Magnetic Phase Transition of $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ ($x \leq 0.10$)

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The electrical resistivity and the thermoelectric power of $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ ($x \leq 0.1$) are measured. The magnetic phase diagram of $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ is determined from the results of these measurements. With increasing iron concentration x , an intermediate magnetic phase(I) appears between the antiferromagnetic(AF) phase and the ferromagnetic(F) one. The AF-I transition temperature T_{I1} and the I-F transition one T_{I2} decrease with increasing the concentration x .

Key words : electrical resistivity, thermoelectric power, order-order magnetic transition, $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$

1. Introduction

Mn_3GaC is an ordered cubic crystal of a perovskite-type structure with lattice parameter $a = 3.894 \text{ \AA}$. The magnetic properties of Mn_3GaC are characterized by an antiferromagnetic (AF) - ferromagnetic (F) transition at a temperature T_f . The first-order transition from the AF state to the F one with increasing temperature is accompanied by a large discontinuous contraction of the lattice parameter. According to the previous results of neutron diffraction the magnetic moment of the Mn atoms is $1.8 \mu_B$ at 4.2 K and $1.2 \mu_B$ at 170 K¹⁾.

Kaneko et al.²⁾ studied the pressure effect on T_f and the Curie temperature T_c of Mn_3GaC at pressures up to 13 kbar. It was found that a new intermediate magnetic (I) phase appears at pressures higher than about 3 kbar. Recently, Kanomata and Kaneko³⁾ have studied the magnetic properties of $\text{Mn}_{3-x}\text{Co}_x\text{GaC}$ ($x \leq 0.5$) and pointed out that the occurrence of the I phase had a close relation to the magnitude of the lattice parameter. To investigate the mechanism of this transition, we have investigated the transport properties of $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ in this study.

2. Experimental

Polycrystalline samples of $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ ($x \leq 0.1$) were prepared by the same method as Ref. 2. X-ray diffraction

studies showed that all diffraction lines are indexed with a cubic perovskite-type structure. It is found that the lattice parameter decreases linearly with Fe concentration x . Electrical resistivity (ρ) and thermoelectric power (S) were measured by the same methods as Ref. 4. The temperature dependence of ρ and S was measured on sintered samples during heating.

3. Results and Discussion

Figure 1(a) shows the temperature dependence of ρ for $\text{Mn}_{2.97}\text{Fe}_{0.03}\text{GaC}$. There appears the abrupt decrease of ρ similar to that observed in Mn_3GaC ⁴⁾ and two kinds of changes in the slope of the ρ - T curve with increasing temperature. Taking account of the results of the magnetic

Fig. 1 Electrical resistivity versus temperature curves for $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ ($x = 0.03$ (a), 0.07 (b))

measurements of $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}^{5)}$, the abrupt change of ρ at $T_{i1} = 135$ K is thought to correspond to the AF $\rightarrow$ I transition. The changes in the slope of the ρ - T curve at $T_{i2} = 154$ K and $T_C = 251$ K are thought to correspond to the I $\rightarrow$ F and F $\rightarrow$ P transitions, respectively. Similar ρ - T curves were obtained for samples with $x = 0.01, 0.02, 0.05$ and 0.07 (see Fig. 1(b)). For $x = 0.10$, we do not find the abrupt decrease of ρ , indicating that the AF state does not appear in the temperature range above 77 K. The total resistivity is given for usual ferromagnetic metals as follows, $\rho(T) = \rho_0 + aT + bT^2$, where ρ_0 is the residual part, the second term is the resistivity part due to electron-phonon scattering and the third term has a magnetic origin. By fitting this equation to the experimental data in the ferromagnetic region, we obtain $\rho_0 = 0.14 \mu\Omega\text{m}$, $a = 1.0 \times 10^{-2} \mu\Omega\text{m/K}$, $b = 1.9 \times 10^{-3} \mu\Omega\text{m/K}^2$ for $\text{Mn}_{2.97}\text{Fe}_{0.03}\text{GaC}$. It is noted that the above value of b is comparable to those observed for typical weak itinerant ferromagnets $\text{Ni}_3\text{Al}^{6)}$ and $\text{ZrZn}_2^{7)}$. A general theory of resistivity in an itinerant system has been proposed by Ueda and Moriya⁸⁾ on the basis of spin fluctuations. Their work predicts a strong T^2 dependence for the resistivity due to spin fluctuations.

Fig. 2 Thermoelectric power S versus temperature for $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ ($x = 0.03$ (a), 0.07 (b)).

Figure 2(a) shows the temperature dependence of S for $\text{Mn}_{2.97}\text{Fe}_{0.03}\text{GaC}$. As seen in the figure, S increases linearly with decreasing temperature and then makes a strong negative dip at T_C . As temperature decreases below T_C , S passes through a minimum and then exhibits an extremely sharp peak. The transition temperatures T_{i1} and T_{i2} determined by the resistivity measurements are shown in the figure. The value of S above T_C for $\text{Mn}_{2.97}\text{Fe}_{0.03}\text{GaC}$ can be expressed as a sum of a linear term in T and a constant, namely, $AT+B$, A and B being constant. The value of B for $\text{Mn}_{2.97}\text{Fe}_{0.03}\text{GaC}$ is found to be $3.5 \mu\text{V/K}$. In Mott's expression, S is simply proportional to T . Sakurai et al.⁹⁾ pointed out that the constant B was the term induced by spin fluctuations in itinerant magnets. In fact, the value B for $\text{Mn}_{2.97}\text{Fe}_{0.03}\text{GaC}$ is comparable to those of the typical itinerant magnets $\text{MnSi}^{9)}$ and $\text{YMn}_2^{10)}$.

On the basis of the experimental results, the values of T_{i1} , T_{i2} and T_C for $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ are plotted against the concentration x in Fig. 3. The I phase appears at atmospheric pressure for the samples with $0 < x \leq 0.1$. The magnetic states were determined from the results of the magnetization measurements.¹¹⁾ The intermediate (I) phase is thought to be a canted magnetic one observed in Mn_3ZnC . As seen in the figure, T_C increases linearly and both T_{i1} and T_{i2} decrease drastically with increasing the concentration x .

Figure 4 shows the lattice parameter dependence of the magnetic transition temperature T_{i1} for several samples of

Fig. 3 Magnetic phase diagram for $\text{Mn}_{3-x}\text{Fe}_x\text{GaC}$ ($x \leq 0.1$).

Fig. 4 The lattice parameter dependence of the first order magnetic transition temperature T_{I1} for $Mn_{3-x}M_xGaC$ ($M = Fe$ and Cr).

$Mn_{3-x}M_xGaC$ ($M = Fe$ and Cr). For $0 \leq x \leq 0.03$, T_I decreases linearly with decreasing the lattice parameter. It seems that for $x = 0.05$ and 0.07 the deviation of the linear relation for T_I vs. lattice parameter is caused by the change of the magnetic states at low temperature as seen in Fig. 3. The tendency of the lattice parameter dependence of the magnetic transition temperatures in $Mn_{3-x}Fe_xGaC$ is similar to those in $Mn_{3-x}Co_xGaC$ ³⁾ and Mn_3GaC under pressure²⁾. It is

considered that the magnetic phase transitions observed may be discussed in terms of the spin-fluctuation model. To investigate the magnetic structure of the I phase in Fig.3 a neutron diffraction study of $Mn_{3-x}Fe_xGaC$ is in progress.

References

- 1) D.Fruchart, E.F.Bertaut, B.L.Clerc, P.Veillet, G.Lorthior, M.E.Fruchart and R.Fruchart, : *J.Solid Chem.*, **8**, 182 (1973)
- 2) T.Kaneko, T.Kanomata and K.Shirakawa : *J. Phys. Soc. Jpn.*, **56**, 4047 (1987).
- 3) T.Kanomata and T.Kaneko : *Physics of Transition Metals* ; ed.by P.M.Oppeneer and J.Kübbler, p 871 (World Scientific, Singapore, 1993).
- 4) T.Harada, T.Makabe, T.Kanomata and T.Kaneko : *J. Magn. Magn. Mat.* , **104-107**, 1955 (1992).
- 5) T.Harada, to be submitted.
- 6) J.H.J.Fruitman, R.Boom, P.F.De Chatel, C.J.Schinkel, J.L.L.Tilanus and B.R.De Vries : *J. Phys. F : Metal Phys.*, **3**, 109 (1973).
- 7) G.Dublon, F.Habbal and J.Bevk : *Solid State Commun.* , **36**, 625 (1980).
- 8) K.Ueda and T.Moriya : *J. Phys. Soc. Jpn.*, **139**, 605 (1975).
- 9) J.Sakurai, Y.Yamamoto and Y.Komura : *J. Phys. Soc. Jpn.* , **57**, 24 (1988).
- 10) H.Kamimura, J.Sakurai, Y.Komura, H.Nakamura, M.Shiga and Y.Nakamura : *J. Magn. Magn. Mater.* , **70**, 145 (1987).
- 11) T.Goto and K.Kamishima : private communication