Pressure Effect on the Curie Temperature of Ni_2In -Type $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$

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The variations of the Curie temperature T_c and ferrimagnetic moment n_f in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ ($0 \leq z \leq 0.35$) have been investigated as functions of z and hydrostatic pressure P up to the maximum 12.5 kbar. Below $z \sim 0.25$, T_c decreases with increasing z , but it increases above $z \sim 0.25$. The pressure derivative of T_c , dT_c/dP decreases and n_f increases with increasing z below $z \sim 0.1$ above which these values are nearly constant. These features are compared with the results of $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ and $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ from the electronic and magnetic points of view.
[pressure effect, Curie temperature, ferrimagnetism, $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$, itinerant electron magnet]

1. Introduction

The Ni_2In -type Mn_7Sn_4 is ferrimagnetic below the Curie temperature $T_c = 230\text{K}$ [1]. According to the neutron diffraction study[2], the magnetic moments at MnI sites are ferromagnetically aligned in the basal plane while those at MnII sites are antiferromagnetically coupled with those at MnI sites. Here, in Mn_7Sn_4 , there are two different types of Mn sites; MnI and MnII, as shown in the inset of Fig.1. The ferrimagnetic moment n_f comes from the difference in Mn moments, $m(\text{Mn})$,

ferrimagnetic materials Mn_7Sn_4 shows that 3d electrons of Mn contribute to the Fermi edge[3]. Pressure contraction raises T_c in Mn_7Sn_4 [4]. On the other hand, the Ni_2In -type $\text{Fe}_{1+\delta}\text{Sn}$ ($\delta > 0$) is ferromagnetic below $T_c \sim 700\text{K}$, in which the saturation magnetization per Fe atom is $1.8 \mu_B$ [5]. From the Mössbauer spectra, the values of $m(\text{FeI}) = 1.9 \mu_B$ and $m(\text{FeII}) = 1.8 \mu_B$ have been estimated[6]. In order to study the stability of magnetic states in metallic solids, we report the z and P (pressure) dependences of T_c , together with the z dependence of n_f , in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$, and compare them with the cases of $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ [7,8] and $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ [9,10].

2. Experimental methods

The methods of sample preparations and magnetic measurements were described in the previous paper[9]. The permeability (μ) of the samples was measured under hydrostatic pressure (P) up to the maximum 12.5 kbar. Here, the generation of pressure was performed with a piston-cylinder type apparatus. Fluorinart(#75) was used as a high-pressure medium. The powder X-ray diffraction pattern for the samples obtained can be explained in terms of the Ni_2In -type crystal structure and no foreign peak was detected.

3. Results

As seen in the upper panel of Figure 1, T_c decreases with increasing z up to a critical concentration $z_c = 0.25$ above which it turns to increase up to $z = 0.35$. Figures 2 and 3 show μ as a function of temperature (T) at typical pressures for the $z = 0.05$ and 0.1 samples and the $z = 0.2$ and 0.3 ones, respectively. As P increases, the sharp peak shifts to the higher temperature range. Here, T_c is assigned from the intersection of two lines drawn in the μ - T curve. Figure 4 shows T_c as a function of P for various z samples, in which T_c increases with increasing P . Below $P \sim 5\text{kbar}$, non reproducible features of T_c have been reported on the data for $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ [10] and $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ [8]. Hence, the data of T_c ($P > 5\text{kbar}$) are discussed hereafter. As seen in the lower panel of Figure 5, the pressure derivative of T_c , dT_c/dP , steeply decreases with increasing z in the range $0 \leq z \leq 0.1$, while it

Fig.1. Curie temperature T_c (upper panel); $\circ$ from the σ - T curve; $\square$ from the μ - T curve and lattice parameters a and c (lower panel) as a function of z in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$. Inset shows the Ni_2In -type structure. Here, open, closed and dotted circles represent for MnI, MnII and Sn atoms, respectively.

between MnI and MnII sites: $m(\text{MnI}) = 0.8 \mu_B$ and $m(\text{MnII}) = 3.8 \mu_B$. An X-ray photoemission investigation on the Ni_2In -type

slowly changes in $0.1 < z \leq 0.3$. The least squares fitting to the dT_c/dP against z in the range $0 \leq z \leq 0.1$ gives the value of $d(dT_c/dP)/dz = -9.5 \times 10^{-2} (\text{K/kbar})/\text{at}\%$. This value is closer to that $(d(dT_c/dP)/du = -6.0 \times 10^{-2} (\text{K/kbar})/\text{at}\%)[8]$ for $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ but considerably larger than that $(d(dT_c/dP)/dx = -1.7 \times 10^{-2} (\text{K/kbar})/\text{at}\%)[10]$ for $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$. Figure 6 shows the magnetization σ at 6K as a function of magnetic field (H) up to 6T. As H increases, σ steeply increases and then saturates in the higher H range. These data are analyzed on the basis of the

Fig.2. Permeability as a function of temperature T for the $z=0.05$ and 0.1 samples in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ at typical pressures P .

Fig.3. Permeability as a function of temperature T for the $z=0.2$ and 0.3 samples in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ at typical pressures P .

Arrott plots which is written as $\sigma^2 = \alpha + \beta (H/\sigma)$ through the least-squares fitting, where α and β are the parameters. The ferrimagnetic moment at 6K, n_f , which is estimated from the square root of α ,

Fig.4. Curie temperature T_c as a function of pressure P for various z samples in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$.

Fig.5. Ferrimagnetic moments at 6K, n_f (upper panel) and the pressure derivative of Curie temperature T_c , dT_c/dP (lower panel), as a function of z for $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$.

initially increases up to about $z=0.1$ above which it hardly depends on z in the range $0.1 < z \leq 0.35$ (the upper panel of Fig.5). As seen in the inset of Fig.6, β for $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ and $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ increase in a similar fashion while that for $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ remains constant.

4. Discussion

The values of lattice parameters a and c at room temperature linearly decrease with increasing z (the lower panel of Fig.1), $u[7]$ and $x[11]$.

The value of n_f only slightly depends on x but it steeply decreases with increasing u [7,11]. Then, it seems that n_f is mainly determined with the number of 3d electrons of metal atoms in the band features. This is because the total valence electrons does not vary with the change in x [10]. The number of 3d electrons of Fe is intermediate between those of Mn and Co. However, it is remarkable that $dn_f/dx < 0$ while $dn_f/dz > 0$ ($0 \leq z \leq 0.1$). Hence, these features of T_c and n_f would be related to the difference in electron correlation (I) nature of 3d electrons in Fe and Co impurities. For $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ [10] and $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ [8], the values of dT_c/dP are fairly described with a

Fig.6. Magnetization σ as a function of magnetic field H at 6K for various z samples in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$. $\circ$: $z=0$; $\bullet$: 0.05; $\blacksquare$: 0.1; $\square$: 0.15; $\blacklozenge$: 0.2; $\diamond$: 0.25; $\blacktriangle$: 0.3; $\triangle$: 0.35. Inset shows the Arrott plot's parameter β (see text) for $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ ($\square$), $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ ($\circ$) and $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ ($\diamond$).

relation[12] concerning electron correlation in itinerant electron ferromagnetic systems:

$$\frac{dT_c}{dP} = AT_c - \frac{B}{T_c} \quad (1)$$

Here, A and B are the parameters. In eq.(1), the first term is dominant in the case of strongly correlated electron ferromagnetic system, while the second term is dominant in weakly correlated one. As seen on the $T_c(dT_c/dP)$ vs T_c^2 curves(Fig.7), the values observed in $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ populate in the significantly lower region from the curves for $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ and $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$. The upper panel of Figure 1 also shows $dT_c/dz > 0$ above $z \sim 0.25$; $T_c(z=1)$ is estimated to be $7 \times 10^2 \text{K}$ by extrapolating to $z=1$. Moreover, the value of $n_f(=1.6 \mu_B)$ in the range $0.25 < z < 0.35$ is comparable with that ($1.8 \mu_B$) in $\text{Fe}_{1-\delta}\text{Sn}$ [5]. Therefore, it is plausible that Fe impurities retain their local magnetic character having the

stronger I than that of Co impurities in Mn_7Sn_4 . More microscopic studies such as NMR and Mössbauer effect are highly desired to discuss the concrete physical states of Fe impurities.

Fig.7. $T_c(dT_c/dP)$ against T_c^2 plots for $(\text{Mn}_{1-z}\text{Fe}_z)_7\text{Sn}_4$ ($\square$), $(\text{Mn}_{1-u}\text{Co}_u)_7\text{Sn}_4$ ($\circ$) and $\text{Mn}_7(\text{Sn}_{1-x}\text{Ge}_x)_4$ ($\diamond$).

According to the spin fluctuation theory[13] for itinerant ferromagnetism, the parameter β gives the information on the density of states at the Fermi level E_F , $D(E_F)$, and its energy derivative. Therefore, it is also noted that $d\beta/dz \approx d\beta/du$ besides $dn_f/dz > 0$ and $dn_f/du < 0$, although the theoretical relation between β and $D(E_F)$ is not known on the itinerant antiferromagnetic system.

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