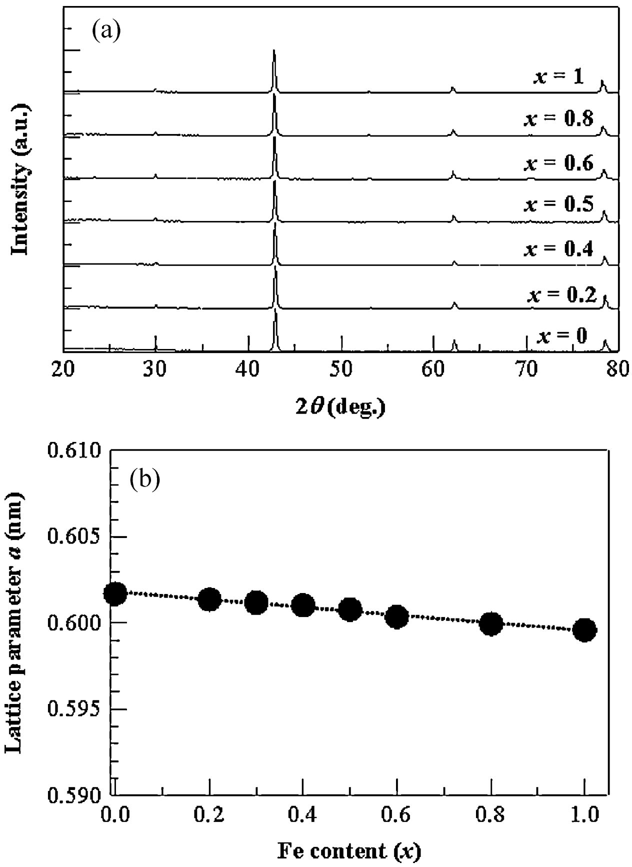

Fig. 1. (a) Powder XRD profiles of $Ru_2Mn_{1-x}Fe_xGe$ and (b) refined cubic lattice parameter a of $Ru_2Mn_{1-x}Fe_xGe$ as a function of Fe content x.

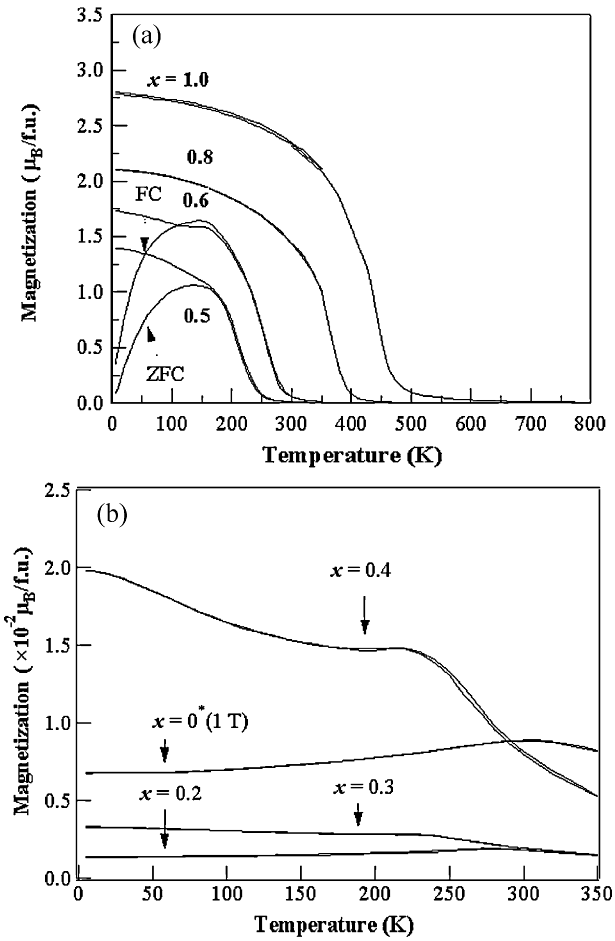

Fig. 2. (a) Temperature dependence of the magnetization of $Ru_2Mn_{1-x}Fe_xGe$ for $x < 0.5$ and (b) that for $x \geq 0.5$.

crystallography, magnetism, and transport properties to be discuss the interplay between magnetism and transport properties.

2. Experiment

Polycrystalline $Ru_2Mn_{1-x}Fe_xGe$ samples were prepared by arc melting. Stoichiometric amounts of the elemental constituents were melted in a zirconium-gettered Ar atmosphere. The polycrystalline ingots were sealed into an evacuated quartz ampoule. The ampoule was heated to 1173 K and after being kept for 72 h, the ampoule was quenched into water. The crystallographic properties were characterized by the X-ray powder diffraction using Cu-Kα radiation. Lattice constants were obtained by the Rietveld method. The magnetic properties were characterized by SQUID magnetometer and VSM. Resistivity measurements were performed by the standard four-probe method under an applied field up to 5 T. The magnetoresistance ratio (MR ratio) is defined as $\Delta\rho^{MR}/\rho = [\rho(B) - \rho(0)]/\rho(0)$.

3. Results and discussion

3.1. Structural properties

The XRD profiles for $Ru_2Mn_{1-x}Fe_xGe$ samples in Fig. 1(a) show that all samples are single phase with $Fm3m$ symmetry structure at room temperature. All samples shows the superlattice reflection from the (1 1 0) plane; however, no reflection of the (1 1 1) plane is shown. The analysis of present XRD profiles indicates that the polycrystalline $Ru_2Mn_{1-x}Fe_xGe$ have the B_2-type ordering states. Lattice parameters were obtained by assuming the space group $Fm3m$ (No. 225) [14] with the S ($=R_{wp}/R_e$) value for all samples to be less than 1.3. The refinement for Ru_2FeGe shows that Fe and Ge atoms are ordered on each crystallographic position with a disorder level of approximately 2%. That for $Ru_2Mn_{0.5}V_{0.5}Ge$ indicates that Mn/Fe atoms mainly are same crystallographic position and that they are partially exchanged to Ge atoms with a disorder level of approximately 5%. The lattice parameter a for Fe-content dependence as shown in Fig. 1(b) decreases monotonically from 0.602 nm for Ru_2MnGe to 0.600 nm for Ru_2FeGe with the 0.284×10^{-4} nm for the error of lattice constants. These results additionally indicate that Fe atom has been substituted mainly to Mn atom.

A recent synchrotron-based XRD study for $Co_2Mn_{1-x}Ti_xSn$ system by Graf et al. [18] suggests that even if the lattice constant change monotonically as function of its content, a phase separation may take place and then, the atom are not uniformly distributed on the crystallographic positions, however, our experimental result shows no peak separation at the (2 2 0) refraction. Then, there is no phase separation on the $Ru_2Mn_{1-x}Fe_xGe$ system.

3.2. Magnetic properties

Temperature dependence of the magnetization for $Ru_2Mn_{1-x}Fe_xGe$ is shown in Fig. 2(a) for the sample of $x = 0$–0.4 and in Fig. 2(b) for that of $x = 0.5$–1, respectively. The samples of $x = 0$, 0.2, and 0.3 show the typical antiferromagnetic temperature dependence, whereas the samples of $x = 0.5$–1 show ferromagnetic behavior. In particular, the Fe rich samples ($x = 0.8$ and 1) show a typical ferromagnetic order. The samples of $x = 0.5$ and 0.6 show a large thermal hysteresis between the zero-field cooled and the field cooled curves. The sample of $x = 0.4$ shows a both antiferromagnetic and ferromagnetic contribution. The temperature dependence of the magnetization indicates that the samples with $x = 0.4$–0.6 has a both antiferromagnetic and ferromagnetic contributions.