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Crystal Distortion and Magnetic Structure of γ-MnT(T = Rh, Ru) Alloys

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We have made X-ray and neutron diffraction experiments and magnetic susceptibility measurements for γ-MnRh alloys and γ-MnRu alloys with a small amount of Cu. The γ-Mn alloy containing 10 at % Rh at 10 K shows an orthorhombic structure with a = 3.806, b = 3.748 and c = 3.671 Å and a non-collinear antiferromagnetic structure with μ_a = 0, μ_b = 1.24 and μ_c = 2.04 μ_B/Mn atom. The crystal transition occurs from the orthorhombic structure to the tetragonal structure with $c/a < 1$ at T_o (= 520 K) and finally to the cubic structure at T_t (= 570 K), which is nearly equal to the Néel temperature T_N. The γ-Mn alloy with 14 at % Ru and 5 at % Cu at 10 K has the similar structure with a = 3.794, b = 3.750 and c = 3.677 Å, and μ_a = 0, μ_b = 1.22 and μ_c = 2.47 μ_B/ Mn atom. The crystal transition temperatures and the Néel temperature are as follows: T_o = 380 K, T_t = 500 K and T_N = 520 K.

KEYWORDS: crystal distortion, antiferromagnetism, neutron diffraction, MnRh, MnRu(Cu)

§1. Introduction

As is well known, most Mn-rich γ-Mn alloys undergo a crystal distortion from the face centered cubic structure [c] to a face centered tetragonal (fct) structure with $c/a < 1$ [t_1] below the Néel temperature.[1)] In some cases a distortion to a fct with $c/a > 1$ [t_2] is observed.[2)] In the γ-MnNi alloys, there is also a face centered orthorhombic (fco) structure [o] in addition to [t_1] and [t_2].[3)]

Jo et al have proposed a theoretical phase diagram for the γ-Mn alloys on the basis of Landau expansion of free energy, suggesting the existence of [o] in a region between [t_1] and [t_2].[4)] They have also suggested some possible magnetic structures; the a, b and c-axis components of the magnetic moment, μ_a, μ_b and μ_c in [o] with $a > b > c$, are as follows: ${\mu_c}^2 > {\mu_b}^2 > {\mu_a}^2 > 0$.

Recently, the present authors have found that the [t_2] and [o] regions exist in the γ-MnGa alloys, and determined a non-collinear magnetic structure with 4 sublattices for [o].[5)] The magnetic structures of the γ-MnNi alloys have been investigated by G. T. Etheridge *et al.*[6)] More recently, we have made determination of the magnetic structure of [o] in the γ-MnAu alloy.[7)] In these results, the a, b and c-axis components of the magnetic moments in [o] with $a > b > c$ are as follows: ${\mu_c}^2 > {\mu_b}^2 > {\mu_a}^2 = 0$. This magnetic structure is consistent with Jo's theory.

One of the present authors [T. H] and his collaborators have reported the existence of [o] in γ-MnRh alloys and presented a phase diagram.[8)] The present authors have also found [o] in γ-MnRu alloys with a small amount of Cu. In the present paper, the results of magnetization measurements and X-ray and neutron diffraction experiments for these alloys are reported.

§2. Sample Preparation and Experimental Procedure

Alloys were prepared by melting electrolytic Mn (3N purity), and Rh or Ru (and Cu) using an argon arc furnace. The ingot was sealed in an evacuated quartz tube with argon gas of 2x10^4 Pa, annealed at 1000 °C for more than 2 days, and quenched in water. The powder sample for X-ray and neutron diffraction experiments was prepared by filing the ingot. In order to remove the stress by filing, the powder sample was re-annealed for 2 hours at 1000 °C, and quenched in water. Magnetizations were measured by the Faraday method using a magnetic balance in an electromagnet. An Fe target was used for the X-ray diffraction. Temperature dependence of the lattice constants between 15 K and 530 K was determined using a cryogenic camera for the X-ray diffraction. Neutron diffraction experiments were made by using the HRPD diffractometer (wave length λ =1.823 or 2.300 Å) and the TAS 2 (λ = 2.360 Å) installed at JRR-3M reactor in JAERI.

§3. Experimental Results and Discussion

3.1 γ-MnRh alloys

The temperature dependence of the lattice constants a, b and c of [o] was determined by X-ray diffraction experiments using the 200, 020 and 002 reflections. Fig. 1 shows the results for the γ-MnRh alloys with 10 at % Rh (γ-$Mn_{0.90}Rh_{0.10}$). The alloy shows [o] at lower temperature. This structure is transformed to [t_1] at T_o (= 520 K). Unfortunately, a transition temperature T_t from [t_1] to [c] is higher than the device limited temperature (= 530 K). We determined the Néel temperature T_N from the temperature dependence of magnetic susceptibility

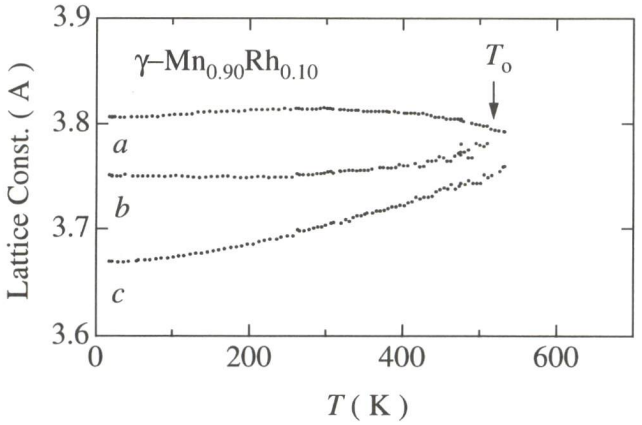

Fig. 1. Temperature dependence of lattice constants for γ-$Mn_{0.90}Rh_{0.10}$.

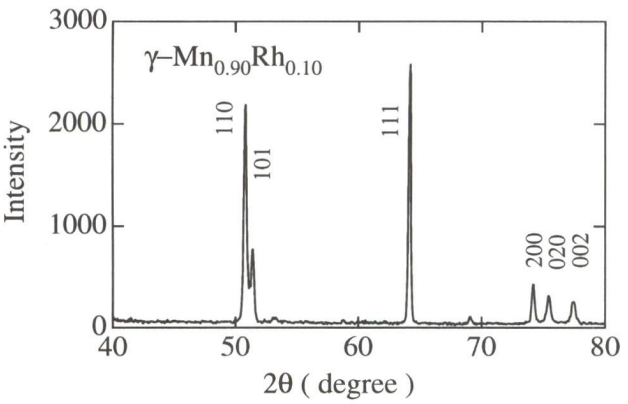

Fig. 2. Neutron diffraction pattern for γ-$Mn_{0.90}Rh_{0.10}$ at 10 K.

as T_N = 568 K.

Figure 2 shows a neutron diffraction pattern for γ-$Mn_{0.90}Rh_{0.10}$ at 10 K obtained by HRPD (λ = 2.300 Å). The pattern is explained by assuming a = 3.806, b = 3.748 and c = 3.671 Å and a non-collinear antiferromagnetic structure similar to that of the γ-MnGa and γ-MnAu alloys. The components of the magnetic moment were determined using the intensities of magnetic reflections 110 and 101 and some nuclear reflections 111 and 200 etc: $\mu_a = 0$, μ_b = 1.24 and μ_c = 2.04 μ_B/Mn atom. We also measured the temperature dependence of these reflections using the TAS 2 diffractometer. Although the 110 and 101 reflections were not appreciably resolved, we obtained the intensities I_{110} and I_{101} by curve fitting using the double Gaussian functions. The results are shown in Fig. 3. We have confirmed that the alloy shows [c] at 600 K from the neutron diffraction experiments, so we suppose that the transition temperature T_t is nearly equal to T_N (= 580 K).

Another γ-MnRh alloy with 16 at % Rh (γ-$Mn_{0.84}Rh_{0.16}$) shows [t_2] with a = 3.748 and c = 3.828 Å at 24 K and [c] above T_t (= 400 K). A detailed phase diagram for the Mn-Rh system is shown in reference 8. We have made neutron diffraction experiments for γ-$Mn_{0.84}Rh_{0.16}$ using TAS 2, and observed the strong 101 reflection and no 110 reflection. The 101 intensity decreases monotonically with increasing temperature without a disturbance at T_t (= 400 K), and vanishes at T_N (= 600 K). The absence of the 110 reflection in the [t_2] region suggests that the magnetic moments lies in the c-plane, i.e. $\mu_c = 0$.

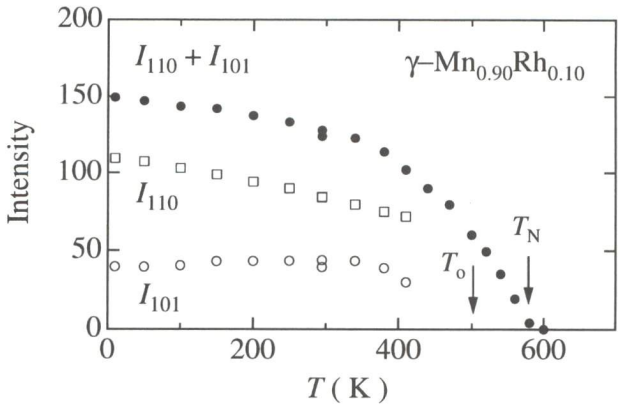

Fig. 3. Temperature dependence of 110 and 101 intensities for γ-$Mn_{0.90}Rh_{0.10}$.

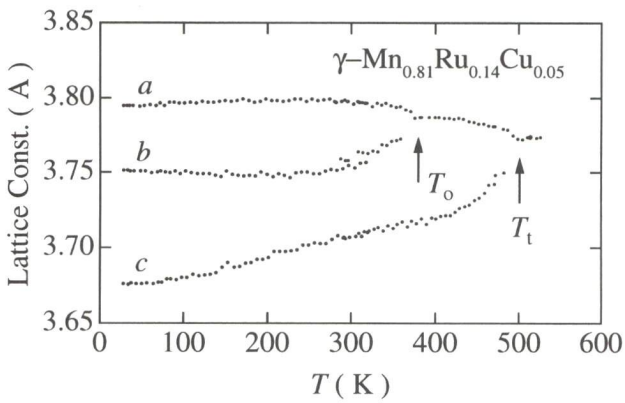

Fig. 4. Temperature dependence of lattice constants for γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$.

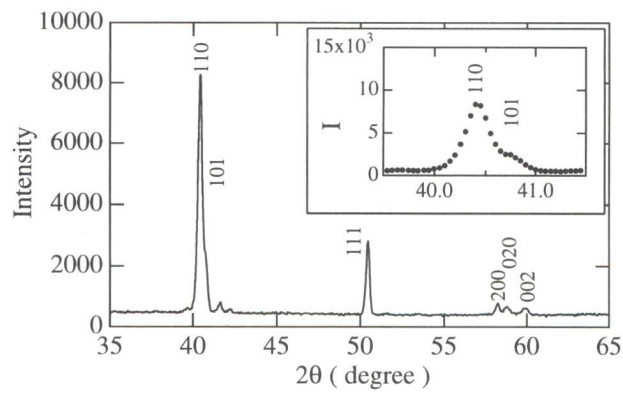

Fig. 5. Neutron diffraction pattern for γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$ at 10 K.

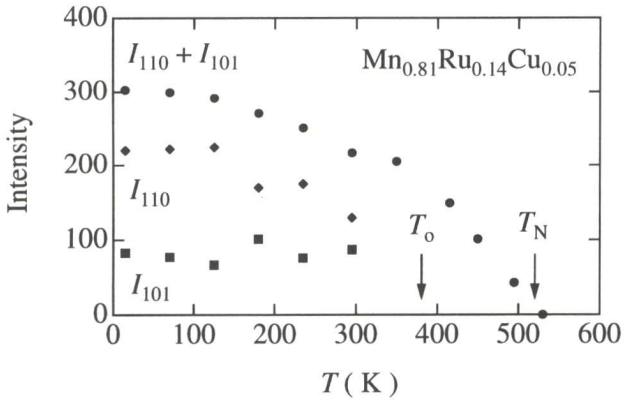

Fig. 6. Temperature dependence of 110 and 101 intensities for γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$.

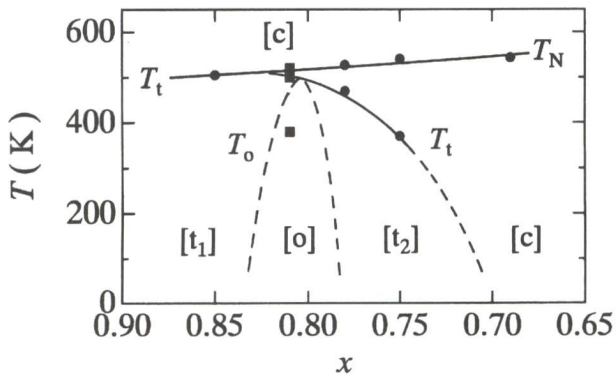

Fig. 7. Tentative phase diagram for γ-MnRu alloys with a small amount of Cu. Closed circles and closed squares show the data for γ-$Mn_xRu_{0.96-x}Cu_{0.04}$ and γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$, respectively.

3.2 γ-MnRu alloys containing a small amount of Cu.

Fig. 4 shows the temperature dependence of the lattice constants for γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$. As seen in Fig. 4, the crystal distortion occurs from [o] to $[t_1]$ at T_o (= 380 K), and from $[t_1]$ to [c] at T_t (= 500 K). We have determined the Néel temperature T_N (= 520 K) from the temperature dependence of the susceptibility; T_N is higher than T_t. We have also made neutron diffraction experiments for this alloy. Fig. 5 shows a neutron diffraction pattern for γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$ at 10 K obtained by using HRPD (λ = 1.823 Å). The pattern is explained by assuming a = 3.794, b = 3.750 and c = 3.677 Å and μ_a = 0, μ_b = 1.33 and μ_c = 2.47 μ_B/Mn atom. Using TAS 2, we also obtain the temperature dependence of the magnetic intensities, I_{110} and I_{101}, by curve fitting the double Gaussian functions, as shown in Fig. 6. The total magnetic intensity decreases monotonically with increasing temperature without a disturbance at T_o (= 380 K) and vanishes at T_N (= 520 K). The results are similar to that of γ-$Mn_{0.84}Rh_{0.16}$.

We have also studied other γ-MnRu alloys with a small amount of Cu, γ-$Mn_xRu_{0.96-x}Cu_{0.04}$. The X-ray diffraction experiments reveal that the alloy with x = 0.85 shows the $[t_1]$ region at low temperatures: a = 3.775, and c = 3.629 Å at 27 K. This is transformed to [c] at T_t (= 505 K). We have also confirmed that the alloy with x = 0.78 shows $[t_2]$ with a = 3.720 and c = 3.799 Å at 28 K and [c] above T_t (= 470 K). Similarly, the alloy with x = 0.75 shows $[t_2]$ with a = 3.733 and c = 3.798 Å at 27 K and [c] above T_t (= 370 K). In addition, more Ru rich alloy with x = 0.69 shows [c] until low temperature: a = 3.756 Å at 23 K and a = 3.768 Å at 293 K. The Néel temperatures of these alloys have been determined from the temperature dependence of susceptibility. The results are as follows: T_N = 527 K for x = 0.78, T_N = 540 K for x = 0.75 and T_N = 543 K for x = 0.69. Fig. 7 shows a phase diagram for γ-$Mn_xRu_{0.96-x}Cu_{0.04}$ alloys, including tentatively the γ-$Mn_{0.81}Ru_{0.14}Cu_{0.05}$ as x = 0.81. This diagram is very similar to that of the γ-MnRh and the γ-MnGa alloys. It is likely that the [o] region is commonly inserted between the $[t_1]$ and $[t_2]$ regions.

§4. Summary

We have found that there is an orthorhombic region for the γ-MnRh alloy with 10 at % Rh and the γ-MnRu alloy containing 14 at % Ru and 5 at % Cu. The lattice constants and magnetic moments at 10 K are as follows: a = 3.806, b = 3.748 and c = 3.671 Å and μ_a = 0, μ_b = 1.24 and μ_c = 2.04 μ_B/Mn atom for the γ -MnRh alloy with 10 at% Rh, and a = 3.794, b = 3.750 and c = 3.677 Å and μ_a = 0, μ_b = 1.33 and μ_c = 2.47 μ_B/Mn atom for γ-MnRu alloy containing 14 at % Ru and 5 at % Cu. We have also determined a tentative phase diagram for γ-MnRu alloys with a small amount of Cu. The present results are consistent with Jo's Theory.

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