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Journal of Alloys and Compounds 383 (2004) 37–39

Journal of ALLOYS AND COMPOUNDS

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Pressure effect on the Curie temperature of the Heusler alloys Rh_2MnZ (Z = Sn, Ge)

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Abstract

The pressure effect on the Curie temperature (T_C) of Rh_2MnZ (Z = Sn, Ge) has been investigated by measuring the temperature dependence of initial permeability at various pressures up to about 1 GPa. The Curie temperature T_C and its pressure coefficients $(1/T_C)dT_C/dp$ were obtained to be 431 K and $+2.6 \times 10^{-2}$ GPa^{-1} for Rh_2MnSn, and 471 K and $+1.7 \times 10^{-2}$ for GPa^{-1} Rh_2MnGe, respectively. It was found that a pressure induced phase transition occurs around 0.6 GPa in Rh_2MnGe.

Keywords: Heusler alloys; Pressure effect; Curie temperature

1. Introduction

The Heusler alloys Rh_2MnZ (Z = Sn, Ge) have the $L2_1$-type crystal structure. Rh_2MnSn is ferromagnetic with the Curie temperature T_C of 412 K and has a total magnetic moment of $3.1\mu_B$/formula [1]. Rh_2MnGe is also ferromagnetic with T_C of 450 K and a magnetic moment of $4.3\mu_B$/formula [2]. The electronic structure and magnetic moment of Rh_2MnX (X = Ge, Sn and Pb) were calculated by Pugacheva and Jezierski [3]. Their results show that mostly magnetic moments are localised on the Mn atom and depend on the local atomic order in the alloys. The magnetic moment on the Rh atom is small (approximately $0.4\mu_B$/atom) and the magnetic properties of these alloys are mainly connected with those of the Mn atoms. The relationship between the Mn–Mn interatomic distance and the Curie temperature has been studied for $L2_1$-type and $C1_b$-type Heusler alloys [4]. The results show that the Curie temperature of both alloys depends on the Mn–Mn interatomic distance and the number of valence electrons.

In this paper we report on the pressure dependence of the Curie temperature and discuss the interatomic distance dependence of the exchange interaction in the ferromagnetic Mn Heusler alloys.

2. Experimental details

The polycrystalline samples of Rh_2MnZ (Z = Sn, Ge) were prepared from Rh (99.9%), Mn (99.99%), Sn (99.9999%) and Ge (99.9999%). They were mixed in the desired proportion and sealed in evacuated silica tubes. To prepare Rh_2MnSn, the mixture of Rh, Mn and Sn was heated at 250 °C for 7 h, annealed at 700 °C for 6 days and quenched in water. The reaction products were pulverized, mixed, heated again in evacuated silica tubes at 700 °C for 6 days, and then quenched in water. To prepare Rh_2MnGe, the mixture of Rh, Mn and Ge was annealed at 950 °C for 6 days and quenched in water. The reaction products were pulverized, mixed, heated again in evacuated silica tubes at 950 °C for 6 days, and then quenched in water.

The powder X-ray diffraction measurements were performed with Cu Kα radiation at room temperature. The obtained diffraction patterns indicated that the prepared samples were single phase with the ordered $L2_1$-type structure. The lattice parameters a of Rh_2MnSn and Rh_2MnGe are determined to be 6.24 and 6.03 Å, respectively.

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doi:10.1016/j.jallcom.2004.04.003

Magnetization measurements were performed at magnetic fields up to 50 kOe by using a superconducting quantum interface device (SQUID) magnetometer.

The initial permeability was measured as a function of temperature under hydrostatic pressures with an ac transformer method. The primary and secondary coils were wound around a cylindrical aggregate of sample. When an ac current of a constant amplitude flows in the primary coil, the voltage induced in the secondary coil is directly proportional to initial permeability. The hydrostatic pressure was applied to a sample in a Teflon pressure cell filled with a liquid pressure medium by using a piston–cylinder type device.

3. Results and discussion

Magnetization of Rh_2MnSn was measured at magnetic fields up to 50 kOe in the temperature range from 5 to 350 K. The obtained magnetization curves are characteristic of ferromagnets. We performed the $\sigma(H, T)^2$ versus $H/\sigma(H, T)$ plot (so-called Arrot plot) based on the results of magnetization curves. The spontaneous magnetization σ_s at each temperature was determined by the linear extrapolation to $H/\sigma = 0$ of the plot at high fields. The value of σ_s and the magnetic moment p_s per formula at 5 K were estimated to be 57.9 emu/g and $3.93\mu_B$/formula, respectively. The plot of σ_s against temperature is well fitted to a Brillouin function for $s = 2$ with $T_C = 431$ K, where T_C is the value estimated by the method mentioned below.

Fig. 1 shows the initial permeability μ versus temperature curves for Rh_2MnSn at various pressures up to about 1 GPa. The value of μ decreases rapidly just below T_C with increasing temperature and then takes almost a constant value above T_C. The Curie temperature T_C was defined as the point intersection of linear extrapolations of the μ–T curves from both higher and lower temperature ranges as shown in the figure.

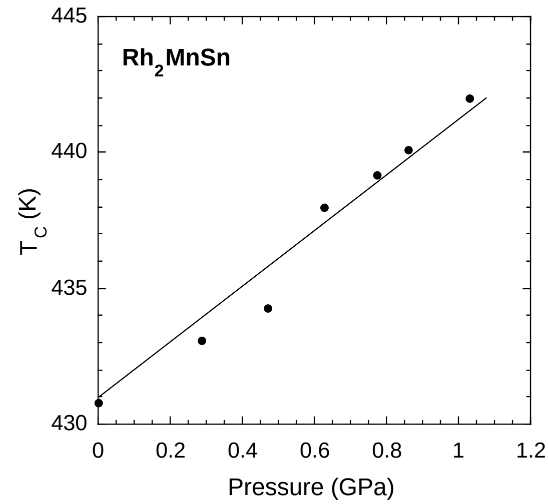

Fig. 2. Pressure dependence of the Curie temperature T_C for Rh_2MnSn.

The Curie temperature was obtained to be $T_C = 431$ K at normal pressure, which is larger than 412 K [1]. The Curie temperature appears to increase with pressure as seen in the figure. The pressure dependence of T_C is shown in Fig. 2. The Curie temperature increases linearly with pressure. The pressure coefficient of the Curie temperature $(1/T_C)dT_C/dp$ is estimated to be $+2.6 \times 10^{-2}$ GPa^{-1} as large as that of Ni_2MnX (X = Al, Ga, In, Sn) [4].

The magnetization of Rh_2MnGe was also measured at magnetic fields up to 50 kOe in the temperature range from 5 to 300 K. The obtained magnetization curves are characteristic of ferromagnets. We determined the spontaneous magnetization σ_s at each temperature using the Arrot plot. The value of σ_s and magnetic moment p_s at 5 K were estimated to be 69.9 emu/g and $4.17\mu_B$/formula, respectively.

Fig. 3 shows the initial permeability μ versus temperature curves for Rh_2MnGe at hydrostatic pressures up to about

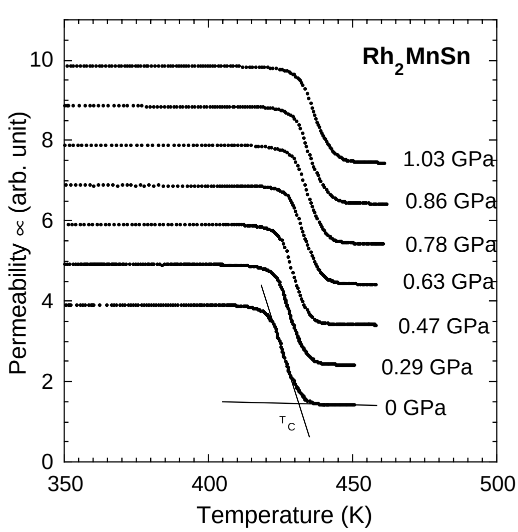

Fig. 1. The initial permeability μ vs. temperature curves at various pressure for Rh_2MnSn.

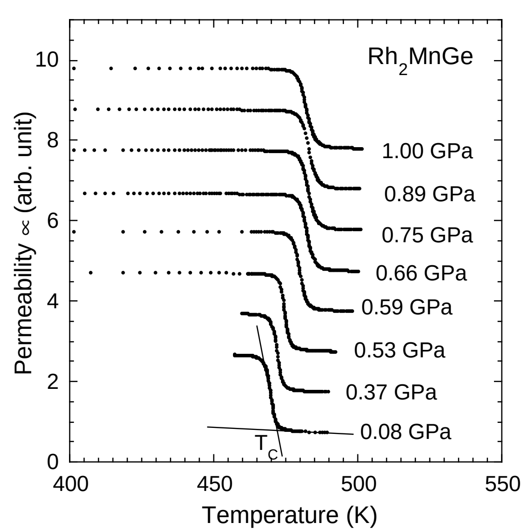

Fig. 3. The initial permeability μ vs. temperature curves at various pressures for Rh_2MnGe.

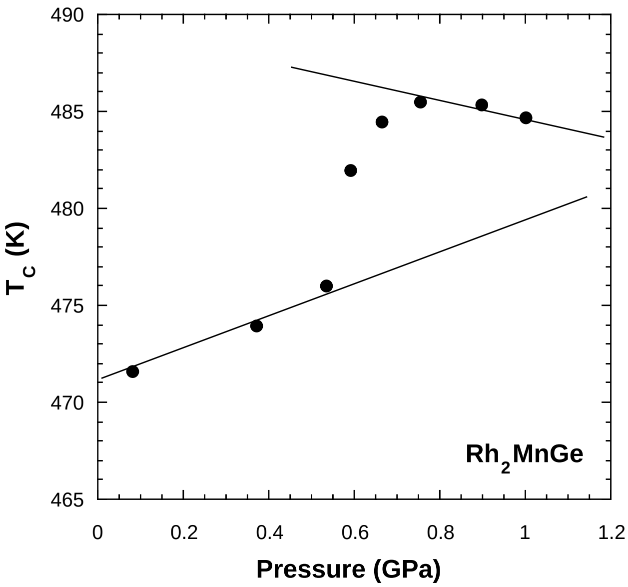

Fig. 4. Pressure dependence of the Curie temperature T_C for Rh_2MnGe.

1 GPa. The Curie temperature was determined by using the same method as for Rh_2MnSn. The pressure dependence of T_C is shown in Fig. 4. The Curie temperature increases linearly with pressure at first, steeply increases around 0.6 GPa, and then decreases above 0.8 GPa. The Curie temperature was estimated to be 471 K at normal pressure. These results suggest that there occurs the pressure induced phase transition around 0.6 GPa ($= P_t$). The pressure coefficient of the Curie temperature is estimated to be $(1/T_C)dT_C/dp = +1.7 \times 10^{-2}\,GPa^{-1}$ below the transition pressure P_t and $(1/T'_C)dT_C/dp = -1.0 \times 10^{-2}\,GPa^{-1}$ above P_t, where T'_C is the Curie temperature of the pressure induced phase.

The results for Rh_2MnSn and Rh_2MnGe below P_t can be understood qualitatively on the basis of the dependence of the exchange interaction on the Mn–Mn distance in the same manner, as it has been assumed by Kanomata et al. [4] for Ni_2MnX (X = Al, Ga, In, Sn), since both Mn–Mn distances, 4.41 Å in Rh_2MnSn and 4.26 Å in Rh_2MnGe are larger than those in Ni_2MnX. There have been proposed so-called interaction curves showing the relationship between T_C and the Mn–Mn interatomic distance R by Yamada [5] and Castelliz [6]. According to Yamada's proposition, the interaction curve has the maximum around $R = 3.5$ Å. This suggests that the pressure coefficient is negative for the alloys with $R < 3.5$ Å. On the other hand, it was recently found that several Heusler alloys like Ni_2MnGa show a crystallographic transition (martensite transition) and that the crystallographic and magnetic transitions have a close relation to each other [7]. If we assume that Rh_2MnGe transforms into a ferromagnetic alloy with smaller R at the critical pressure P_t by a pressure induced crystallographic transition, the negative pressure coefficient of the Curie temperature for Rh_2MnGe above P_t can be ascribed to that of the Curie temperature T'_C for a new ferromagnetic phase induced by pressure. Thus, it is interesting to examine whether there occurs a crystallographic transformation at P_t. X-ray diffraction measurements under pressure are now in progress to reveal the pressure induced phase transition of Rh_2MnGe.

Acknowledgements

This work was carried out under the Visiting Researcher's Program of the Laboratory for Advanced Materials, Institute for Materials Research, Tohoku University.

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