Electrical-resistance maximum near the Curie point in $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$

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The electrical resistivity of the $D0_3$-type $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ 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 3*d* 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 3*d* alloys and that the residual resistivity increases at an extremely large rate of 12 $\mu\Omega$ cm for at.% V and 14 $\mu\Omega$ cm for at.% Ti in the respective alloys. It is concluded that $(Fe_{1-x}M_x)_3Ga$ (M=V and 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 Fe_3Ga forms an $L1_2$ crystal structure in a low-temperature equilibrium state,[1,2] unlike Fe_3Al and Fe_3Si, which have a $D0_3$ crystal structure. However, a slight substitution of Fe in Fe_3Ga by any 3*d* transition metal causes a more remarkable stabilization of the $D0_3$ phase.[3] The structural and magnetic properties of the $(Fe_{1-x}M_x)_3Ga$ system (M=Cr, Mn, Co, and Ni) have been studied by means of x-ray diffraction, magnetic measurements, Mössbauer spectroscopy,[3] and band calculations.[4] These investigations have demonstrated that the stabilization of the $D0_3$ phase in $(Fe_{1-x}M_x)_3Ga$ 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 $(Fe_{1-x}M_x)_3Si$, as has extensively been examined by Niculescu, Burch, and Budnick.[5] Recently, we have also found in the $(Fe_{1-x}M_x)_3Ga$ system that the substitution of M=V or Ti leads to the stabilization of the $D0_3$ phase,[6] as is the case for M=Cr, Mn, Co, or Ni.[3]

Meanwhile, electrical resistivity of these alloys has not been studied except for Fe_3Ga.[7] The resistivity measurements of $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ 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 $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ 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

$(Fe_{1-x}M_x)_3Ga$ 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% 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 $0.5\times0.5\times15$ 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 $D0_3$ 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 $D0_3$ 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,[8] was also detected in the $D0_3$ 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 $0.5\times0.5\times5$ 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.[9] 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.[10,11] 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

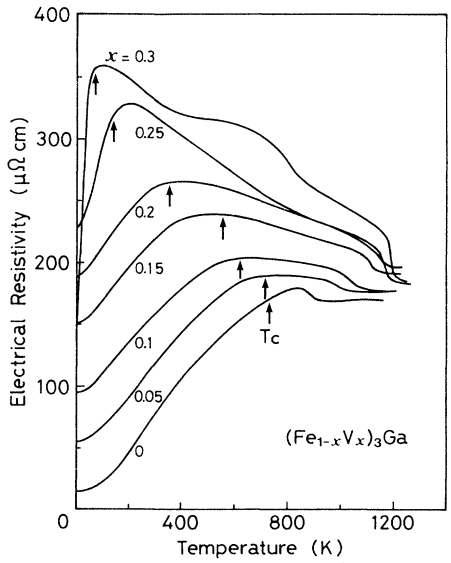

FIG. 1. Temperature dependence of electrical resistivity in $(Fe_{1-x}V_x)_3Ga$. The arrows indicate the Curie temperatures T_C.

resistivity curves showed a stepwise reduction at about 800 K or higher temperatures. This is considered to be related to the order-disorder transformation,[6] 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 $D0_3$ 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 $\mu\Omega$ cm. Therefore, the drop in the resistivity seems to imply an important change in the

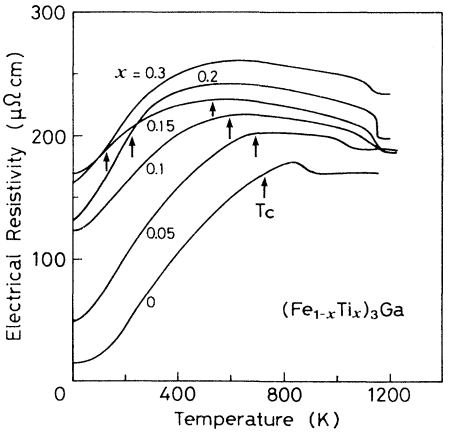

FIG. 2. Temperature dependence of electrical resistivity in $(Fe_{1-x}Ti_x)_3Ga$. The arrows indicate the Curie temperatures T_C.

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 σ versus applied magnetic field up to 16 kOe saturated in a manner characteristic of ferromagnetism for $(Fe_{1-x}V_x)_3Ga$ with $0 \leq x \leq 0.25$ and for $(Fe_{1-x}Ti_x)_3Ga$ 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.[12] The saturation magnetization σ_0 was calculated by the use of $T^{3/2}$ extrapolation to 0 K and is plotted against composition x in Fig. 3. Now, σ_0 gives the average atomic moment μ in units of the Bohr magneton (μ_B) as follows:

$$\mu = \sigma_0[(Fe_{1-x}M_x)_3Ga]/4N_0\mu_B = \tfrac{3}{4}\mu_T \ , \tag{1}$$

where N_0 denotes Avogadro's number, $[(Fe_{1-x}M_x)_3Ga]$ is in g mol per formula unit, and μ_T is the moment per transition-metal atom on assuming that Ga atoms have no moment. The last term μ_T consists of the weighted sum of the Fe and M moments:

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

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

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

where the first term in the right side is the initial Fe moment in Fe_3Ga, 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 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 μ_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.

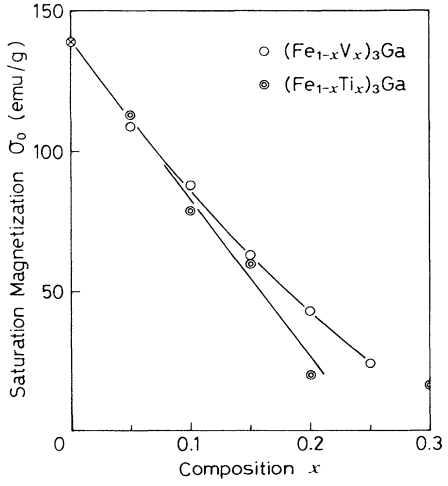

FIG. 3. Saturation magnetization σ_0 (in emu/g) as a function of composition x in $(Fe_{1-x}V_x)_3Ga$ (○) and $(Fe_{1-x}Ti_x)_3Ga$ (◎).

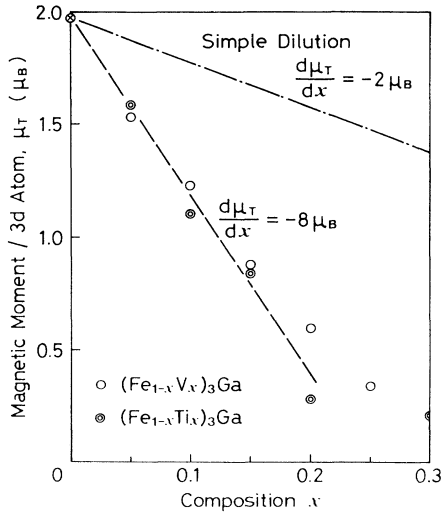

FIG. 4. Magnetic moment per $3d$ transition-metal atom, μ_T (in μ_B), as a function of composition x in $(Fe_{1-x}V_x)_3Ga$ (○) and $(Fe_{1-x}Ti_x)_3Ga$(◎). The observed gradient, $d\mu_T/dx = -8\mu_B$, is shown by the dashed line and a simple dilution case, $d\mu_T/dx = -2\mu_B$, is shown by the dash-dotted line.

IV. DISCUSSION

A. General aspects of electrical resistivity

The temperature dependence of the electrical resistivity observed in $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ 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 ρ_0, the phonon scattering term $\rho_{ph}(T)$, and the magnetic scattering term $\rho_m(T)$, as shown by the dashed lines in Fig. 5. The total resistivity ρ_t, is the sum of these three terms, of which the first two are observed even in nonmagnetic alloys, i.e.,

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

In reference to these three terms, the various components of the resistivity of $(Fe_{1-x}M_x)_3Ga$ are considered. Each term has the following temperature dependence.

Firstly, the residual resistivity ρ_0 is temperature independent and corresponds to the resistivity extrapolated to 0 K. As shown in Fig. 6, the value of ρ_0 increases linearly with composition x in the range up to $x = 0.25$ for $M = V$ and $x = 0.15$ for $M = Ti$, following

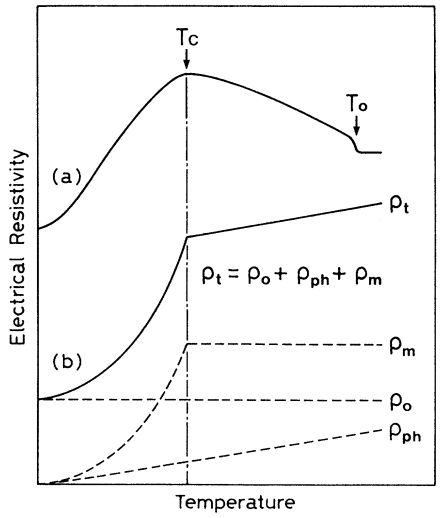

FIG. 5. Schematic illustration of the temperature dependence of electrical resistivity for $(Fe_{1-x}M_x)_3Ga$ [curve (a)] and an ordinary ferromagnet [curve (b)]. The total resistivity ρ_t is the sum of residual resistivity ρ_0, phonon scattering term $\rho_{ph}(T)$, and magnetic scattering term $\rho_m(T)$. T_C and T_0 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

$$d\rho_0/dc = \tfrac{4}{3} d\rho_0/dx \ . \tag{5}$$

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

Secondly, the phonon scattering term $\rho_{ph}(T)$ is proportional to T^5 at very low temperatures and to T at high temperatures. Phonon scattering is the only cause for the temperature dependence of resistivity in ordinary nonmagnetic metals and alloys and in magnetic ones above the Curie point T_C or the Néel point T_N. Therefore, the $\rho_{ph}(T)$ term is usually separable from the other terms, i.e., ρ_0 and $\rho_m(T)$, in consideration of the Grüneisen formula.[13] This is not the case, however, for $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ 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 $d\rho_{ph}(T)/dT$, is usually of the order of 0.01 $\mu\Omega$ cm/K, the $\rho_{ph}(T)$ term can be negligible in the first approximation as far as a much larger temperature dependence is concerned.

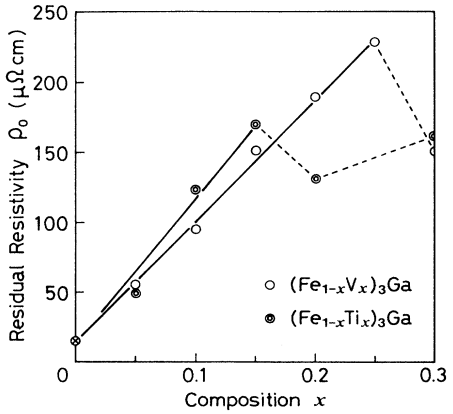

FIG. 6. Residual resistivity ρ_0, as a function of composition x in $(Fe_{1-x}V_x)_3Ga$ (○) and $(Fe_{1-x}Ti_x)_3Ga$(◎). The resistivity increases per 1 at. % V and Ti are 12 and 14 $\mu\Omega$ cm, respectively.

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, ρ_m^*, is estimated as the difference between the maximum resistivity and ρ_0. Since the value of ρ_m^* is of the order of 50–150 $\mu\Omega$ cm, the neglect of the $\rho_{ph}(T)$ term in this estimation cannot cause a large error.

B. Anomaly in magnetic scattering resistance

The maximum value of the magnetic resistivity, ρ_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:[14]

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

where a subscript i denotes the Van Vleck configuration[15] representing each $3d$ electronic state and p_i is its populational probability. It is known that α is almost constant, i.e., $\alpha = 31.2 \pm 1$ for most $3d$ metals and alloys,[14] 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.,

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

where m and $\hbar$ are the electron mass and Planck's constant over 2π, respectively. In consideration of the relation between ρ_m^* and the atomic moments, the saturation magnetization of $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ is represented as a function of composition x in Fig. 3. The result enables one to express ρ_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.[15] However, we introduce an approximation to use μ_j/g $(g=2)$ instead of S_i, where μ_j is the saturation moment of atom j at 0 K and g denotes the Landé g factor. Thus, Eq. (6) is modified to

$$\rho_m^* = \alpha \sum_j p_j \left[\frac{\mu_j}{g} \right] \left[\frac{\mu_j}{g} + 1 \right] . \quad (8)$$

And Fig. 7 represents the relation between ρ_m^* and $\sum p_j(\mu_j/g)(\mu_j/g+1)$. Noticeably, the linear dependence of ρ_m^* on $\sum p_i S_i(S_i+1)$ in Eq. (6) is well reproduced despite the approximation of μ_j/g, as shown by the dashed line that accommodates most data on ordinary $3d$ ferromagnets. In contrast, the results on $(Fe_{1-x}M_x)_3Ga$ remarkably deviate from those on ordinary $3d$ alloys: the value of ρ_m^* of $(Fe_{1-x}M_x)_3Ga$ is extremely large as compared with those of ordinary $3d$ alloys, where μ_j of $(Fe_{1-x}M_x)_3Ga$ is calculated on the simplest assumption that Fe atoms alone have equal magnetic moments to compose the bulk saturation magnetization given in Fig. 3.

There are two possible interpretations for the above discrepancy in ρ_m^*: "large α" and "large μ_j" in reference to Eq. (8), both of which are not necessarily incompatible. The first case is that $(Fe_{1-x}M_x)_3Ga$ may have an exceptionally large α, which directly suggests a small conduction electron number, n, in Eq. (7) because G is 15.5 ± 3.5 eV A^3 for most $3d$ elements[14] and the atomic density N of $(Fe_{1-x}M_x)_3Ga$ is close to that of Fe and other $3d$ ferromagnets. The second case is that μ_j may have much larger values than those calculated from the saturation moment of $(Fe_{1-x}M_x)_3Ga$, which means an emergence of nonparallel spin moments due to the doped M atoms. It should be noted that several $(Fe_{1-x}M_x)_3Si$ alloys have ferrimagnetic or canted spin moments.[16] This is especially likely to be the case for $(Fe_{0.7}V_{0.3})_3Ga$ or nearly the Heusler-type alloy Fe_2VGa, which has a small moment of about $0.3\mu_B$/Fe and yet a very large ρ_m^* of about 200 $\mu\Omega$ cm, in addition to the anomaly in ρ_0.

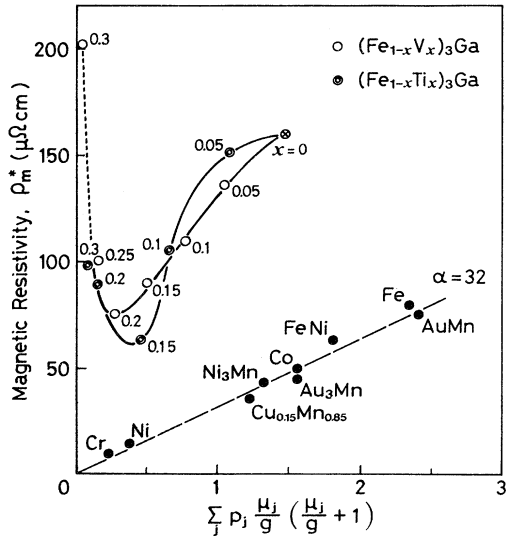

FIG. 7. The maximum value of magnetic scattering resistivity, ρ_m^*, versus $\sum p_j(\mu_j/g)(\mu_j/g+1)$ for $(Fe_{1-x}V_x)_3Ga$ (○) and $(Fe_{1-x}Ti_x)_3Ga$(◎). The dashed line represents the relation for $\alpha=32$ in Eq. (8), which accommodates most data on ordinary $3d$ ferromagnets.

Now, the negative temperature dependence of the resistivity above T_C is the most characteristic feature of $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$. The absolute value of the temperature coefficient of the resistivity increases with increasing x, as observed in Figs. 1 and 2. The value is found to be especially large for $(Fe_{0.75}V_{0.25})_3Ga$ and $(Fe_{0.7}V_{0.3})_3Ga$ that also have an extremely large value of ρ_m^*, as stated above. This suggests a close correlation between the anomaly in the magnetic scattering and the negative temperature dependence of the resistivity.

C. Comparison with other metallic conductors

In order to consider the resistance maximum observed in $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$, let us compare these alloys with various metallic conductors having a negative temperature coefficient of resistivity, of which typical examples are classified into the following types (a) to (f).

(a) The well-known Kondo effect gives rise to a resistance minimum in dilute alloys, where the resistivity increases in proportion to $|\ln T|$ with lowering temperature.[17] The Kondo anomaly occurs only when localized spin moments of $3d$ electrons are effectively noninteracting and therefore disappears when $3d$-metal impurities become dense enough to form the spin glass. Accordingly, this case seems to be in contrast to $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ in which the anomaly occurs at high $3d$-metal concentrations and at high temperatures.

(b) Intermetallic Ce compounds exhibit an anomaly which is interpreted in terms of the dense Kondo effect.[18,19] Since the $4f$ spin moment of Ce atoms is small, i.e., $\frac{1}{2}$, and highly localized, its exchange interaction is small enough to afford the Kondo effect despite a high concentration of magnetic elements. Ce compounds sometimes show a resistance maximum at a temperature where the formation of heavy fermions[20] begins. It is evident that this resistance maximum is intrinsically different from that observed in $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ which contain no $4f$ elements.

(c) A negative temperature dependence of resistivity is observed in Cr and its alloys which form spin-density-wave (SDW) type antiferromagnetism. The anomaly occurs below T_N because the conduction electron density decreases due to a truncation of the Fermi surface accompanying the SDW.[21,22] This case is rather exceptional in that it is caused by a long-period spin ordering.

(d) Inoue *et al.*[23,24] have found an anomaly in a series of noble metals containing a high concentration of Cr or Mn, where the concentration ranges to about 30 at.% and the negative temperature dependence holds up to 1000 K. They analyzed their results in terms of Suhl's formula,[25] an extension of the Kondo theory. It is not certain, however, that such theories are applicable to antiferromagnets containing nondilute $3d$ metals and having T_N higher than 100 K. The temperature dependence of resistivity in these alloys also shows a resistance maximum above T_N. Therefore the shape of the

resistivity curve resembles those of $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$, 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.[26,27] Mooij[28] has suggested that there is a possible correlation between a high resistance and a negative temperature coefficient in solid solutions and amorphous alloys. Isino[27] 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.[27]

(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.*[29,30] 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.[26] Such an interpretation helps to understand the anomalous resistance behavior of $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$. 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 $(Fe_{1-x}V_x)_3Ga$ and $(Fe_{1-x}Ti_x)_3Ga$ 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 $(Fe_{1-x}M_x)_3Ga$ 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 $D0_3$-type Fe_3D (D=Ga, Al, 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 $(Fe_{1-x}M_x)_3Ga$ with the transition metals M other than V and Ti, and also $(Fe_{1-x}M_x)_3Al$ and $(Fe_{1-x}M_x)_3Si$.

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[1]W. Köster and T. Gödecke, Z. Metallkd. **68**, 661 (1977).
[2]J. Bras, J. J. Couderc, M. Fogot, and J. Ferre, Acta Metall. **25**, 1077 (1977).
[3]N. Kawamiya and K. Adachi, J. Magn. Magn. Mater. **31-34**, 145 (1983).
[4]S. Ishida, S. Fujii, N. Shinmura, and S. Asano, J. Phys. Condens. Matter **1**, 5115 (1989).
[5]V. A. Niculescu, T. J. Burch, and J. I. Budnick, J. Magn. Magn. Mater. **39**, 223 (1983), and references therein.
[6]Y. Nishino, M. Matsuo, S. Asano, and N. Kawamiya, Scr. Metall. Mater. (to be published).
[7]N. Kawamiya, K. Adachi, and Y. Nakamura, J. Phys. Soc. Jpn. **33**, 1318 (1972).
[8]V. Ya. Markiv and P. I. Kripyakevich, Kristallografiya **11**, 859 (1967) [Sov. Phys. Crystallogr. **11**, 733 (1967)].
[9]The resistivity curves on heating and on cooling almost coincided with each other. Figures 1 and 2 show the curves on heating.
[10]T. Takezawa, T. Miwa, and T. Yokoyama, J. Jpn. Inst. Metals **51**, 285 (1987).
[11]T. Takezawa and K. Takebayashi, J. Jpn. Inst. Metals **51**, 791 (1987).
[12]A. Arrott, Phys. Rev. **108**, 1394 (1957).
[13]See, e.g., A. H. Wilson, *The Theory of Metals* (Cambridge University Press, Cambridge, England, 1953), p. 287.
[14]R. J. Weiss and A. S. Marotta, J. Phys. Chem. Solids **9**, 302 (1959).
[15]J. H. Van Vleck, Rev. Mod. Phys. **25**, 220 (1953).
[16]V. Niculescu, K. Raj, J. I. Budnick, T. J. Burch, W. A. Hines, and A. H. Menotti, Phys. Rev. B **14**, 4160 (1976).
[17]J. Kondo, Prog. Theor. Phys. **32**, 37 (1964).
[18]H. Schneider and D. Wohlleben, Z. Phys. B **44**, 193 (1981).
[19]A. Sumiyama, Y. Oda, H. Nagano, Y. Ônuki, K. Shibutani, and T. Komatsubara, J. Phys. Soc. Jpn. **55**, 1294 (1986).
[20]G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).
[21]S. Maki and K. Adachi, J. Phys. Soc. Jpn. **46**, 1131 (1979).

[22]S. Maki and K. Adachi, Trans. Jpn. Inst. Metals **22**, 182 (1980).
[23]K. Inoue, Y. Nakamura, and N. Kunitomi, J. Phys. Soc. Jpn. **27**, 1159 (1969).
[24]K. Inoue and Y. Nakamura, J. Phys. Soc. Jpn. **32**, 441 (1972).
[25]H. Suhl, Phys. Rev. Lett. **20**, 656 (1968).
[26]M. Isino and Y. Muto, J. Phys. Soc. Jpn. **54**, 3839 (1985).
[27]M. Isino, J. Phys. Soc. Jpn. **54**, 3848 (1985).
[28]J. H. Mooij, Phys. Status Solidi A **17**, 521 (1973).
[29]U. Mizutani, M. Sasaura, Y. Yamada, and T. Matsuda, J. Phys. F **17**, 667 (1987).
[30]U. Mizutani, in *Proceedings of JIMIS-5: Non-Equilibrium Solid Phases of Metals and Alloys, Kyoto, 1988,* edited by Y. Nakamura [Trans. Jpn. Inst. Metals Suppl. **29**, 275 (1988)].