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Pressure Effect on the Magnetic Transition Temperatures of Mn_7Sn_3Ge and $Mn_7Sn_2Ge_2$

Satoru OHTA, Naoto HAGIWARA[1], Seiji FUJII[1], Hajime YOSHIDA[2], Takejiro KANEKO[2], Shuichiro ANZAI[1] and Masanori MATOBA[1]

Hachinohe Institute of Technology, Hachinohe 031, Japan
[1]*Faculty of Science and Technology, Keio University, Hiyoshi, Yokohama 223, Japan*
[2]*Institute for Materials Research, Tohoku University, Katahira, Sendai 980, Japan*

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The pressure (P) derivative of the ferrimagnetic Curie temperature dT_C/dP for the Ni_2In-type $Mn_7Sn_{4-z}Ge_z$ is determined to be $dT_C/dP=(1.8\pm0.1)-(0.4\pm0.1)z$ K/kbar. The signs of the pressure derivatives dT_f/dP are negative for $z=0$ and 0.4, where T_f is the transition temperature from the spin-glass like state to the ferrimagnetic one. These pressure effects are discussed on the basis of the interaction curve for antiferromagnetic Mn alloys.

KEYWORDS: ferrimagnetic Curie temperature, spin glass, pressure effect, exchange interaction, Mn_7Sn_4, $Mn_7Sn_{4-z}Ge_z$

The Ni_2In-type Mn_7Sn_4 is of ferrimagnetic below the Curie temperature $T_C=230$ K.[1] A neutron diffraction investigation[2] has revealed the magnetic structure of Mn_7Sn_4 (at 90 K) which consists of alternatively stacked MnI($0.8\mu_B$) c-planes and c-planes of MnII($3.8\mu_B$) and Sn atoms (Fig. 1(a)). A spin-glass like behavior has been found below a magnetic transition temperature $T_f=99$ K.[3] Then, it has been suggested that the competition between effective magnetic interactions exists among the Mn atoms. It is of interest to study the magnetic and electronic properties of such a mixed moment system, whose magnetic moments are very different each other, including competitive interactions. In the free atomic states Ge and Sn are of equielectronic in their valence shells. The replacement of Sn with Ge in Mn_7Sn_4 decreases the hexagonal lattice parameters a and c, and lowers T_C.[4] However, the hydrostatic compression of Mn_7Sn_4 raises T_C.[5] In the present paper the pressure (P) effect on T_C, together with on T_f, is investigated on $Mn_7Sn_{4-z}Ge_z$.

Sintered samples of $Mn_7Sn_{4-z}Ge_z$ ($z\leq2$) were prepared with the procedure reported previously.[3] The measurements of the shift of T_C as a function of P for various z were carried out by using the piston cylinder-type apparatus in heating runs. The pressure transmitting fluid was Fluorinert.

As the temperature (T) increases, the initial permeability μ at ambient pressure steeply increases up to T_f above which it gradually decreases and shows a sharp peak (Hopkinson peak) around T_C. Here, T_C is assigned from the intersection of two lines drawn in the μ-T curve. As P increases, T_C increases and T_f decreases. The pressure behavior of T_C and T_f is consistent with those reported

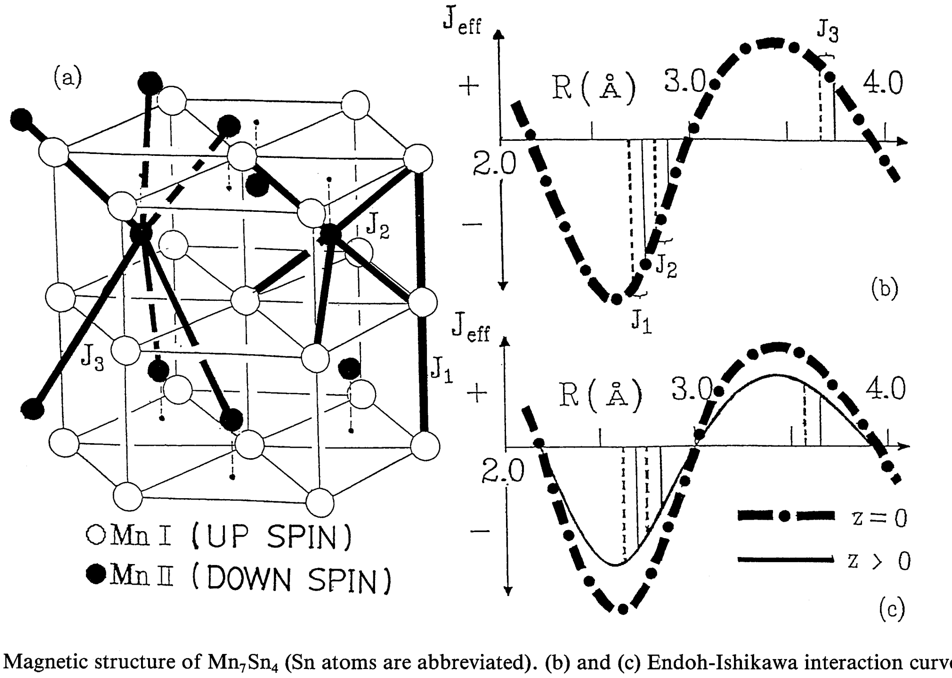

Fig. 1. (a) Magnetic structure of Mn_7Sn_4 (Sn atoms are abbreviated). (b) and (c) Endoh-Ishikawa interaction curve. See text.

previously.[5,6]

Figure 2 shows that the higher pressures give the higher T_C's of Mn_7Sn_3Ge and $Mn_7Sn_2Ge_2$. The pressure dependence of T_C for $Mn_7Sn_{4-z}Ge_z$ is shown in Fig. 3. Below about 5 kbar there is a non-linear variation of T_C. For example, the values of T_C (□) for Mn_7Sn_3Ge measured in increasing pressure of the second run differ from those of the virgin run (○), but above 5 kbar they coincide with those of the virgin runs of increasing pressure. (The P-hysteresis of T_C below about 5 kbar would be attributed to a state of internal stress in the samples quenched from high temperature.) Hence, the pressure derivatives dT_C/dP are evaluated from the data for pressures higher than 5 kbar. Figure 4(a) shows that dT_C/dP decreases with increasing z. The least-squares fitting to the dT_C/dP vs z line gives $dT_C/dP=(1.8\pm0.1)-(0.4\pm0.1)z$.

Figure 4(b) shows the T_f vs P curves for Mn_7Sn_4 and $Mn_7Sn_{3.6}Ge_{0.4}$. Above 5 kbar, the derivatives dT_f/dP are both negative (Mn_7Sn_4: -3 K/kbar and $Mn_7Sn_{3.6}Ge_{0.4}$: -2 K/kbar).

Our previous XPS investigation has disclosed the metallic character of $Mn_7Sn_{4-z}Ge_z$[4] so that the magnetic interactions are discussed on the basis of the following interaction curve proposed on the antiferromagnetic Mn alloys. Based on the magnetic structure of α-Mn, Yamada *et al.*[7] have led a Mn-Mn interaction (J_{eff}) curve as a function of Mn-Mn distance (R). Endoh and Ishikawa[8] have extended it to higher R range by taking into account the Néel temperatures and the nearest and the next nearest neighboring Mn-Mn distances for NiMn, $PtMn_3$, PdMn, $RhMn_3$ and PtMn. Their Interaction curve is redrawn by the thick dot-and-dash curves in Figs. 1(b) and 1(c). Recently, Kanomata and Kaneko[9] have shown that the Cu_2Sb-type Mn alloys having smaller Mn moments ($<2\mu_B$) range on the curve below the critical Mn-Mn distance $R_c=2.85$ Å while those having larger ones ($>2\mu_B$) distribute above R_c.

In Mn_7Sn_4, the first, the second and the third nearest neighboring Mn-Mn distances are $R_1=2.76$ Å, $R_2=2.89$ Å and $R_3=3.75$ Å, respectively. The Mn-Mn interactions for R_1, R_2 and R_3 are J_1 for MnI-MnI, J_2 for MnI-MnII and J_3 for MnII-MnII, respectively. They are shown by thick solid lines in Fig. 1(a). In Fig. 1(b), vertical solid lines schematically represent J_1, J_2 and J_3. The ferrimagnetic structure of Mn_7Sn_4[2] is maintained by six pairs (per one Ni_2In-type unit cell, and so on) of J_2 (<0) and six ones of J_3 (>0). The interaction curve indicates the sign of J_1 to be negative. However, the observed spin arrangment between MnI moments is of ferromagnetic. The MnI moments are forced to align ferromagnetically each other by J_2 and J_3. Under this situation, MnI moments frustrate each other through J_1. Therefore, these competing interactions cause the spin-glass like state in Mn_7Sn_4 below T_f.

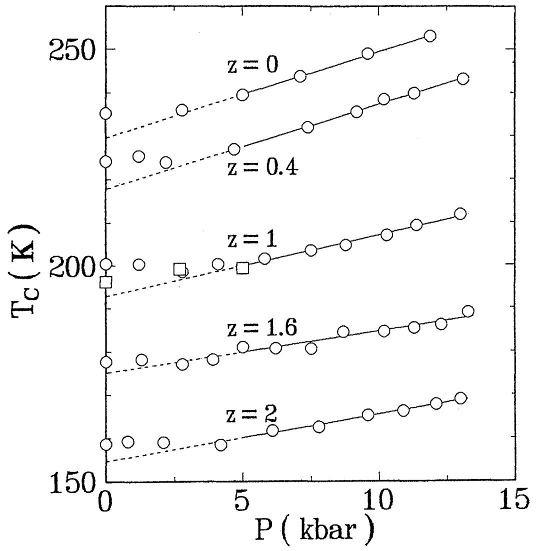

Fig. 3. Ferrimegntic Curie temperatures (T_C) as a function of P for $Mn_7Sn_{4-z}Ge_z$ ($0\le z\le 2$). Here, ○ represents in the first run and □ in the second run.

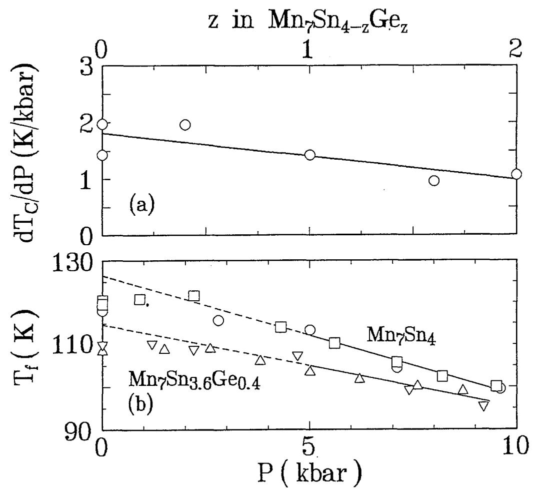

Fig. 4. (a) dT_C/dP as a function of z for $Mn_7Sn_{4-z}Ge_z$. (b) Spin-glass like to ferrimagnetic transition temperature (T_f) as a function of P for Mn_7Sn_4 and (○ and □) $Mn_7Sn_{3.6}Ge_{0.4}$ (△ and ▽). Different signs represent the different runs.

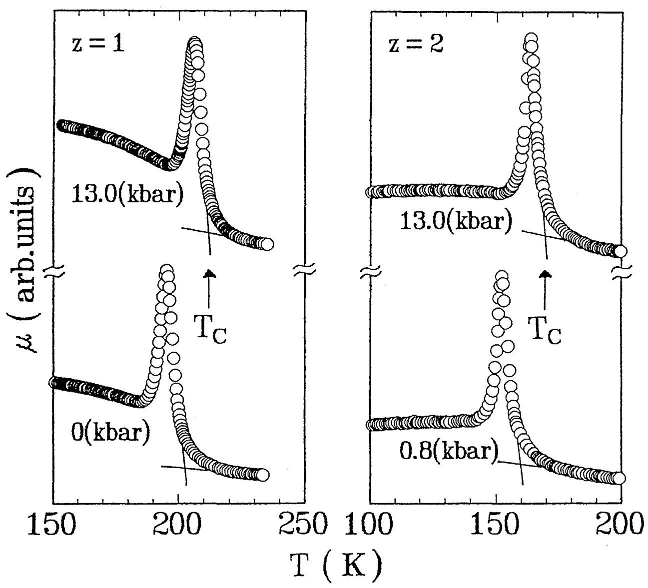

Fig. 2. Permeability (μ) vs temperature (T) curves for Mn_7Sn_3Ge ($z=1$) and $Mn_7Sn_2Ge_2$ ($z=2$) at typical pressures (P).

When the inter-atomic distance of Mn_7Sn_4 contracts by pressure, the magnitudes of $6\times J_2$ and $6\times J_3$ are enhanced rather than that of $2\times J_1$. This feature is shown by the vertical dashed lines in Fig. 1(b). This pressure effect on J_{eff} makes the ferrimagnetic state to be more stable. Namely, the pressure expands the ferrimagnetic region; $dT_C/dP>0$ and $dT_f/dP<0$. If the amplitude of the interaction curve decreases with the increase in z, one can explain the observed features $dT_C/dz<0$ and $d(dT_C/dP)/dz<0$ with $dR_1/dz<0$, $dR_2/dz<0$ and $dR_3/dz<0$. Here, a typical curve with reduced amplitude is shown by the solid curve in Fig. 1(c). The vertical solid and dashed lines represent $J_1<0$, $J_2<0$ and $J_3>0$ for $P=0$ and those for $P>0$, respectively.

Kübler *et al.*[10] have carried out self-consistent spin-polarized energy-band calculations for X_2MnM Heusler alloys with X=Co, Ni, etc. and M=Al, Sn etc. They have proposed the covalent mechanism in which the M–Mn p-d hybrid states in the vicinity of the Fermi level play an important role in determining the details of the moment-alignment geometry and the change in Mn moment. The small variation of the interaction curve for Ge-subsituted Mn_7Sn_4 system might be related to this situation. Further neutron diffraction and XPS investigations are desired to elucidate a more detailed electronic feature for this behaviour.

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