

## Electrical-resistance maximum near the Curie point in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$ and $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$

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The electrical resistivity of the  $D0_3$ -type  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  alloys has been measured over the temperature range from 4.2 to 1250 K. The temperature dependence of the resistivity shows an anomaly that has been found in ferromagnetic  $3d$  alloys: an appearance of the resistance maximum near the Curie point and the negative temperature dependence at high temperatures up to 1000 K and above. The tendency of the negative temperature dependence of the resistivity increases markedly with increasing the V or Ti composition. Such an anomalous resistance behavior cannot be accounted for in terms of an order-disorder transformation, since the decrease in the resistivity occurs even below room temperature. At a temperature higher than 800 K, a stepwise reduction of the resistivity appears as a sign of the transformation, which terminates the negative-temperature-coefficient range. Close examination of various components of the resistivity reveals that the magnetic scattering resistance is five to ten times as large as those of ordinary ferromagnetic  $3d$  alloys and that the residual resistivity increases at an extremely large rate of  $12 \mu\Omega \text{ cm}$  for at.% V and  $14 \mu\Omega \text{ cm}$  for at.% Ti in the respective alloys. It is concluded that  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  ( $M = \text{V}$  and  $\text{Ti}$ ) is a type of conductor not to be classified into several other types of metallic conductors hitherto known to exhibit a negative temperature coefficient of resistivity.

### I. INTRODUCTION

The intermetallic compound  $\text{Fe}_3\text{Ga}$  forms an  $L1_2$  crystal structure in a low-temperature equilibrium state,<sup>1,2</sup> unlike  $\text{Fe}_3\text{Al}$  and  $\text{Fe}_3\text{Si}$ , which have a  $D0_3$  crystal structure. However, a slight substitution of Fe in  $\text{Fe}_3\text{Ga}$  by any  $3d$  transition metal causes a more remarkable stabilization of the  $D0_3$  phase.<sup>3</sup> The structural and magnetic properties of the  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  system ( $M = \text{Cr}$ ,  $\text{Mn}$ ,  $\text{Co}$ , and  $\text{Ni}$ ) have been studied by means of x-ray diffraction, magnetic measurements, Mössbauer spectroscopy,<sup>3</sup> and band calculations.<sup>4</sup> These investigations have demonstrated that the stabilization of the  $D0_3$  phase in  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  is closely related to a selective sublattice occupation of substituting  $M$  atoms: that is, Cr or Mn enters the Fe site with eight Fe nearest neighbors in the  $D0_3$  lattice, while Co or Ni enters the Fe site with four Fe and four Ga nearest neighbors. Such a site preference is well known to occur in  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Si}$ , as has extensively been examined by Niculescu, Burch, and Budnick.<sup>5</sup> Recently, we have also found in the  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  system that the substitution of  $M = \text{V}$  or  $\text{Ti}$  leads to the stabilization of the  $D0_3$  phase,<sup>6</sup> as is the case for  $M = \text{Cr}$ ,  $\text{Mn}$ ,  $\text{Co}$ , or  $\text{Ni}$ .<sup>3</sup>

Meanwhile, electrical resistivity of these alloys has not been studied except for  $\text{Fe}_3\text{Ga}$ .<sup>7</sup> The resistivity measurements of  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  have revealed another interesting feature as shown in the present paper.

(1) The resistivity decreases with rising temperature above the Curie point  $T_C$ , which implies an occurrence of

a negative temperature coefficient. (2) Near  $T_C$ , the magnetic scattering resistance shows a gradual change resulting in a broad peak, instead of an angular change observed in ordinary ferromagnetic metals and alloys. (3) The residual resistivity is generally proportional to the V or Ti composition but increases at a rate much larger than that expected from the case of bcc iron solid solutions.

In addition to the electrical resistivity measurements, magnetic properties of  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  have also been measured and are discussed in regard to the anomaly in the resistivity. Further, the electrical resistance maximum and the negative temperature dependence in these alloys are compared with several other metallic conductors which exhibit a similar type of resistance behavior.

### II. EXPERIMENT

#### A. Sample preparation

$(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  alloys were made from 99.99% pure Fe and Ga, and 99.9% pure V or Ti. The properly weighed constituents were preheated to 1373 K in an atmosphere of Ar-3%  $\text{H}_2$  and then melted in an argon arc furnace. The weight loss during the melting procedure was less than 0.5%, which limits the accuracy of the nominal composition assigned to each sample. The ingots were annealed at 1273 K for more than 260 ks in vacuum to ensure homogeneity. Samples for resistivity measurements were cut from the ingots with an alumina-blade saw to the size of  $0.5 \times 0.5 \times 15 \text{ mm}^3$ . Each sample,

sealed in an evacuated quartz capsule, was annealed at 1273 K for 3.6 ks and then quenched into ice water. Subsequently, they were heat-treated at 773 K for 1.2 Ms to form an ordered structure and slow-cooled to room temperature.

### B. X-ray analysis

X-ray diffraction was measured using Co  $K\alpha$  radiation on powder samples prepared as above. For  $M=V$ , a single phase of the  $DO_3$  structure was formed in the composition range  $x=0.05-0.4$ , but the (111) and (200) superlattice reflections were drastically reduced in intensity for  $x > 0.3$ . For  $M=Ti$ , a single phase of the  $DO_3$  structure was found in the range  $x=0.05-0.15$ , whereas at higher compositions a small amount of additional phase, probably a Laves phase,<sup>8</sup> was also detected in the  $DO_3$  matrix.

### C. Measurements

The electrical resistivity was measured by a conventional dc four-terminal method over the temperature range from 4.2 to 1250 K at a rate of 0.05 K/s. The measurements at high temperature were carried out in an atmosphere of Ar-3%  $H_2$ .

The magnetization below room temperature down to 77 K was measured by a vibrating-sample magnetometer using samples of  $0.5 \times 0.5 \times 5 \text{ mm}^3$  in dimension. Thermomagnetic measurements were made on powder samples by a magnetic balance method in the temperature range between 300 and 1000 K. Magnetization was calibrated in reference to the room-temperature value of 55.0 emu/g for Ni.

## III. EXPERIMENTAL RESULTS

### A. Electrical resistivity

The electrical resistivity was measured as a function of temperature for  $(Fe_{1-x}V_x)_3Ga$  and  $(Fe_{1-x}Ti_x)_3Ga$  as shown in Figs. 1 and 2, respectively.<sup>9</sup> The arrows indicate the Curie temperatures  $T_C$ . In these alloys, the electrical resistivity increased rapidly at low temperatures, reached a maximum at or near  $T_C$  and, above  $T_C$ , decreased with rising temperature. The tendency of the negative temperature dependence of the resistivity increased markedly with increasing  $x$ . This negative temperature dependence above  $T_C$  is the most remarkable feature, which has first been found in ferromagnetic  $3d$  alloys. As can be seen in Fig. 1, the decrease in the resistivity occurred even below room temperature for  $x=0.25$  and  $0.3$  in  $(Fe_{1-x}V_x)_3Ga$ . This means that the negative temperature dependence of the resistivity cannot be associated with an order-disorder transformation, unlike the resistance anomaly found in Fe-Pd alloys, which has been explained by the formation of  $L1_2$ -type  $FePd_3$  of long-range order on cooling.<sup>10,11</sup> Other characteristics in the conduction phenomena are a high residual resistivity and a large magnetic scattering resistance, as will be discussed later.

Another feature to be noted in Figs. 1 and 2 is that the

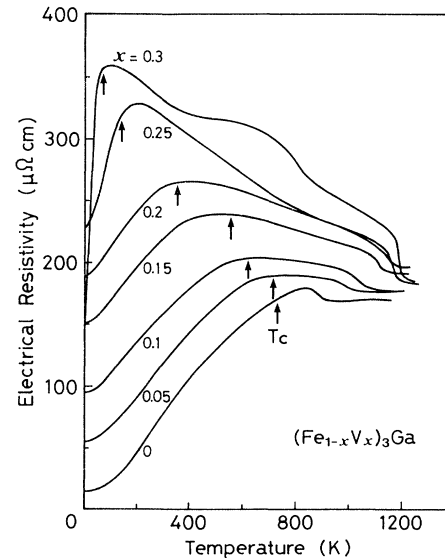


FIG. 1. Temperature dependence of electrical resistivity in  $(Fe_{1-x}V_x)_3Ga$ . The arrows indicate the Curie temperatures  $T_C$ .

resistivity curves showed a stepwise reduction at about 800 K or higher temperatures. This is considered to be related to the order-disorder transformation,<sup>6</sup> although it seems unusual that the resistivity decreases when disordering occurs. Then, the transformation temperature, taken as a point of the drop, rises rapidly with increasing  $x$ , thus indicating a remarkable stabilization of the  $DO_3$  phase over a wide temperature range. It should also be remarked that this change terminates the negative temperature-coefficient range. At temperatures higher than the transformation temperature, the resistivity appears to be almost constant, independent of composition, and is equal to about  $200 \mu\Omega \text{ cm}$ . Therefore, the drop in the resistivity seems to imply an important change in the

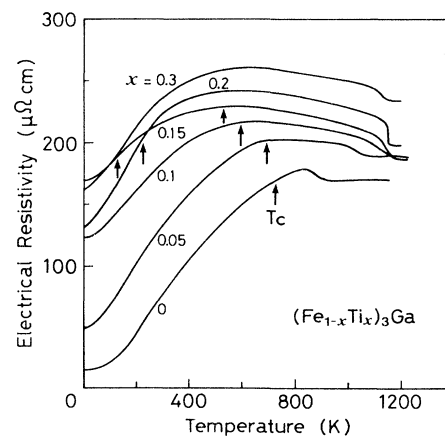


FIG. 2. Temperature dependence of electrical resistivity in  $(Fe_{1-x}Ti_x)_3Ga$ . The arrows indicate the Curie temperatures  $T_C$ .

conduction mechanism. To fully understand the nature of the transformation, however, it is necessary to make *in situ* structural studies.

### B. Magnetic properties

The measured bulk magnetization  $\sigma$  versus applied magnetic field up to 16 kOe saturated in a manner characteristic of ferromagnetism for  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  with  $0 \leq x \leq 0.25$  and for  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  with  $0 \leq x \leq 0.3$  at 80 K. The values of the Curie temperature  $T_C$ , were obtained from the thermomagnetic measurements using a modified Arrott-plot method.<sup>12</sup> The saturation magnetization  $\sigma_0$  was calculated by the use of  $T^{3/2}$  extrapolation to 0 K and is plotted against composition  $x$  in Fig. 3. Now,  $\sigma_0$  gives the average atomic moment  $\mu$  in units of the Bohr magneton ( $\mu_B$ ) as follows:

$$\mu = \sigma_0 [(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}] / 4N_0\mu_B = \frac{3}{4}\mu_T, \quad (1)$$

where  $N_0$  denotes Avogadro's number,  $[(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}]$  is in g mol per formula unit, and  $\mu_T$  is the moment per transition-metal atom on assuming that Ga atoms have no moment. The last term  $\mu_T$  consists of the weighted sum of the Fe and  $M$  moments:

$$\mu_T = (1-x)\mu_{\text{Fe}} + x\mu_M. \quad (2)$$

The result is shown in Fig. 4, where the gradient represents the derivative of  $\mu_T$  with respect to  $x$ :

$$\frac{d\mu_T}{dx} = -\mu_{\text{Fe}}(0) + \frac{d\mu_{\text{Fe}}}{dx} + \mu_M, \quad (3)$$

where the first term in the right side is the initial Fe moment in  $\text{Fe}_3\text{Ga}$ , the second is the Fe moment perturbation induced by  $M$  substitution, and the third is the moment to be assigned to the  $M$  atom. The first term would give a simple dilution case and is equal to  $-2\mu_B$ . The observed

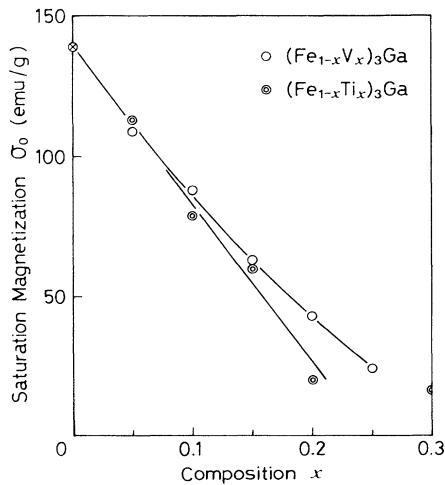


FIG. 3. Saturation magnetization  $\sigma_0$  (in emu/g) as a function of composition  $x$  in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  ( $\circ$ ) and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  ( $\odot$ ).

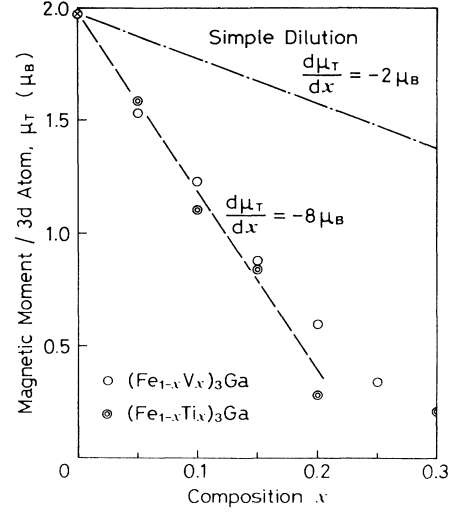


FIG. 4. Magnetic moment per 3d transition-metal atom,  $\mu_T$  (in  $\mu_B$ ), as a function of composition  $x$  in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  ( $\circ$ ) and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  ( $\odot$ ). The observed gradient,  $d\mu_T/dx = -8\mu_B$ , is shown by the dashed line and a simple dilution case,  $d\mu_T/dx = -2\mu_B$ , is shown by the dash-dotted line.

gradient  $d\mu_T/dx$ , however, is  $-8\mu_B$  per  $M$  atom, which is much larger in magnitude than that expected from a simple dilution, so that the difference is too large to be ascribed to  $\mu_M$  alone. Therefore, this discrepancy in  $d\mu_T/dx$  should be ascribed mainly to the second term which can take a large value because a substituting  $M$  atom perturbs many neighboring Fe atoms simultaneously.

## IV. DISCUSSION

### A. General aspects of electrical resistivity

The temperature dependence of the electrical resistivity observed in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  can be represented schematically by the upper curve (a) in Fig. 5, as compared with that of an ordinary ferromagnetic metal shown by the lower curve (b). The latter curve consists of three components: the residual resistivity  $\rho_0$ , the phonon scattering term  $\rho_{\text{ph}}(T)$ , and the magnetic scattering term  $\rho_m(T)$ , as shown by the dashed lines in Fig. 5. The total resistivity  $\rho_t$ , is the sum of these three terms, of which the first two are observed even in non-magnetic alloys, i.e.,

$$\rho_t = \rho_0 + \rho_{\text{ph}}(T) + \rho_m(T). \quad (4)$$

In reference to these three terms, the various components of the resistivity of  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  are considered. Each term has the following temperature dependence.

Firstly, the residual resistivity  $\rho_0$  is temperature independent and corresponds to the resistivity extrapolated to 0 K. As shown in Fig. 6, the value of  $\rho_0$  increases linearly with composition  $x$  in the range up to  $x=0.25$  for  $M=\text{V}$  and  $x=0.15$  for  $M=\text{Ti}$ , following

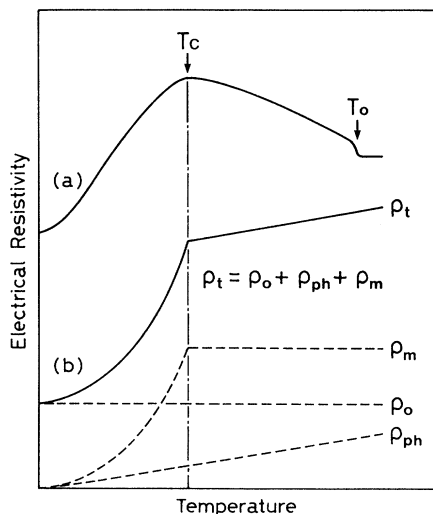


FIG. 5. Schematic illustration of the temperature dependence of electrical resistivity for  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  [curve (a)] and an ordinary ferromagnet [curve (b)]. The total resistivity  $\rho_t$  is the sum of residual resistivity  $\rho_o$ , phonon scattering term  $\rho_{\text{ph}}(T)$ , and magnetic scattering term  $\rho_m(T)$ .  $T_C$  and  $T_o$  represent a Curie temperature and an order-disorder transformation temperature, respectively.

Matthiessen's rule. The concentration  $c$  (at. %) of  $M$  atoms is  $3x/4$  and the resistivity increase per 1 at. %  $M$  is given by

$$d\rho_0/dc = \frac{4}{3}d\rho_0/dx. \quad (5)$$

From Fig. 6, this coefficient has been found to be an extremely large value of  $12 \mu\Omega \text{ cm}$  for at. % V and  $14 \mu\Omega \text{ cm}$  for at. % Ti in the respective alloys.

Secondly, the phonon scattering term  $\rho_{\text{ph}}(T)$  is proportional to  $T^5$  at very low temperatures and to  $T$  at high

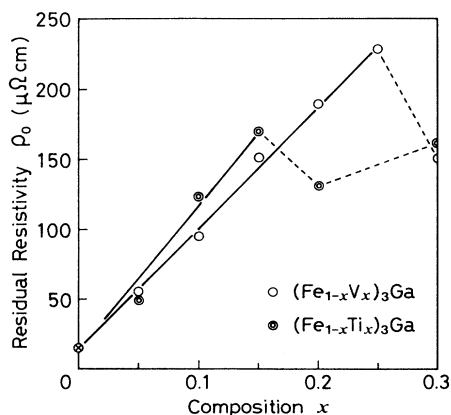


FIG. 6. Residual resistivity  $\rho_o$ , as a function of composition  $x$  in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  (○) and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  (●). The resistivity increases per 1 at. % V and Ti are  $12$  and  $14 \mu\Omega \text{ cm}$ , respectively.

temperatures. Phonon scattering is the only cause for the temperature dependence of resistivity in ordinary non-magnetic metals and alloys and in magnetic ones above the Curie point  $T_C$  or the Néel point  $T_N$ . Therefore, the  $\rho_{\text{ph}}(T)$  term is usually separable from the other terms, i.e.,  $\rho_o$  and  $\rho_m(T)$ , in consideration of the Grüneisen formula.<sup>13</sup> This is not the case, however, for  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  because the anomalous temperature dependence occurs only above  $T_C$ , as can be seen in Figs. 1 and 2. In any way, this term must also exist in these alloys in the form superimposed with  $\rho_m(T)$  or an anomalous resistance term. However, since the temperature coefficient  $d\rho_{\text{ph}}(T)/dT$ , is usually of the order of  $0.01 \mu\Omega \text{ cm/K}$ , the  $\rho_{\text{ph}}(T)$  term can be negligible in the first approximation as far as a much larger temperature dependence is concerned.

Thirdly, the magnetic scattering term  $\rho_m(T)$  is a function of  $M(0)$  and  $M(T)$ , i.e., the saturated magnetization at 0 K and at  $T$  K, where the most important term is  $M(0)^2 - M(T)^2$ . Magnetic scattering is the main cause of the resistivity change below  $T_C$ . The value of  $\rho_m(T)$  reaches its maximum at  $T_C$  and remains constant because  $M(T)$  is always zero above  $T_C$  within an approximation of localized moments. The maximum value,  $\rho_m^*$ , is estimated as the difference between the maximum resistivity and  $\rho_o$ . Since the value of  $\rho_m^*$  is of the order of  $50-150 \mu\Omega \text{ cm}$ , the neglect of the  $\rho_{\text{ph}}(T)$  term in this estimation cannot cause a large error.

### B. Anomaly in magnetic scattering resistance

The maximum value of the magnetic resistivity,  $\rho_m^*$ , is considered to be proportional to the weighted sum of  $S(S+1)$ , i.e., the quantum-mechanical square of the atomic spin, as discussed by Weiss and Marotta.<sup>14</sup>

$$\rho_m^* = \alpha \sum_i p_i S_i(S_i + 1), \quad (6)$$

where a subscript  $i$  denotes the Van Vleck configuration<sup>15</sup> representing each  $3d$  electronic state and  $p_i$  is its population probability. It is known that  $\alpha$  is almost constant, i.e.,  $\alpha = 31.2 \pm 1$  for most  $3d$  metals and alloys,<sup>14</sup> although it comprises various factors such as the  $s-d$  interaction energy coefficient  $G$ , the conduction electron number per atom,  $n$ , and the atomic density  $N$ , i.e.,

$$\alpha = (3\pi^2 N)^{1/3} (mG)^2 / 4\pi e^2 n \hbar^3, \quad (7)$$

where  $m$  and  $\hbar$  are the electron mass and Planck's constant over  $2\pi$ , respectively. In consideration of the relation between  $\rho_m^*$  and the atomic moments, the saturation magnetization of  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  is represented as a function of composition  $x$  in Fig. 3. The result enables one to express  $\rho_m^*$  as a function of moments. Proper calculation of  $S_i(S_i + 1)$  requires the use of half integer  $S_i$  after the Van Vleck model.<sup>15</sup> However, we introduce an approximation to use  $\mu_j/g$  ( $g=2$ ) instead of  $S_i$ , where  $\mu_j$  is the saturation moment of atom  $j$  at 0 K and  $g$  denotes the Landé  $g$  factor. Thus, Eq. (6) is modified to



resistivity curve resembles those of  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$ , despite the difference in the type of spin ordering.

(e) Some V-based alloys with a high resistance have been found to exhibit a negative temperature coefficient by Isino and Muto.<sup>26,27</sup> Mooij<sup>28</sup> has suggested that there is a possible correlation between a high resistance and a negative temperature coefficient in solid solutions and amorphous alloys. Isino<sup>27</sup> explained the anomalous resistance behavior as a result of strong scattering by impurity atoms, which is observed to occur rather commonly in disordered, amorphous or liquid alloys with a high resistance. This phenomenon is characterized by inapplicability of Matthiessen's rule and Nordheim's relation and also by a short mean-free-path of conduction electrons, which is inversely proportional to solute concentration.<sup>27</sup>

(f) A temperature coefficient of resistivity decreases regularly in nonmagnetic amorphous metals, e.g., Ca-Mg-Al as their resistivity becomes higher, as found by Mizutani *et al.*<sup>29,30</sup> In contrast to all the types mentioned above and the present case, they have observed the negative temperature dependence in alloys consisting of simple metals alone, i.e., containing no transition or rare-earth elements.

The cases of (e) and (f) show a common feature that a high electrical resistance in metallic conductors generally tends to reduce a temperature coefficient of resistivity, finally making it negative above a critical value of resistivity. This might occur regardless of details of the electronic structure of conductors, as suggested by Isino and Muto.<sup>26</sup> Such an interpretation helps to understand the anomalous resistance behavior of  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$ . In the present case, magnetism also plays an important role: the negative temperature dependence appears only above  $T_C$ . Further, the elements with less than half-filled  $d$  bands, e.g., Ti and V seem to be more effective for the anomaly than those with more than half-filled ones, in common with the cases (c), (d) and (e).

## V. CONCLUSIONS

The temperature dependence of the electrical resistivity in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$  and  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  shows peculiar characteristics unlike ordinary ferromagnetic  $3d$  alloys: (i) an appearance of the resistance maximum near the Curie point, (ii) the negative resistivity slope at high temperatures up to 1000 K and above, despite its metallic bonding, (iii) the extremely large spin disorder scattering five to ten times as large as those in ordinary ferromagnetic  $3d$  alloys, (iv) the extremely large residual resistivities of 12 and 14  $\mu\Omega$  cm for at. % V and Ti, respectively, and (v) a stepwise reduction in the resistivity at a temperature probably corresponding to an order-disorder transformation.

Comparison among various metals and alloys with a negative temperature coefficient of resistivity has shown that  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  is a new type of metallic conductor not to be classified into the types hitherto known. Such an anomalous temperature dependence of the resistivity seems to take place in a series of  $D\text{O}_3$ -type  $\text{Fe}_3D$  ( $D = \text{Ga}, \text{Al}, \text{Si}$ , etc.) ferromagnetic alloys in which Fe atoms are partly replaced by the transition metals. Extensive measurements of resistivity and magnetism are in progress, as to  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Ga}$  with the transition metals  $M$  other than V and Ti, and also  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Al}$  and  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Si}$ .

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