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2000 J. Phys.: Condens. Matter 12 1769

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Electronic, magnetic and transport properties of $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ alloys

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Received 10 September 1999

Abstract. We report electronic, magnetic and transport properties of $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ alloys with $x = 0-0.38$ and analyse the results on the basis of the measured electronic specific-heat coefficient, the Debye temperature and the magnetic stiffness constant. As the V composition increases, the electrical resistivity increases rapidly at low temperatures and the magnetization decreases significantly in parallel with a sharp reduction in the Curie temperature. In particular, the Heusler-type Fe_2VAl compound ($x = 0.33$) 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. Low-temperature specific heat studies demonstrate a substantial decrease in carrier concentration with the V substitution, being consistent with recent band calculations, which predict that Fe_2VAl is a nonmagnetic semimetal with a sharp pseudogap at the Fermi level. A large mass enhancement deduced from the electronic specific-heat measurements suggests that Fe_2VAl is a possible candidate for a 3d heavy-fermion system. The unusual electron transport is mainly attributed to the effect of strong spin fluctuations, in addition to the existence of very low carrier concentrations.

1. Introduction

The intermetallic compound Fe_3Al is a well ordered ferromagnet with a D0_3 crystal structure. When Fe atoms in Fe_3Al are partly replaced by other 3d transition elements, the electrical resistivity increases rapidly at low temperatures and shows an anomalous temperature dependence [1, 2]. As reviewed by Nishino [3], these alloys possess common features characterized by (1) a resistance maximum near the Curie point T_C and (2) a negative resistivity slope at high temperatures above T_C up to 1000 K and above. The substitution of 3d elements to the left of Fe in the periodic table seems to be responsible for the occurrence of the negative temperature dependence of resistivity. Since the substitution of Ti, V, Cr and Mn always causes a sharp reduction in T_C and in magnetization, the anomalous resistance behaviour could be attributed to a weakening of ferromagnetism compelled by the substituents.

The most spectacular feature of the resistance anomaly has been found for $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ [1–3] as well as for $(\text{Fe}_{1-x}\text{V}_x)_3\text{Si}$ [4] and $(\text{Fe}_{1-x}\text{V}_x)_3\text{Ga}$ [5]. In particular, the Heusler-type Fe_2VAl compound ($x = 0.33$) exhibits a semiconductor-like behaviour with the resistivity reaching $3000 \mu\Omega \text{ cm}$ at 2 K [2, 3]. Furthermore, as will be discussed later, the Fe_2VAl compound is proved to be in a marginally magnetic state. In this regard, Fe_2VAl shows an apparent similarity to a nonmagnetic narrow-gap semiconductor, FeSi [6], which has been

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classified as a unique d-electron system among the family of Kondo insulators [7]. The anomaly in FeSi appears only at low temperatures below 200 K, while the resistivity of Fe₂VAl continues to decrease with temperature up to 1200 K and above. In order to create a framework for understanding the unusual transport properties, electronic structure calculations have been reported quite recently [8–11], all of which consistently predict that Fe₂VAl is a nonmagnetic semimetal with a narrow pseudogap at the Fermi level. Moreover, measurements of nuclear magnetic resonance (NMR) [12] and Hall effect [13] strongly support that Fe₂VAl is a low-carrier-concentration semimetal. While a clear Fermi cutoff was observed in valence-band photoemission spectra [2, 14], the presence of the pseudogap has been experimentally confirmed in optical conductivity spectra [15]. The unusual electron transport properties are most likely attributable to the enhanced localization tendency of a small number of carriers in the pseudogap of the marginally magnetic Fe₂VAl compound.

Despite the possession of a very low carrier concentration, low-temperature specific-heat measurements for Fe₂VAl revealed a large electronic specific-heat coefficient $\gamma = 14 \text{ mJ mol}^{-1} \text{ K}^{-2}$ [2]. Remarkably, plots of the specific heat over temperature, C/T , versus T^2 show an anomalous upturn with decreasing temperature, suggesting a large mass enhancement as commonly found in f-electron heavy-fermion systems [16]. It is of great interest to study the effect of the V substitution on the density of states at the Fermi level in (Fe_{1-x}V_x)₃Al alloys. The low-temperature specific heat measurements certainly provide useful information, since the electronic, magnetic and lattice contributions to the specific heat can be well separated into each component by differentiating their temperature dependence. In the present study, we report the experimental studies on the electronic and magnetic properties of (Fe_{1-x}V_x)₃Al alloys including the Heusler-type Fe₂VAl and analyse the results on the basis of the measured electronic specific-heat coefficient, the Debye temperature and the spin-wave stiffness constant. In particular, we discuss the V composition dependence of the electronic specific heat with reference to the magnetic states deduced from magnetic measurements.

2. Experiment

2.1. Sample preparation

The pseudobinary alloys (Fe_{1-x}V_x)₃Al were prepared by repeated melting of appropriate mixtures of 99.99% pure Fe and Al, and 99.7% pure V in an argon arc furnace. 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 ingot with a SiC blade saw to the size of $1 \times 1 \times 15 \text{ mm}^3$ for resistivity measurements, and to the size of $6 \times 6 \times 18 \text{ mm}^3$ for specific-heat measurements. Each sample, sealed in an evacuated quartz capsule, was held at 1273 K for 3.6 ks and then annealed for the D0₃ ordering at 673 K for 14.4 ks followed by furnace cooling.

2.2. X-ray analysis

X-ray diffraction spectra were taken with Cu $K\alpha$ radiation on powdered samples prepared as above. The formation of the D0₃ single phase was confirmed in the V composition range $0 \leq x \leq 0.38$. The lattice parameter first decreases with increasing x but turns to increase above $x = 0.33$. The V composition of $x = 0.33$ at which the lattice parameter reaches a minimum is very close to the constitution of the Heusler-type Fe₂VAl compound [2, 3]. This L2₁-ordered structure is known to be formed when Fe and V atoms separate on the two transition-metal sublattices. Taking the site preference into consideration, the

degree of long-range order can be determined by comparing the superlattice and fundamental peak intensities of x-ray diffraction. It has been found that the degree of order, S , increases monotonically with increasing x from $S \approx 0.8$ at $x = 0$ reaching $S \approx 1.0$ near $x = 0.33$ [13]. The above results demonstrate that the V substitution substantially contributes to the stabilization of the D0_3 phase in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$.

2.3. Measurements

The electrical resistivity was measured by a standard dc four-terminal method with a current of 100 mA over the temperature range 2–1273 K and with a rising rate of 0.03 K s^{-1} . The measurements at high temperatures were carried out in a vacuum of $6 \times 10^{-4} \text{ Pa}$. The low-temperature specific heat was measured from 1.5 to 6 K using a dc adiabatic method. The magnetization was measured from 2 to 300 K in magnetic fields up to 5 T using a superconducting quantum interference device (SQUID) magnetometer. The Curie temperature was also determined by means of differential thermal analysis.

3. Results and discussion

3.1. Electrical resistivity

Figure 1 shows the temperature dependence of electrical resistivity in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with V compositions $x = 0$ –0.38: the logarithm of resistivity is plotted on the ordinate. The arrows denoted by T_C indicate the Curie temperatures determined by using thermomagnetic measurements (section 3.2) and differential thermal analysis. Fe_3Al ($x = 0$) shows a metallic behaviour with the residual resistivity of about $20 \mu\Omega \text{ cm}$, and a relatively large residual resistivity may be simply attributed to the Fe–Al site disorder [17]. When Fe atoms are replaced by V atoms, the electrical resistivity increases rapidly at low temperatures, forming a maximum at or near T_C and, above T_C , decreases dramatically with rising temperature. The resistance maximum can be clearly seen for the samples up to $x = 0.30$, and the temperature corresponding to the resistance maximum coincides well with the Curie temperature T_C , as shown by the arrows. As the V composition increases, the temperature region having the negative resistivity slope expands markedly in parallel with a sharp reduction in T_C , so that the electrical resistivity becomes strongly dependent on temperature.

The resistance maximum disappears for Fe_2VAl ($x = 0.33$) and the resistivity continues to increase down to the lowest temperature of 2 K. Remarkably the resistivity for $x = 0.33$ and 0.38 reaches about 3000 and 9000 $\mu\Omega \text{ cm}$, respectively, which is two or three orders of magnitude higher than that of ordinary 3d transition-metal alloys. In other words, the Heusler-type Fe_2VAl compound exhibits a seemingly semiconductor-like behaviour over the whole temperature range up to 1200 K and above. The $\ln \rho$ versus $1/T$ plots (ρ , resistivity; T , temperature) for the data on $x = 0.33$ and 0.38 become almost linear in the temperature interval 400–800 K, and the energy gap of 0.10–0.13 eV was deduced from the slope. A similar thermal excitation behaviour has been confirmed by NMR measurements [12], which yield an energy gap of about 0.27 eV. We consider the thermal excitation to originate from the presence of a pseudogap at the Fermi level in Fe_2VAl , as predicted by the band calculations [8–11] and confirmed in optical conductivity spectra [15].

Such a semiconductor-like resistivity has also been reported for Fe_2VGa by Endo *et al* [18, 19], who have extended the earlier work of Kawamiya *et al* [5] and discussed the observed giant magnetoresistance of 30–50% in terms of the metal–insulator transition. More recently, Matsushita and Yamada [20] have examined the effect of heat treatment on the electrical

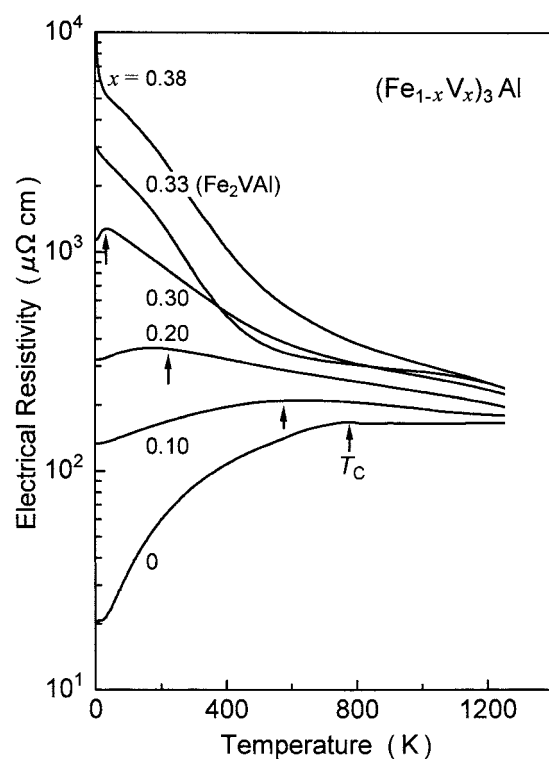


Figure 1. Temperature dependence of electrical resistivity in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with $0 \leq x \leq 0.38$. The arrows indicate the Curie temperatures T_C .

resistivity of Fe_2VAl : the semiconductor-like behaviour is still observed for a quenched sample but is surprisingly suppressed in a slowly cooled sample. This is apparently at variance with a dramatic increase in the low-temperature resistivity of the well annealed Fe_2VAl [2].

3.2. Magnetic properties

Figure 2 shows the temperature dependence of the magnetization measured under a magnetic field of 1.0 T for $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with $x = 0$ –0.38: the logarithm of magnetization is plotted on the ordinate. In the V composition range $0 \leq x \leq 0.30$, the magnetization versus applied magnetic fields up to 1.0 T below the Curie temperature is almost saturated in a manner characteristic of ordinary ferromagnets. The Curie temperature T_C was determined by the thermomagnetic measurements using a modified Arrott-plot method [21]. The saturation magnetization, as well as T_C , decreases significantly with increasing x , so that the magnetization for Fe_2VAl ($x = 0.33$) is three orders of magnitude smaller than that for Fe_3Al ($x = 0$). The magnetizations for $x = 0.33$ and 0.38 are still enhanced at low temperatures although the Arrott plots do not show any evidence of a ferromagnetic transition. Further, the variation of the magnetic susceptibility for Fe_2VAl cannot be fitted to the Curie–Weiss behaviour, being consistent with the recent work [20], whereas, according to Webster and Ziebeck [22], the magnetization is proportional to an applied magnetic field and the magnetic susceptibility can be explained by the Curie–Weiss law with a temperature-independent term.

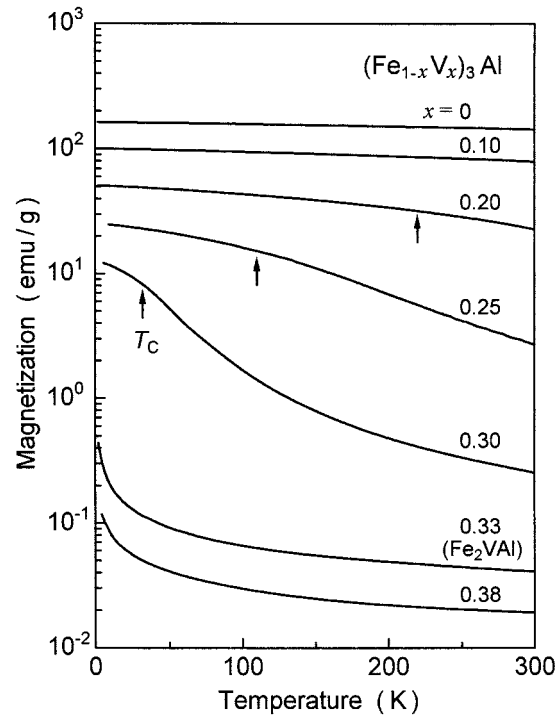


Figure 2. Temperature dependence of magnetization in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with $0 \leq x \leq 0.38$. The arrows indicate the Curie temperatures T_C .

Popiel *et al* [23] have reported that there is no long-range magnetic order in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with $x > 0.20$ and that the superparamagnetic states persist down to 4.2 K. However, our magnetization measurements clearly demonstrate that $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ alloys are ferromagnetic at least up to $x = 0.30$. The temperature dependence of the magnetization below T_C may be described in terms of the well known Bloch law:

$$M(T)/M(0) = 1 - BT^{3/2} \quad (1)$$

where $M(0)$ is the saturation magnetization at 0 K. Then the spin-wave stiffness constant, D , is deduced by fitting the magnetization curve to equation (1) and applying the following expression:

$$B = [2.61g \mu_B/M(0)](k_B/4\pi D)^{3/2} \quad (2)$$

where g is the g -factor equal to 2, μ_B is the Bohr magneton and k_B is the Boltzmann constant. We have found that $D = 122 \text{ meV } \text{\AA}^2$ for Fe_3Al ($x = 0$), being in good agreement with that measured by using neutron spectrometry [24]. Figure 3 shows (a) the average magnetic moment per Fe atom, μ_{Fe} , (b) the Curie temperature, T_C , and (c) the spin-wave stiffness constant, D , as a function of the V composition x . The magnetic moment μ_{Fe} is determined by extrapolating the saturation magnetization data down to 0 K. The value of T_C decreases almost linearly with increasing x and goes to zero in the vicinity of $x = 0.33$. Thus the Heusler-type Fe_2VAl compound is found to be on the verge of magnetic ordering. The magnetic moment μ_{Fe} decreases more sharply than a simple dilution of Fe moments by V atoms carrying no magnetic moment. We consider that this accelerated decrease occurs because the substituting V atom affects many neighbouring Fe atoms simultaneously. Similarly, the

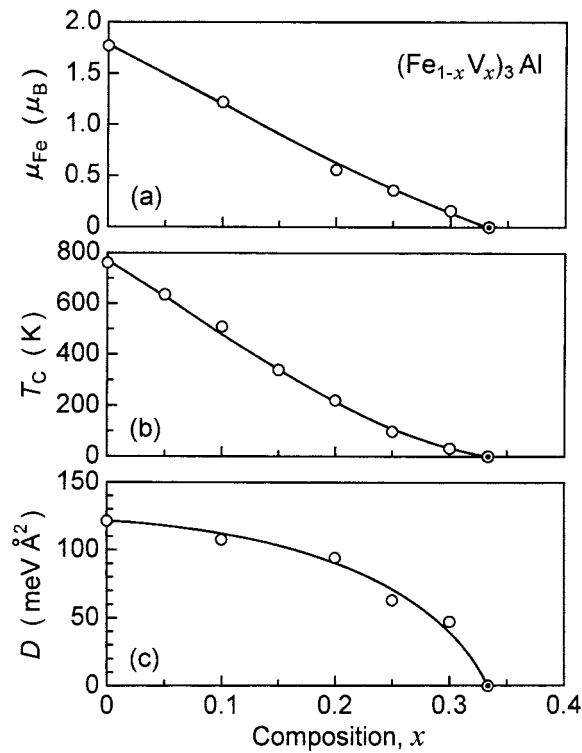


Figure 3. Magnetic properties of $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ alloys: (a) average magnetic moment per Fe atom, μ_{Fe} , (b) Curie temperature, T_{C} , and (c) spin-wave stiffness constant, D , as a function of V composition x .

spin-wave stiffness constant, D , first decreases moderately but diminishes rapidly near the composition of Fe_2VAl ($x = 0.33$). It is seen that the composition dependence of D is very different from that of T_{C} , indicating the absence of a linear relationship between D and T_{C} , in contrast to the prediction by the Heisenberg model. This is, we consider, reasonable, since the distance between nearest Fe atoms increases significantly with the V substitution and the ferromagnetism in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ would not be explained simply by taking into account the nearest-neighbour exchange interaction.

3.3. Specific heat

In figure 4, the specific heat C measured on $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with $x = 0-0.38$ is shown in the ordinary form of C/T against T^2 . Note here that the present C value is defined in per mole atom and should be multiplied by a factor of four when expressed in terms of ‘chemical formula’ moles. In the V composition range $0 \leq x \leq 0.30$, a downward deviation of the specific heat is evidently present at low temperatures, owing to the spin-wave contribution characteristic of ferromagnetic alloys [25]. It is found that the measured specific heat decreases significantly with increasing V composition. Unlike the ordinary case for $x = 0-0.30$, an anomalous upturn in C/T can be observed for Fe_2VAl ($x = 0.33$), which is reminiscent of heavy-fermion systems and is most likely related to the spin fluctuations [2, 3]. It is noted that the specific heat data on $x = 0.38$ also exhibit an anomaly similar to that for Fe_2VAl .

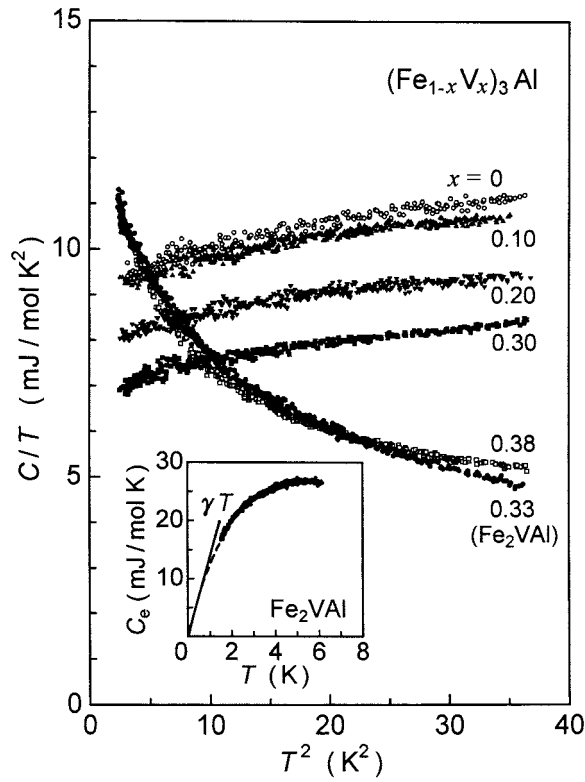


Figure 4. Specific heat over temperature, C/T , versus T^2 measured for $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ with $0 \leq x \leq 0.38$. The inset shows the electronic specific heat C_e versus T plots for Fe_2VAl ($x = 0.33$). The electronic specific heat is approximately of the form $C_e = \gamma T$ for $T \rightarrow 0$.

The low-temperature specific heat, C , of a ferromagnetic metal is generally expressed as

$$C = \gamma T + \alpha T^3 + \beta T^{3/2} \quad (3)$$

where γ , α and β represent the electronic, lattice and spin-wave specific-heat coefficients, respectively. The specific heat data on the ferromagnetic $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ alloys with $x = 0-0.30$ can be analysed by using equation (3). However, it is difficult to determine the three coefficients simultaneously by the method of least squares. We deduce the value of β independently from the aforementioned spin-wave stiffness constant, D , through the relation,

$$\beta = 39.43 (W/d) D^{-3/2} \quad (4)$$

where W is the average atomic weight in g and d is the measured mass density in g cm^{-3} . Once the value of β is given, the remaining coefficients, α and γ , are reliably determined by a least-squares fitting.

The lattice specific-heat coefficient, α , can be related to the Debye temperature θ_D as follows:

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

where R is the gas constant. We thus obtain $\theta_D = 377$ K for Fe_3Al , which agrees fairly well with that derived from measurements of the heat capacity [26] and the thermal expansion [27]. Figure 5 shows the Debye temperature θ_D as a function of the V composition x . It can be seen

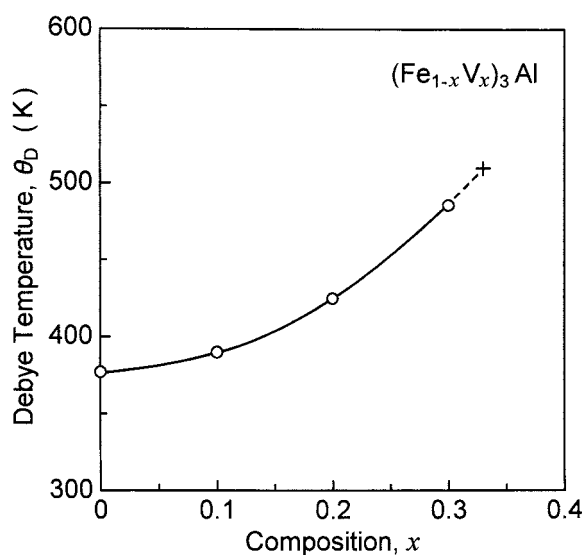


Figure 5. Debye temperature θ_D as a function of V composition x in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$. Also shown is the extrapolated θ_D value (+) for Fe_2VAl ($x = 0.33$).

that the Debye temperature increases with increasing x , suggesting a substantial enhancement of the atomic bonding in the D0_3 structure. Furthermore, the Debye temperature for Fe_2VAl is reasonably extrapolated to $\theta_D = 510$ K at $x = 0.33$ in figure 5.

The Heusler-type Fe_2VAl compound exhibits the anomalous upturn in C/T with decreasing temperature as shown in figure 4. Since the electronic specific-heat coefficient is strongly dependent on temperature, the data can no longer be analysed in the ordinary manner. From equation (5), the Debye temperature θ_D estimated for Fe_2VAl yields the lattice specific-heat coefficient, $\alpha = 0.013 \text{ mJ mol}^{-1} \text{ K}^{-4}$. Therefore, the electronic specific heat C_e ($\equiv \gamma T$) can be obtained by subtracting the lattice specific-heat contribution, αT^3 , from the measured C , and the C_e versus T plots for Fe_2VAl are shown in the inset of figure 4. The electronic specific-heat coefficient, γ , may be evaluated by extrapolating the C_e versus T data to absolute zero and applying the linear relation $C_e = \gamma T$ for $T \rightarrow 0$. The limiting γ value for Fe_2VAl is about $14 \text{ mJ mol}^{-1} \text{ K}^{-2}$, being in agreement with our previous result [2] where the lattice contribution was ignored.

Figure 6 shows the electronic specific-heat coefficient, γ ($\equiv C_e/T$), of $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ as a function of the V composition x . The present γ value for Fe_3Al ($x = 0$) is $9.3 \text{ mJ mol}^{-1} \text{ K}^{-2}$, being consistent with that previously published [28, 29], but a slight difference in γ may arise from the influence of the degree of order [29]. The γ value for Fe_3Al appears to be larger than that for typical transition metals, e.g. $\gamma = 5.0 \text{ mJ mol}^{-1} \text{ K}^{-2}$ for Fe [28]. It may be worthwhile mentioning that the value of γ for binary Fe–Al alloys is sharply enhanced at Al contents above 20 at.% [28, 29], which is apparently accompanied by a more rapid decrease in the magnetic moment than that expected from the simple dilution rule [30]. Also plotted in figure 6 are the values of C_e/T (open squares) obtained at the highest measuring temperature of 6 K. In the V composition range $0 \leq x \leq 0.30$, the γ value is in good agreement with C_e/T ($T = 6$ K), indicating that the γ value is temperature independent as in ordinary metals. A decrease in γ demonstrates a substantial decrease in the density of states at the Fermi level with increasing V composition. However, the γ value for Fe_2VAl ($x = 0.33$) is significantly dependent on temperature and its limiting value extrapolated to 0 K abruptly rises to a large

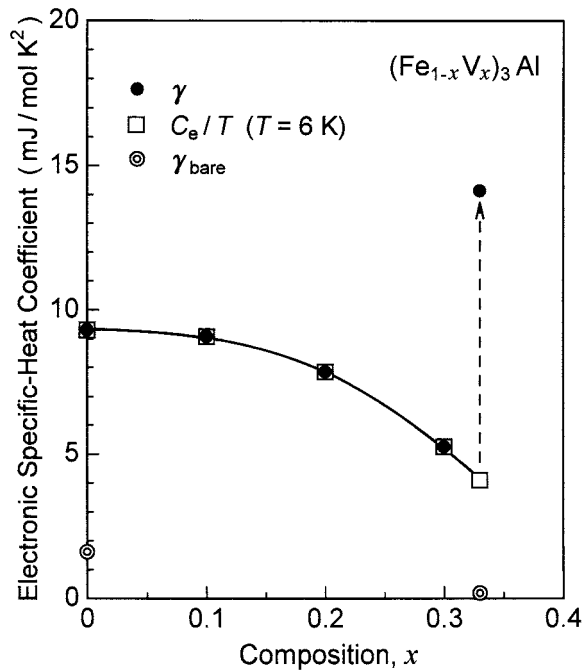


Figure 6. Electronic specific-heat coefficient of $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ as a function of V composition x . The measured γ and C_e/T ($T = 6$ K) are represented by solid circles and open squares, respectively, and the bare-band γ_{bare} values derived from the band calculations [8] are represented by double circles.

value of $14 \text{ mJ mol}^{-1} \text{ K}^{-2}$, in contrast to a small value of C_e/T ($T = 6$ K). A further decrease in C_e/T may be expected for Fe_2VAl , since the specific heat of heavy-fermion compounds is known to exhibit a broad peak [31] when plotted as in the inset to figure 4.

The measured electronic specific-heat coefficient, γ is generally larger than that derived from the band calculations. The value of γ can be related to the density of states at the Fermi level, $N(E_F)$, as follows:

$$\gamma = \frac{\pi^2}{3} k_B^2 N(E_F) (1 + \lambda_{\text{ep}} + \lambda_{\text{sf}}). \quad (6)$$

where λ_{ep} and λ_{sf} are electron-phonon coupling and spin-fluctuation enhancement, respectively. Using $N(E_F)$ derived from the band calculations [8], the bare-band electronic specific-heat coefficients, γ_{bare} corresponding to $\lambda_{\text{ep}} = \lambda_{\text{sf}} = 0$, can be obtained for Fe_3Al and Fe_2VAl and are plotted by double circles in figure 6. The measured γ value for Fe_3Al is approximately five times larger than γ_{bare} so that $\lambda_{\text{sf}} \approx 4$ if $\lambda_{\text{ep}} \approx 0$. The decrease in C_e/T ($T = 6$ K) with increasing x is in line with a reduction in λ_{bare} for Fe_2VAl , although their absolute values are considerably different from each other. Accordingly we conclude that the carrier concentration in $(\text{Fe}_{1-x}\text{V}_x)_3\text{Al}$ substantially decreases with increasing V composition.

It should be remarked in figure 6 that the calculated γ_{bare} value for Fe_2VAl is only $0.19 \text{ mJ mol}^{-1} \text{ K}^{-2}$. Compared to the experimental γ value, this results in a large mass enhancement of more than 70 times. Taking the observed upturn in C/T into consideration, Fe_2VAl could certainly be regarded as a possible 3d heavy-fermion system, although f-electron heavy-fermion compounds often exhibit a gigantic mass enhancement of two or three orders of magnitude [16]. Since, according to Guo *et al* [8], the estimated λ_{ep} value for Fe_2VAl is

relatively small, consistent with the high θ_D , the electron–phonon coupling contribution to the mass enhancement is of minor importance. In view of this fact combined with the large Fe (V) d-dominant density of states immediately above and below the Fermi level [8], we believe that the observed large enhancement of the electronic specific-heat coefficient is mainly caused by the strong spin fluctuations in the marginally magnetic Fe₂VAl compound. Such a heavy-fermion-like behaviour in d-electron systems has also been reported for (Co_{1-x}Ni_x)₂ScSn with an L2₁ Heusler structure [32]: γ is significantly enhanced only in the vicinity of Ni composition where T_C reaches zero. While the mechanism to explain the electronic mass enhancement for d-electron systems may not be as clear as for f-electron systems, Moriya [33] has proposed a self-consistent renormalization theory of spin fluctuations for nearly and weakly itinerant magnets. According to his theory, heavy-fermion effects should be observed in d-electron systems that are close to the magnetic instability.

Singh and Mazin [9] have suggested that, due to nonstoichiometry or to antisite defects, Fe atoms on the nominally V sites in Fe₂VAl would exhibit local magnetic moments. Given the variation of the lattice parameter showing a sharp upturn and a wide range of V (Fe) composition that can be made [2], there could be significant Fe occupation on the V sites in the experimentally studied samples. In fact, our magnetization data on Fe₂VAl provide some evidence of the local moment magnetism, even though the band calculations [8, 9] have shown that the stoichiometric compound is intrinsically nonmagnetic. Although little is known about such defects, they have proposed that the unusual transport properties of Fe₂VAl may be caused by strong spin fluctuations of Fe impurities on the V sites [9]. We believe that Fe atoms on the nominally V sites may be a common defect, accompanied by either nonstoichiometry or compensating vacancies, in the measured Fe₂VAl samples. Further experiments are now in progress in order to clarify the effect of nonstoichiometry on the electronic properties of the marginally magnetic Fe₂VAl compound.

Acknowledgments

The authors are grateful to Professor K Soda, Nagoya University, for his valuable comments and discussion. Thanks are also due to the Research Centre for Molecular Materials, the Institute for Molecular Science, for assistance in obtaining the low-temperature magnetization data.

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