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The Curie temperature in Heusler alloys Ni_2MnZ (Z = Ga, Sn and Sb) under high pressure

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ABSTRACT

The pressure effect on the Curie temperature T_C of the Heusler alloys Ni₂MnZ (Z=Ga, Sn and Sb) has been investigated by measuring the temperature dependence of initial permeability at various pressures up to 100 kbar. It was found that the Curie temperature of Ni₂MnZ (Z=Ga, Sn and Sb) increases almost linearly with increasing pressure and then shows the tendency of saturation at high pressure. The pressure derivatives of T_C in the pressure range below about 60 kbar were estimated to be 0.36 K/kbar for Ni₂MnGa, 0.68 K/kbar for Ni₂MnSn. The value of dT_C/dp at 5 kbar for Ni₂MnSb was estimated to be ~ 1.2 K/kbar. © 2012 Published by Elsevier B.V.

1. Introduction

In recent years, Heusler alloys have become the subject of intensive experimental and theoretical investigations as potential candidates for smart materials. Heusler alloys are usually defined at the stoichiometric composition X_2YZ (X, Y = transition element; Z = s, p element) with the $L2_1$ structure. Most Mn-based Heusler alloys X_2MnZ show the ferromagnetic behavior. The pressure effect on the Curie temperature T_C of these Heusler alloys was studied intensively for Cu₂MnAl, Ni₂MnZ (Z = Al, Ga, Sn and Sb), Au₂MnAl, Pd₂MnZ (Z = Sn and Sb) and Rh₂MnZ (Z = Sn and Ge) [1–10]. For all of Mn-based Heusler alloys mentioned above, T_C increased linearly with increasing pressure in the pressure range below about 10 kbar. On the basis of the results, the interatomic distance dependence of the exchange interactions was discussed qualitatively for a number of the Mn-based Heusler alloys [3,4].

Recently, Şaşıoğlu et al. investigated the pressure dependence of electronic structure, exchange interactions and Curie temperature in the ferromagnetic Ni₂MnSn Heusler alloy theoretically within the framework of the density-functional theory [11]. They showed that the character of the pressure dependence of the exchange

interactions is a consequence of the complex interplay of competing trends in the electronic properties of Ni₂MnSn. According to their results, T_C of Ni₂MnSn increases with increasing pressure up to about 100 kbar. Extending their theoretical study to a larger pressure interval, they obtained the non-monotonic pressure dependence of T_C .

Measurements of pressure shift on T_C for the ferromagnetic Mnbased Heusler alloys under pressures up to about 10 kbar have been given by many authors as mentioned above. As far as we know, there is no data on the pressure shift on T_C of the ferromagnetic Mn-based Heusler alloys under pressures up to 100 kbar.

In this paper, we report the pressure effect on the Curie temperature of the Heusler alloys Ni₂MnZ (Z=Ga, Sn and Sb) under pressure up to 100 kbar (120 kbar for Ni₂MnSn) and discuss the interatomic distance dependence of the exchange interaction. Preliminary results have been reported previously by our group [7].

2. Experimental

The polycrystalline samples of Ni₂MnZ (Z = Ga, Sn and Sb) were prepared from Ni (99.9%), Mn (99.9%), Ga (99.9999%), Sn (99.999%) and Sb (99.9%). They were mixed in the desired proportion and sealed in evacuated silica tubes. To prepare Ni₂MnGa, the mixture of Ni and Mn powders and Ga grains was heated at 850 °C for 6 days. The reaction product was pulverized, mixed, heated again in evacuated silica tubes at 820 °C for 4 days, and then quenched in water. Polycrystalline Ni₂MnSn and Ni₂MnSb were also synthesized by the chemical reaction method as well as the case of Ni₂MnGa. The X-ray powder diffraction measurements were carried out using



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Fig. 1. Temperature dependence of the initial permeability μ at 5, 50 and 100 kbar for Ni₂MnGa. The arrows in the figure show the Curie temperature $T_{\rm C}$.

Cu K α radiation at room temperature. The obtained diffraction patterns of Ni₂MnZ (Z=Ga, Sn and Sb) indicated that the prepared samples were in single phase with the ordered $L2_1$ -type structure.

High-pressure apparatus used in this study is a double-stage "6-8 type" multianvil operated with a large hydraulic press. All anvils are made of tungsten carbide, and the six first-stage anvils drive the eight second-stage anvils toward a sample. A quasi-hydrostatic pressure can be generated in an octahedral pressure cell with an edge of 7 mm. The pressure was calibrated by using the phase transition pressures of pure Bi. The initial permeability μ was measured as a function of temperature at various pressures with an ac transformer method. The primary and secondary coils were wound around a cylindrical aggregate of sample in the pressure cell. When an ac current of a constant amplitude with about 80 Hz flows in the primary coil, the voltage induced in the secondary coil is directly proportional to initial permeability μ . The amplitude of the ac magnetic field produced by the primary coil is about 20.1 Oe. The Curie temperature was determined from the μ versus *T* curves, where temperature decreased from the paramagnetic state to the ferromagnetic state.

3. Results and discussion

Fig. 1 shows the initial permeability μ versus *T* curves at 5 kbar, 50 kbar and 100 kbar for Ni₂MnGa. The value of μ takes a low value above T_C with decreasing temperature and then increases rapidly around T_C . The T_C was defined as the cross point of linear extrapolation lines from both higher and lower temperature ranges on the μ versus *T* curve. The Curie temperature is found to be 358 K at 5 kbar. This value is in good agreement with reported earlier [3]. The T_C of Ni₂MnGa increases with increasing pressure as shown in the figure. Similar μ versus *T* curves for Ni₂MnGa were observed at various pressures. The pressure dependence of T_C for Ni₂MnGa is shown in Fig. 2. As shown in Fig. 2, T_C increases almost linearly with increasing pressure up to about 60 kbar and then the gradient of T_C versus *p* curve becomes smaller at pressures higher than about 60 kbar. The pressure derivative of T_C in the pressure range below about 60 kbar, dT_C/dp , is found to be 0.36 K/kbar.

The pressure dependence of $T_{\rm C}$ for Ni₂MnSn is shown in Fig. 3. As seen in the figure, $T_{\rm C}$ increases almost linearly with increasing pressure up to about 60 kbar, but the pressure dependence of $T_{\rm C}$ shows the tendency of saturation at pressures higher than 60 kbar. The value of $dT_{\rm C}/dp$ in the pressure range below about 60 kbar is estimated to be 0.68 K/kbar. Fig. 4 shows the pressure dependence of $T_{\rm C}$ for Ni₂MnSb under pressures up to 100 kbar. As shown in the figure, the tendency of saturation for the pressure dependence of $T_{\rm C}$ is observed from the low pressure range. The Curie temperature at 5 kbar is found to be 353 K. The value of $dT_{\rm C}/dp$ at 5 kbar is estimated to be about 1.2 K/kbar. This value is about 3 times smaller



Fig. 2. Pressure dependence of the Curie temperature T_C for Ni₂MnGa. The solid line in the figure is a guide for the eyes.

than that reported by Kaneko et al. [2]. On the other hand, Austin and Mishra reported that $T_{\rm C}$ of Ni₂MnSb is independent of pressure [1]. They determined the pressure change of $T_{\rm C}$ for Ni₂MnSb by measuring the temperature dependence of magnetization at various pressures up to 12 kbar. Kanomata et al. reported the value of $dT_C/dp = 4.0 \text{ K/kbar}$ at the limit of p = 0 kbar for Ni₂MnSb. Thus, the values of dT_C/dp for Ni₂MnSb scatter by the reported authors. Recently Şaşıoğlu et al. carried out the first-principles band calculation for a model system where the atoms of Mn sublattices in Ni₂MnSn are interchanged with the atoms of the Ni sublattices [11]. With Mn-Ni interchange, they obtained a substantial difference in the electronic structure of the system. On the basis of the results of calculation, they showed that the value of dT_C/dp exceeds strongly the corresponding value for the system without Mn-Ni atomic interchange. The scattering of the value of dT_C/dp in Ni₂MnSb may be attributed to the degree of Ni-Mn disorder in the samples.

As shown in Figs. 2–4, the pressure change of $T_{\rm C}$ for the Heusler alloys Ni₂MnZ (Z=Ga, Sn and Sb) was investigated up to the



Fig. 3. Pressure dependence of the Curie temperature $T_{\rm C}$ for Ni₂MnSn [7]. The solid line in the figure is a guide for the eyes.



Fig. 4. Pressure dependence of the Curie temperature T_C for Ni₂MnSb. The solid line in the figure is a guide for the eyes.

pressure range of 100 kbar (120 kbar for Ni₂MnSn) in this study. It was found that the pressure dependences of T_{C} for Ni₂MnZ (Z=Ga, Sn and Sb) show the tendency of saturation at high pressure. The increased pressure leads to broadening of the bands that stems from the decreasing interatomic distance. It is well known that the band broadening leads to the decrease of the magnetic moments. Şaşıoğlu et al. calculated the pressure dependence of the interatomic exchange parameters of Ni₂MnSn as mentioned above [11]. According to their analysis, the leading contribution to $T_{\rm C}$ of Ni₂MnSn is given by the Mn–Mn exchange interaction within the first three coordination spheres. Namely, the first neighbor Mn-Mn exchange interaction increases with pressure below about 110 kbar and decreases with further increase of pressure. The second and third Mn-Mn exchange interactions increase with increasing pressure. These peculiar behaviors of the exchange interactions are due to the competition of the two opposite trends, namely the effect of the increasing hopping and the effect of the decreasing atomic moments [11]. It should be noted that the Mn and Ni magnetic moments decrease with increasing pressure and the electron hopping increases with increasing pressure as Sasioğlu et al. pointed out in their report [11]. As a result, Şaşıoğlu et al. observed a flat feature in the pressure dependence of the T_C for Ni₂MnSn in the pressure range from 90 to 160 kbar. The pressure dependence of the measured Curie temperature for Ni_2MnSn in this study is in qualitatively agreement with the calculated one. A theoretical investigation of the pressure dependence of the Curie temperature for Ni_2MnGa and Ni_2MnSb will also be necessary.

4. Summary

The pressure effect on the Curie temperature $T_{\rm C}$ of the ferromagnetic Heusler alloys Ni₂MnZ (Z = Ga, Sn and Sb) has been investigated by measuring the temperature dependence of initial permeability at various pressures up to 100 kbar (120 kbar for Ni₂MnSn). High pressure was generated by the double-stage "6-8 type" multi-anvil operated with a large hydraulic press. The $T_{\rm C}$ of Ni₂MnZ (Z = Ga, Sn and Sb) increases almost linearly with increasing pressure and then shows the tendency of saturation at high pressure. The values of $dT_{\rm C}/dp$ in the pressure range below about 60 kbar were found to be 0.36 K/kbar for Ni₂MnGa, 0.68 K/kbar for Ni₂MnSn. The value of $dT_{\rm C}/dp$ at 5 kbar for Ni₂MnSb is estimated to be about 1.2 K/kbar. The experimental results of Ni₂MnSn in this study are in qualitative agreement with the calculated one reported by Şaşıoğlu et al. [11].

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