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## Temperature dependence of electrical resistivity in $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$ alloys

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**Abstract.** We report on the temperature dependence of electrical resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  alloys with Ti compositions  $x = 0\text{--}0.33$ . Samples in the composition range  $0 \leq x \leq 0.15$  are found to exhibit ferromagnetism with the Curie temperature  $T_C$  decreasing from 770 K for  $x = 0$  to 145 K for  $x = 0.15$ . The electrical resistivity below about 400 K for these Ti-poor samples increases rapidly with increasing  $x$ , but a negative temperature derivative of resistivity ( $d\rho/dT$ ) dominates above  $T_C$  up to 1000 K and above. In contrast, samples in the range  $0.20 \leq x \leq 0.33$  are in a paramagnetic state, at least down to 2 K, and exhibit a rapid decrease in the low-temperature resistivity with increasing Ti composition  $x$ . In particular, the Heusler-type  $\text{Fe}_2\text{TiAl}$  ( $x = 0.33$ ) shows a large positive  $d\rho/dT$  with the residual resistivity of only about  $20 \mu\Omega \text{ cm}$ , in sharp contrast to a closely related system  $\text{Fe}_2\text{VAl}$  reminiscent of a semiconductor-like behaviour with the resistivity reaching  $3000 \mu\Omega \text{ cm}$  at 2 K. This can be explained by the fact that  $\text{Fe}_2\text{TiAl}$  possesses a much higher density of states at the Fermi level than  $\text{Fe}_2\text{VAl}$ , as deduced from the low-temperature specific-heat measurements supplemented by the band calculations in literature. The reason for the possession of a large positive  $d\rho/dT$  in  $\text{Fe}_2\text{TiAl}$  is discussed in relation to the Bloch–Grüneisen law.

### 1. Introduction

The intermetallic compound  $\text{Fe}_3\text{Al}$  is a well ordered ferromagnet with a  $\text{D0}_3$  crystal structure. We have recently found an anomalous temperature dependence of electrical resistivity in a series of the pseudobinary alloys  $(\text{Fe}_{1-x}\text{M}_x)_3\text{Al}$  in which Fe atoms are partly replaced by other 3d transition elements such as  $\text{M} = \text{Ti}, \text{V}, \text{Cr},$  and  $\text{Mn}$  [1, 2]. As reviewed by Nishino [3], these alloys possess general features characterized by (1) a resistance maximum near the Curie point  $T_C$  and (2) a negative temperature derivative of resistivity ( $d\rho/dT$ ) at high temperatures above  $T_C$  up to 1000 K and above. Such a resistance anomaly tends to appear more strongly with increasing composition  $x$  of the substituted element M, in parallel with a sharp reduction in  $T_C$ .

The most spectacular feature of the resistance anomaly has been found for  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  [2]. In particular, the Heusler-type  $\text{Fe}_2\text{VAl}$  ( $x = 0.33$ ) is found to be in a marginally magnetic state and to exhibit a semiconductor-like behaviour with the resistivity reaching  $3000 \mu\Omega \text{ cm}$  at 2 K. This unusual resistivity behaviour is accompanied by the presence of a clear Fermi cutoff in photoemission spectra and an anomalous enhancement in

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the electronic specific-heat coefficient at low temperatures. Recent band-structure calculations [4–9] consistently predicted that  $\text{Fe}_2\text{VAl}$  is a nonmagnetic semimetal having a deep pseudogap centred right at the Fermi level  $E_F$ . Indeed, nuclear magnetic resonance (NMR) [10, 11] and Hall-effect measurements [12] have confirmed the possession of a very low carrier concentration. Low-temperature specific-heat studies have also demonstrated a substantial decrease in the density of states (DOS) at  $E_F$  with the V substitution [13, 14]. More recently, the existence of a pseudogap of 0.1–0.2 eV in width has been experimentally confirmed in  $\text{Fe}_2\text{VAl}$  by optical conductivity measurements [15].

In order to deepen the understanding of the observed unusual transport properties of  $\text{Fe}_2\text{VAl}$ , it is of crucial importance to compare the results with those for systems with the substitution of M other than V. Indeed, electronic band-structure calculations have also been reported for  $\text{Fe}_2\text{TiAl}$  [7, 9]. Botton *et al* [9] noted great similarities in the overall shape of the DOS between  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_2\text{VAl}$ , including the possession of a deep pseudogap. However, it must be emphasized here that the Fermi level  $E_F$  sits in the middle of the sharp pseudogap in  $\text{Fe}_2\text{VAl}$  whereas  $E_F$  substantially shifts from the pseudogap in  $\text{Fe}_2\text{TiAl}$ . As a result,  $\text{Fe}_2\text{TiAl}$  has a very large DOS at  $E_F$ , indicating the possession of a metallic band structure, in sharp contrast to a semimetallic nature as in  $\text{Fe}_2\text{VAl}$ . As far as the low Ti composition range ( $0 \leq x \leq 0.15$ ) is concerned, the electrical resistivity at temperatures below 400 K in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  increases rapidly with increasing  $x$  in the same way as that found in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  [1]. It is, therefore, of great interest to study the resistivity behaviour of Ti-rich samples with  $x > 0.15$  including  $\text{Fe}_2\text{TiAl}$  ( $x = 0.33$ ), since the substantial difference in the electronic structure at the Fermi level is most likely reflected in the transport properties. The purpose of the present work is to investigate systematically the temperature dependence of electrical resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  with  $x = 0$ –0.33. The low-temperature specific heat was also measured for both  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_2\text{VAl}$  to allow a direct comparison of the DOS at the Fermi level between these two Heusler-type compounds. We also discuss why a large positive  $d\rho/dT$  dominates in the temperature range roughly below the Debye temperature for  $\text{Fe}_2\text{TiAl}$  on the basis of the Bloch–Grüneisen law.

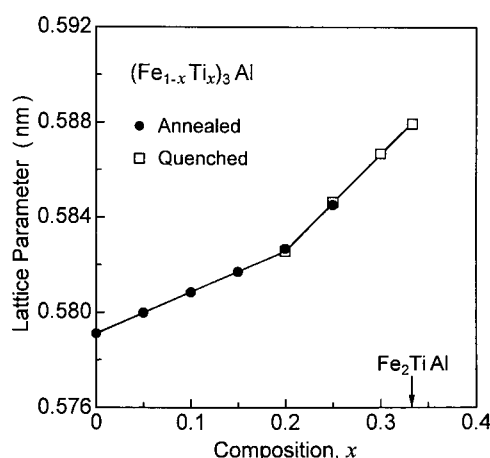
## 2. Experiment

### 2.1. Sample preparation

Ingots of  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  alloys with  $x = 0$ –0.33 were prepared by repeating arc-melting of appropriate amounts of 99.99% pure Fe and Al, and 99.9% pure Ti in an argon gas atmosphere. Since the weight loss after melting was less than 0.3%, the nominal composition was accepted as being accurate. The ingots were homogenized at 1273 K for more than 170 ks in vacuum. Samples were cut from the ingots with a SiC blade saw to the size of  $1 \times 1 \times 15 \text{ mm}^3$  for resistivity measurements, and  $7 \times 7 \times 15 \text{ mm}^3$  for specific heat measurements. Each sample was sealed in an evacuated quartz capsule and was annealed at 1273 K for 3.6 ks and then at 673 K for 14.4 ks followed by furnace cooling. This sample preparation is the same as employed previously for  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  [2, 12, 14]. Further, as will be discussed later, some of Ti-rich samples with  $x \geq 0.20$  were quenched into water after annealing at 1073 K for 18 ks.

### 2.2. Measurements

X-ray diffraction spectra were taken with Cu  $K\alpha$  radiation on powdered samples prepared as above. The electrical resistivity was measured by a standard d.c. four-terminal method with a current of 100 mA over the temperature range from 4.2 to 1373 K: the measurements at high



**Figure 1.** Lattice parameter as a function of Ti composition  $x$  in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$ . The solid circles and open squares represent the data on annealed and quenched samples, respectively.

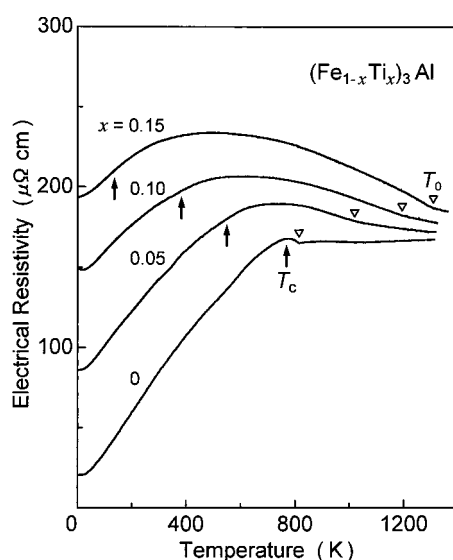
temperatures were carried out in a vacuum of  $4 \times 10^{-4}$  Pa with a rising rate of  $0.05 \text{ K s}^{-1}$ . The specific heat was measured by using the d.c. adiabatic method from 1.5 to 25 K. The Curie temperature was determined from thermomagnetic measurements using a superconducting quantum interference device (SQUID) magnetometer and also by means of differential thermal analysis.

### 3. Results

#### 3.1. X-ray diffraction analysis

The x-ray diffraction measurements could identify annealed samples of  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  to be in a single-phase  $\text{D0}_3$  structure in the composition range  $0 \leq x \leq 0.25$ . However, a small amount of an additional phase, probably the Laves phase ( $\text{Fe}_2\text{Ti}$ ), was detected for  $x \geq 0.30$ . The volume fraction of the Laves phase was estimated by comparing the peak intensities of x-ray diffraction and was found to be approximately 0.2 and 3.0 vol% for  $x = 0.30$  and 0.33, respectively. On the other hand, the  $\text{D0}_3$  single phase was maintained up to  $x = 0.30$  for quenched samples, while the amount of the Laves phase was reduced to only 1.1 vol.% for  $x = 0.33$  ( $\text{Fe}_2\text{TiAl}$ ).

The lattice parameter of the  $\text{D0}_3$  phase is plotted in figure 1 as a function of the Ti composition  $x$  in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$ . The lattice parameters of annealed samples shown by solid circles almost coincide with those of quenched samples ( $x = 0.20$  and  $0.25$ ) shown by open squares. The lattice parameter increases with increasing  $x$ , but the lattice dilatation is much smaller than that estimated from the Fe–Ti primary (bcc) solid solution [16]. This suggests that the substitution of Ti substantially contracts the lattice spacing as a result of the distinctive stabilization of the  $\text{D0}_3$  phase. The lattice parameter of the quenched  $\text{Fe}_2\text{TiAl}$  sample is equal to 0.588 nm, being in good agreement with the earlier report [17], in which the formation of  $\text{Fe}_2\text{TiAl}$  with the Heusler-type ( $\text{L2}_1$ ) structure has been confirmed by neutron diffraction. We believe that the transport properties of the quenched  $\text{Fe}_2\text{TiAl}$  sample would be practically unaffected by the presence of a very small amount of the unavoidable Laves phase.

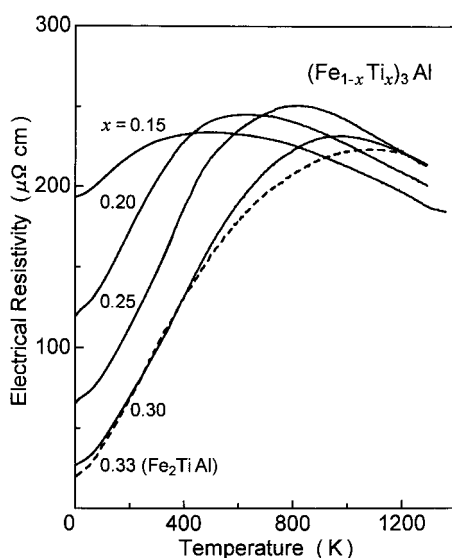


**Figure 2.** Temperature dependence of electrical resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  for low Ti compositions  $x = 0$ – $0.15$ . The arrows denoted by  $T_C$  indicate the Curie temperatures. The open triangles denoted by  $T_0$  indicate the  $\text{D0}_3$ – $\text{B2}$  transformation temperatures.

### 3.2. Electrical resistivity

Figure 2 shows the temperature dependence of the electrical resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  for low Ti compositions  $x = 0$ – $0.15$ . These curves were obtained on both heating and cooling runs, which almost coincided with each other. The arrow denoted by  $T_C$  indicates the Curie temperature. The value of  $T_C$  for  $x = 0.15$  was determined by the thermomagnetic measurements in magnetic fields up to 5 T, using a modified Arrott plot method, and was found to be 145 K. We see from figure 2 that, upon substitution of Ti for Fe, the electrical resistivity in the low temperature range increases rapidly but tends to saturate and forms a maximum at temperatures above  $T_C$  with the subsequent decrease with further increase in temperature. In parallel with a sharp decrease in  $T_C$ , the resistance maximum is shifted to a lower temperature. A similar resistance maximum has also been reported to occur in engineered alloys of  $\text{Fe}_3\text{Al}$  containing Ti and Mo [18]. The magnitude of the observed negative  $d\rho/dT$  at higher temperatures increases as the Ti composition  $x$  increases, which is consistent with our earlier report [1]. The negative  $d\rho/dT$  has also been observed for  $\text{Fe}_3\text{Ga}$ - [19] and  $\text{Fe}_3\text{Si}$ -based alloys [20] with the substitution of Ti for Fe. Also noted in figure 2 is that the resistivity curves show an inflection at temperatures above 800 K, as indicated by open triangles denoted by  $T_0$ . This inflection is known to be caused by the  $\text{D0}_3$ – $\text{B2}$  phase transformation and, hence, the transformation temperature can be precisely determined from it. The transformation temperature increases remarkably with increasing Ti composition at the rate of 50–55 K/at. %Ti, as already reported in previous investigations [1, 21–25].

Figure 3 shows the temperature dependence of the electrical resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  for high Ti compositions  $x = 0.15$ – $0.33$ : the curve for  $x = 0.15$  is the same as that shown in figure 2. Note here that the Arrott plots for  $x \geq 0.20$  did not show any evidence of a ferromagnetic transition, although the magnetizations measured under a magnetic field of 1.0 T were still enhanced at low temperatures at least down to 2 K. We believe, therefore, that

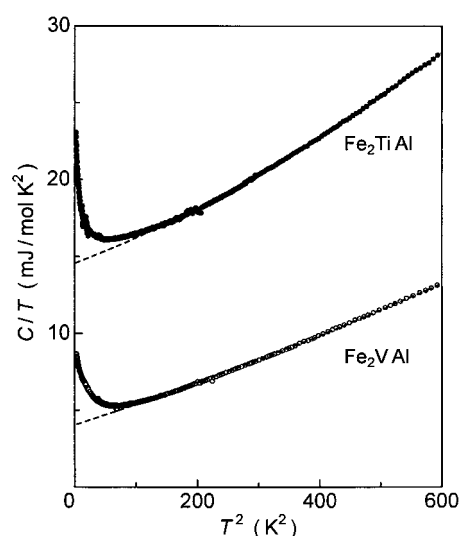


**Figure 3.** Temperature dependence of electrical resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  for high Ti compositions  $x = 0.15$ – $0.33$ . The resistivity curve for the quenched  $\text{Fe}_2\text{TiAl}$  sample ( $x = 0.33$ ) is shown by the dashed line.

Ti-rich alloys with  $x \geq 0.20$  are in a paramagnetic state over the whole temperature range examined. The resistivity curves were measured on annealed samples for  $0.15 \leq x \leq 0.25$  and on quenched samples for  $x = 0.30$  and  $0.33$ . It may be worthwhile mentioning that resistivity curves for quenched samples with  $x = 0.20$  and  $0.25$  were similar to those shown in figure 3 but their residual resistivities were always slightly higher than those in annealed ones owing to the existence of vacancies introduced by quenching. In striking contrast to the results in figure 2, the electrical resistivity decreases significantly at low temperatures as the Ti composition  $x$  exceeds 0.15, resulting in widening of the temperature range having a positive  $d\rho/dT$ . The resistivity curves of Ti-rich samples again clearly tend to saturate at higher temperatures and form a broad maximum for all samples studied. In particular, the Heusler-type  $\text{Fe}_2\text{TiAl}$  shows the smallest residual resistivity of only about  $20 \mu\Omega \text{ cm}$ , but a large positive  $d\rho/dT$  is responsible for an increase in resistivity up to above  $200 \mu\Omega \text{ cm}$  in the temperature range above about 800 K.

### 3.3. Specific heat

We measured the low-temperature specific heat of  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_2\text{VAl}$  in order to compare their DOSs at the Fermi level. In figure 4, the specific heat  $C$  is shown in the ordinary form of  $C/T$  against  $T^2$ . Note here that the present value of  $C$  is defined in units of per mole atom and should be multiplied by a factor of four when expressed in units of ‘chemical formula’ moles. It is seen that the specific heat of  $\text{Fe}_2\text{TiAl}$  is definitely higher than that of  $\text{Fe}_2\text{VAl}$  over the whole temperature range studied. Remarkably,  $C/T$  for  $\text{Fe}_2\text{TiAl}$ , as well as for  $\text{Fe}_2\text{VAl}$ , is found to be enhanced significantly with decreasing temperature below 10 K. We believe that the observed upturn in  $C/T$  at low temperatures is due to strong spin fluctuations associated with nonstoichiometry or with antisite defects, as suggested by Singh and Mazin [5]. The low-temperature upturn for  $\text{Fe}_2\text{VAl}$  has also been discussed in term of a Schottky anomaly arising from magnetic clusters [13].



**Figure 4.** Specific heat over temperature,  $C/T$ , versus  $T^2$  measured for  $\text{Fe}_2\text{TiAl}$  (dots) and  $\text{Fe}_2\text{VAl}$  (circles). The dashed curves represent a least squares fitting to equation (1).

Although the electronic specific-heat coefficient is strongly temperature dependent at low temperatures,  $C/T$  increases almost linearly with  $T^2$  in the temperature range above 10 K, as is clearly seen in figure 4. The specific heat at higher temperatures is believed to be composed of a combination of electronic and lattice contributions. We fit the data in the range 10 and 25 K to the ordinary equation

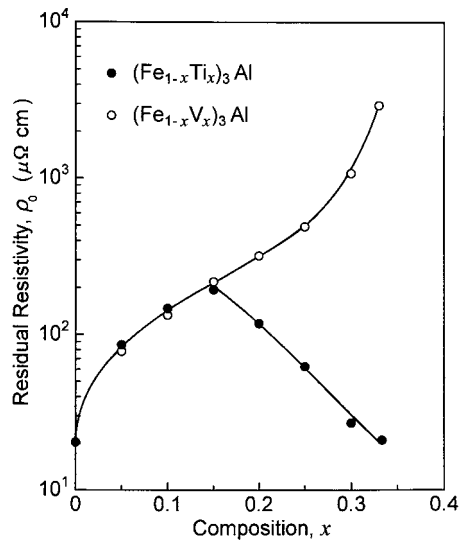
$$C = \gamma T + \alpha T^3 + \delta T^5 \quad (1)$$

where the first term represents the electronic specific heat and the remaining two the lattice contributions [26]. The electronic specific-heat coefficient  $\gamma$  was determined by a least squares fitting, as shown by the dotted curves in figure 4:  $\gamma = 12.7 \text{ mJ mol}^{-1} \text{ K}^{-2}$  for  $\text{Fe}_2\text{TiAl}$  and  $\gamma = 3.9 \text{ mJ mol}^{-1} \text{ K}^{-2}$  for  $\text{Fe}_2\text{VAl}$ . The measured value of  $\gamma$  for  $\text{Fe}_2\text{VAl}$  is very close to that deduced from the electronic specific-heat data at 6 K [14], but is still larger than that reported by Lue *et al* [13]. Note that the electronic specific-heat coefficient deduced from the band calculations for  $\text{Fe}_2\text{VAl}$  is only  $0.2 \text{ mJ mol}^{-1} \text{ K}^{-2}$  [4] and is much smaller than the experimental value because of the mass enhancement effect discussed previously [2, 14]. Similarly we found that the measured value of  $\gamma$  for  $\text{Fe}_2\text{TiAl}$  is larger by a factor of three or four than the calculated one [7, 9]. Apart from this discrepancy, we could confirm experimentally from the specific heat measurements that the DOS at the Fermi level,  $N(E_F)$ , of  $\text{Fe}_2\text{TiAl}$  is about three times as high as that of  $\text{Fe}_2\text{VAl}$  as a result of the shift of the Fermi level off from the centre of the pseudogap.

The least squares fitting also determines the lattice specific-heat coefficient,  $\alpha$ , which can be related to the Debye temperature  $\theta_D$  as follows:

$$\theta_D = (12\pi^4 R/5\alpha)^{1/3} \quad (2)$$

where  $R$  is the gas constant. We obtained  $\theta_D = 425 \text{ K}$  for  $\text{Fe}_2\text{TiAl}$  and  $\theta_D = 510 \text{ K}$  for  $\text{Fe}_2\text{VAl}$ . The value of  $\theta_D$  for  $\text{Fe}_2\text{VAl}$  agrees well with that extrapolated from those for  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  in our previous study [14]. Both the  $\theta_D$  values for  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_2\text{VAl}$  are higher than that for  $\text{Fe}_3\text{Al}$ , i.e.  $\theta_D = 377 \text{ K}$  [14], suggesting a substantial enhancement of the atomic bonding in the Heusler structure.



**Figure 5.** Residual resistivity  $\rho_0$  as a function of Ti and V compositions  $x$  in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  (solid circles) and  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  (open circles), respectively.

## 4. Discussion

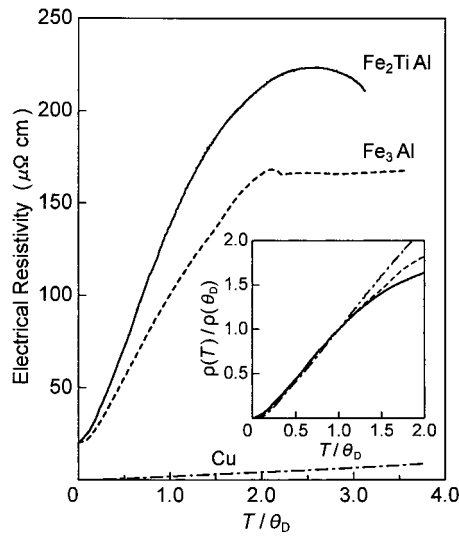
### 4.1. Saturation effect in the temperature dependence of resistivity

We discuss first why the resistivity tends to saturate and a negative  $d\rho/dT$  occurs at high temperatures. Indeed, this is a quite universal feature observed for many crystalline and amorphous alloys and intermetallic compounds, and a positive  $d\rho/dT$  switches to a negative one when the resistivity exceeds about  $200 \mu\Omega$  cm. This phenomenon is often referred to as Mooij's criterion [27]. As has been discussed by Mizutani [28], an increase in resistivity with increasing temperature is brought about by the reduction in the electron mean free path and the Boltzmann transport mechanism fails when its value becomes comparable to the interatomic distance. This condition is generally fulfilled when the resistivity reaches about  $150$ – $200 \mu\Omega$  cm. As will be discussed further in section 4.3, we believe that the mechanism for the appearance of a positive  $d\rho/dT$  at temperatures, say, below the Debye temperature is caused by a decreasing mean free path due to increasing electron–phonon interaction with increasing temperature. This means that the saturation effect observed in the present alloys can be taken as the termination of the mean free path effect. The saturation effect does not necessarily mean the occurrence of a negative  $d\rho/dT$  at the highest temperature range where the measuring temperature far exceeds the Debye temperature. Here the elastic scattering of electrons with ions is expected to dominate in the same way as in liquid metals, where the occurrence of a negative  $d\rho/dT$  is often discussed in relation to the Ziman theory [29]. Of course, we cannot directly apply the Ziman theory to a crystal metal as in the present case. Further discussion on the occurrence of a negative  $d\rho/dT$  in a crystal at very high temperatures like 1200 K is still premature at this stage.

### 4.2. Composition dependence of residual resistivity

The residual resistivity  $\rho_0$  is shown in figure 5 as a function of the Ti composition  $x$  in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$ : the logarithm of resistivity is plotted on the ordinate. Also shown in this





**Figure 6.** Electrical resistivity as a function of the temperature which is normalized by the respective Debye temperatures,  $T/\theta_D$ , for  $\text{Fe}_2\text{TiAl}$  (solid curve),  $\text{Fe}_3\text{Al}$  (dashed curve) and pure Cu (dash-dotted curve). The inset shows the relation between  $\rho(T)/\rho(\theta_D)$  and  $T/\theta_D$  for the three systems:  $\rho(T)$  represents the temperature-dependent electron-phonon contribution. The Debye temperatures for  $\text{Fe}_2\text{TiAl}$ ,  $\text{Fe}_3\text{Al}$  and pure Cu are chosen to be 425, 377 and 342 K, respectively.

figure are the  $\rho_0$  values for  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  taken from our data reported earlier [12, 14]. In the composition range  $0 \leq x \leq 0.15$ , the residual resistivity increases with increasing  $x$  in the same manner as that for the V substitution. However, the  $\rho_0$  value turns out to decrease sharply for  $x > 0.15$  and is reduced to only  $20 \mu\Omega \text{ cm}$  at  $x = 0.33$  ( $\text{Fe}_2\text{TiAl}$ ). In contrast, the residual resistivity in  $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$  increases continuously with increasing  $x$  and reaches about  $3000 \mu\Omega \text{ cm}$  at  $x = 0.33$  ( $\text{Fe}_2\text{VAl}$ ). We can attribute the observed large difference in the resistivity behaviour between  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_2\text{VAl}$  to the difference in the DOS at the Fermi level, as discussed in section 3.3.

It should be remarked in figure 5 that the  $x$ -dependence of the residual resistivity in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  exhibits a maximum at  $x = 0.15$ . As discussed by Weinert and Watson [7], the presence of a hybridization pseudogap may be regarded as a common feature in transition-metal aluminides including both  $\text{Fe}_2\text{VAl}$  and  $\text{Fe}_2\text{TiAl}$ . They suggested that the Fermi level falls in the pseudogap when there are five 3d electrons per transition-metal atom, i.e. 15 3d electrons in the three transition-metal spheres for the present Heusler-type compounds. In fact, since the total number of 3d electrons is equal to 15 for  $\text{Fe}_2\text{VAl}$ , the Fermi level falls right at the centre of the pseudogap. However, the total number of 3d electrons is equal to 14 for  $\text{Fe}_2\text{TiAl}$ . Hence, the Fermi level is shifted off from the centre of the pseudogap and, according to the band calculations [7, 9], it is located at an energy approximately 0.5 eV below the pseudogap. A simple manipulation yields a total of exactly 15 3d electrons per chemical formula unit when  $x = 0.25$ . This would roughly explain why the residual resistivity exhibits a maximum at an intermediate Ti composition. Finally, it is noted that a similar Ti composition dependence of the residual resistivity has been observed in  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Ga}$  [19].

#### 4.3. Temperature dependence of resistivity in $\text{Fe}_2\text{TiAl}$

As emphasized earlier, we have observed a large positive  $d\rho/dT$  at temperatures below approximately the Debye temperature in the present  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  alloys. The electrical

resistivity data for the three representative systems  $\text{Fe}_2\text{TiAl}$ ,  $\text{Fe}_3\text{Al}$  and pure Cu are plotted in figure 6 as a function of the temperature, which is normalized with respect to the respective Debye temperatures,  $T/\theta_D$ . It is seen that the value of  $d\rho/dT$  for  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_3\text{Al}$  is much larger than that of Cu, which is shown by the dash-dotted curve. Moreover, the saturation effect can be clearly observed for both  $\text{Fe}_3\text{Al}$  and  $\text{Fe}_2\text{TiAl}$ , the origin of which has been discussed in section 4.1. First, the temperature-independent impurity scattering contribution is subtracted from the measured total resistivity. The resulting  $\rho(T)$  data represent the temperature-dependent electron-phonon contribution and may be well analysed by using the well known Bloch-Grüneisen law [29]. The inset to figure 6 shows the relation between  $\rho(T)/\rho(\theta_D)$  and  $T/\theta_D$ . Surprisingly, all the curves coincide quite well in the temperature range  $T \leq \theta_D$ , whereas the data in the range  $T > \theta_D$  for  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_3\text{Al}$  show strong downward deviation from a linear dependence observed for Cu due to the saturation effect discussed above. The inset to figure 6 implies that the electron-phonon contribution to the resistivity in the range  $T \leq \theta_D$  is essentially the same irrespective of the alloy systems. It is also interesting to note that the Bloch-Grüneisen law holds equally well for both ferromagnetic  $\text{Fe}_3\text{Al}$  and paramagnetic  $\text{Fe}_2\text{TiAl}$ , suggesting the electron-magnon interaction to be of minor importance.

We are now ready to consider why a large positive  $d\rho/dT$  occurs for  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_3\text{Al}$ , as is seen in figure 6. As is clear from the argument above, we can equally address the question to why  $\rho(\theta_D)$  for  $\text{Fe}_2\text{TiAl}$  is much larger than that for Cu. In general, the electrical resistivity of an isotropic metallic system can be expressed as follows:

$$\frac{1}{\rho} = \frac{e^2}{3} \Lambda_F v_F N(E_F) \quad (3)$$

where  $\Lambda_F$  is the mean free path and  $v_F$  is the Fermi velocity. Here the product of  $v_F N(E_F)$  is purely electronic in origin and has very little to do with the temperature dependence of resistivity, which, instead, obviously arises from that of the mean free path. As discussed above in relation to the inset to figure 6,  $d\Lambda_F^{-1}/dT$  is essentially the same among the three systems shown there. According to equation (3), we see that the magnitude of  $d\rho/dT$  is biased by the magnitude of the coefficient  $v_F N(E_F)$ . A free-electron-like large value of  $v_F$  for Cu is definitely responsible for the possession of a low  $d\rho/dT$  as shown in figure 6. In contrast, the value of  $v_F$  must be small enough to enhance  $d\rho/dT$  for  $\text{Fe}_2\text{TiAl}$  and  $\text{Fe}_3\text{Al}$  because of the location of the Fermi level in the 3d-transition-metal band. The averaged Fermi velocity may be evaluated by taking the derivative of the energy dispersion curve with respect to the wavevector in different directions at the Fermi energy and averaging them over the Fermi surface. Such band calculations are now in progress in order to calculate the product of  $v_F N(E_F)$  in the Heusler-type  $\text{Fe}_2\text{TiAl}$  compound.

## 5. Conclusions

The temperature dependence of electrical resistivity has been investigated for  $(\text{Fe}_{1-x}\text{Ti}_x)_3\text{Al}$  alloys with Ti compositions  $x = 0-0.33$ . In the low Ti composition range ( $0 \leq x \leq 0.15$ ), the electrical resistivity increases rapidly at low temperatures with increasing  $x$ , showing a resistance maximum at temperatures above  $T_C$ . In contrast, samples with Ti compositions  $x = 0.20-0.33$  are in a paramagnetic state and exhibit a rapid decrease in the low-temperature resistivity with increasing  $x$ . In particular, the Heusler-type  $\text{Fe}_2\text{TiAl}$  shows the smallest residual resistivity of only about  $20 \mu\Omega \text{ cm}$ , but a large positive  $d\rho/dT$  is responsible for an increase in resistivity above  $200 \mu\Omega \text{ cm}$  at high temperatures, where the resistivity saturates and a negative  $d\rho/dT$  occurs in accord with the Mooij criterion. By analysing the  $\rho(T)$  data on the

basis of the Bloch–Grüneisen law, we could point out that the Fermi velocity must be small enough to cause a large positive  $d\rho/dT$  for  $\text{Fe}_2\text{TiAl}$ .

The resistivity behaviour of  $\text{Fe}_2\text{TiAl}$  mentioned above is in sharp contrast to a closely related system  $\text{Fe}_2\text{VAl}$ , which shows a semiconductor-like behaviour with a resistivity reaching  $3000 \mu\Omega \text{ cm}$  at 2 K. The observed large difference in the resistivity behaviour can be attributed to the fact that  $\text{Fe}_2\text{TiAl}$  possesses a much higher DOS at the Fermi level than  $\text{Fe}_2\text{VAl}$ , as confirmed by the low-temperature specific-heat measurements. Thus, the present results strengthen our previous conclusion that unique transport properties observed for  $\text{Fe}_2\text{VAl}$  originate from the possession of low carrier concentrations as a result of the location of the Fermi level at the centre of the deep pseudogap in the presence of spin fluctuation effects.

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