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Thermal and transport properties of the Heusler-type Fe$_2$VAI$_{1-x}$Ge$_x$ (0 ≤ x ≤ 0.20) alloys: Effect of doping on lattice thermal conductivity, electrical resistivity, and Seebeck coefficient

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We report on the thermoelectric properties of the Heusler-type Fe$_2$VAI$_{1-x}$Ge$_x$ alloys with compositions 0 ≤ x ≤ 0.20. While Fe$_2$VAI (x=0) exhibits a semiconductorlike behavior in electrical resistivity, a slight substitution of Ge for Al causes a significant decrease in the low-temperature resistivity and a large enhancement in the Seebeck coefficient, reaching −130 μV/K for x=0.05 at around room temperature. Comparison with the Fe$_2$VAI$_{1-x}$Si$_x$ system demonstrates that the compositional variation of the Seebeck coefficient falls on a universal curve irrespective of the doping elements (Ge and Si), both of which are isoelectronic elements. The net effect of doping is most likely to cause a rigid-bandlike shift of the Fermi level from the central region in the pseudogap. In spite of a similar decrease in the electrical resistivity with composition of Ge and Si, the thermal conductivity decreases more rapidly for the Ge substitution. It is concluded that doping of heavier atoms such as Ge reduces more effectively the lattice thermal conductivity while retaining the low electrical resistivity as well as the large Seebeck coefficient.

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I. INTRODUCTION

The Heusler-type intermetallic compound Fe$_2$VAI has received intense attention because of the occurrence of a semiconductorlike temperature dependence of electrical resistivity over a wide temperature range up to 1200 K and above. Band structure calculations so far reported consistently predicted the presence of a deep pseudogap at the Fermi level due to the hybridization effects. Nuclear magnetic resonance (NMR) (Ref. 7) and Hall effect measurements strongly support that Fe$_2$VAI is characterized as a low carrier-density semimetal. Optical conductivity and photoelectron spectroscopy measurements clearly manifested the existence of a pseudogap of 0.1–0.2 eV in width. While the pseudogap scenario itself could not account for the semiconductorlike behavior, a steep rise of resistivity at low temperatures has been attributed to spin fluctuations of magnetic antiferromagnetic order in combination with a very low carrier density originating from the pseudogap at the Fermi level.

Because of the possession of a sharp pseudogap across the Fermi level, Fe$_2$VAI-based alloys have attracted a great deal of interest as a potential candidate for new thermoelectric materials. In metallic systems, the Seebeck coefficient $S$ at a temperature $T$ is often discussed using the well-known formula:

$$ S(T) = \frac{\pi^2 k_B^2}{3} (-e) \left( \frac{\partial \ln \sigma(E)}{\partial E} \right)_{E=E_F}, $$

where $\sigma(E)$ is the electrical conductivity and $E_F$ represents the Fermi energy. Provided $\sigma(E)$ is proportional to the density of states (DOS), $N(E)$, it is argued that a large Seebeck coefficient is brought about by a low $N(E)$ coupled with its steep slope, $\partial^2 N(E)/\partial E^2$, near $E_F$. Thus we expect the Seebeck coefficient of Fe$_2$VAI to be well enhanced by doping or off-stoichiometry, since the DOS rises sharply in both sides of the pseudogap. If $E_F$ shifts to a sharply rising portion of the conduction band DOS, $S(T)$ becomes negative because of a positive value of $\partial^2 N(E)/\partial E^2$. Indeed for off-stoichiometric (Fe$_{2+3/5}$V$_{1+2/5}$)$_{100-y}$Al$_y$ (23.8 ≤ y ≤ 26.3) alloys, a small deviation of the Al content from stoichiometry ($y=25.0$) causes a large enhancement in the Seebeck coefficient up to −130 μV/K for the Al-poor sample and 75 μV/K for the Al-rich sample, both of which are accompanied with a significant decrease in the electrical resistivity. Recently, high-resolution x-ray photoemission spectroscopic study of (Fe$_{2+3/5}$V$_{1+2/5}$)$_{100-y}$Al$_y$ alloys has demonstrated that the Seebeck coefficient evaluated from the spectroscopic data coupled with the theoretical band calculations agrees well with the experimental one, including the change in its sign. While off-stoichiometric Fe$_2$-$x$V$_{1+x}$Al alloys also exhibit a large Seebeck coefficient, the sign of $S$ is negative or positive for the V-rich or V-poor samples, respectively, which is inconsistent with the rigid-band model and implies a remarkable modification of the electronic structure.

We have also found that doping of quaternary elements into Fe$_2$VAI causes a large enhancement in the Seebeck coefficient. In particular, a slight substitution of Si for Al results in a significant decrease in the low-temperature resistivity $\rho$ in parallel with an enhancement in the Seebeck coefficient $S$ with a negative sign. Comparison of the calculated band structures between Fe$_2$VAI and Fe$_2$VSi demonstrates that there are overall similarities in their DOSs including the presence of a pseudogap but, for Fe$_2$VSi, $E_F$ lies at a higher energy than the center of the pseudogap, since Si possesses one valence higher than trivalent Al. Therefore, the partial substitution of Si in Fe$_2$VAI$_{1-x}$Si$_x$ would result in a rigid-bandlike shift of $E_F$ from the central region in the pseudogap, though band calculations for Fe$_2$VAI$_{12/5}$Si$_{12/5}$ predict a ferrimagnetic ground state. It should be remarked that...
the low-temperature resistivity decreases more significantly due to doping than that for the off-stoichiometric alloys mentioned above. Remarkably, Fe$_2$VAl$_{1-x}$Si$_x$ alloys possess a large power factor, $P=S^2/\rho$, of $5.4 \times 10^{-3}$ W/m K$^2$ for $x=0.10$ at room temperature,\textsuperscript{20} which is actually higher than that for conventional thermoelectric materials such as Bi$_2$Te$_3$.

To produce good thermoelectric materials with a large figure of merit, $Z=P/\kappa$ ($\kappa$, thermal conductivity), it is further necessary to reduce the value of $\kappa$, although investigation on the doping effect of quaternary elements in Fe$_2$VAl-based alloys is still lacking. Thermal conductivity is generally determined by contributions both from conduction electrons and phonons, and the former can be roughly estimated from the electrical resistivity by using the Wiedemann-Franz law. One of the current challenges is therefore to reduce the lattice thermal conductivity while retaining the low electrical resistivity as well as the large Seebeck coefficient. Taking into account the large power factor for Fe$_2$VAl$_{1-x}$Si$_x$ alloys, we expect the substitution of isoelectronic elements such as heavy Ge to be beneficial for reducing the lattice thermal conductivity due to the atomic mass effect without a substantial modification of the electronic structure. The purpose of the present study is to investigate the temperature dependence of the Seebeck coefficient and electrical resistivity in Fe$_2$VAl$_{1-x}$Ge$_x$ alloys, in addition to the measurement of thermal conductivity at room temperature, and to clarify the doping effect on the thermal and transport properties in comparison with Fe$_2$VAl$_{1-x}$Si$_x$ alloys.

II. EXPERIMENTS

Ingots of Fe$_2$VAl$_{1-x}$Ge$_x$ alloys ($x \leq 0.20$) including the stoichiometric Fe$_2$VAl ($x=0$) were prepared by repeating arc melting of appropriate mixtures of 99.99% pure Fe and Al, and 99.9% pure V and Ge, in an argon atmosphere. We also prepared Fe$_2$VAl$_{1-x}$Si$_x$ alloys ($x \leq 0.20$) in the same manner as described in Ref. 20, where the transport properties were already reported except for the thermal conductivity. Chemical composition was determined within the accuracy of $\pm 0.2\%$ by inductively coupled argon plasma atomic-emission spectroscopy. In particular, special care was taken to ensure that the composition thus determined agrees with the nominal one for all samples studied. The ingots were homogenized at 1273 K for 48 h in vacuum. Samples were cut from the ingots with a SiC blade saw to the size of $1 \times 1 \times 15$ mm$^3$ for resistivity measurements, $0.5 \times 0.5 \times 6$ mm$^3$ for thermoelectric measurements, $0.5 \times 1 \times 7$ mm$^3$ for Hall-effect measurements, and $3.5 \times 3.5 \times 4$ mm$^3$ for thermal-conductivity measurements. Each sample was sealed in an evacuated quartz capsule and was annealed at 1273 K for 1 h and then at 673 K for 4 h followed by furnace cooling.

Powder x-ray diffraction (XRD) spectra were measured with Cu $K\alpha$ radiation for Fe$_2$VAl$_{1-x}$Ge$_x$ and Fe$_2$VAl$_{1-x}$Si$_x$ alloys thus prepared. The overall XRD patterns essentially remain unaltered on replacement of Al by Ge and Si, all of which were identified as a single-phase Heusler ($L_2_1$) structure. Figure 1 shows the lattice parameter of the $L_2_1$ phase as a function of the composition $x$. The lattice parameter for the Ge substitution slightly increases with the composition $x$, whereas it decreases linearly with increasing $x$ for the Si substitution as reported previously.\textsuperscript{24,25} Since the variation in the lattice parameter shown in Fig. 1 is qualitatively consistent with those estimated from Al-Ge and Al-Si solid solutions,\textsuperscript{26} both Ge and Si atoms are believed to occupy preferentially the Al site in the Heusler-type Fe$_2$VAl lattice.

The electrical resistivity was measured by a standard dc four-terminal method with a current of 100 mA over the temperature range 4.2–1273 K and with a rising rate of 0.05 K/s: the measurements at high temperatures were carried out in a vacuum of $4 \times 10^{-4}$ Pa. The Seebeck coefficient was measured with a commercially available apparatus (MMR Technologies inc., SB-100) in the temperature range 100–400 K. The thermal conductivity was measured in a vacuum of $5 \times 10^{-4}$ Pa by the longitudinal steady-state method with Al$_2$O$_3$ as a standard sample. The temperature difference was controlled to be less than 2 K to minimize the heat loss through radiation.

III. RESULTS AND DISCUSSION

A. Seebeck coefficient

The Seebeck coefficient $S$ for a series of Fe$_2$VAl$_{1-x}$Ge$_x$ alloys with $x=0–0.20$ is shown in Fig. 2 as a function of temperature. The value of $S$ for Fe$_2$VAl ($x=0$) is positive and centered around 20–30 $\mu$V/K over the whole temperature range examined. A relatively small value of $S$ most likely reflects the fact that the numbers of carriers of electron and hole pockets are nearly compensated. Also a positive sign of $S$ indicates that a majority carrier must be holes, being consistent with Hall-effect measurements as will be discussed later. When substituted by Ge, the sign of $S$ turns out to be negative and the absolute value increases remarkably with increasing $x$, reaching $|S|=136$ $\mu$V/K at 260 K for $x=0.05$. Moreover, $|S|$ increases gradually as the temperature increases, forming a broad maximum at 250–350 K, and then turns to decrease at higher temperatures. The occurrence of a maximum in $|S|$ has been observed not only for Fe$_2$VAl$_{1-x}$Si$_x$ alloys\textsuperscript{20} but also for (Fe$_{2/3}$V$_{1/3}$)$_{100-x}$Al$_x$ alloys.\textsuperscript{15} The temper-
temperature at which $|S|$ exhibits a maximum increases with increasing $x$. This behavior cannot be explained in terms of Eq. (1), but the decrease in $|S|$ at higher temperatures is believed to be due to an increasing number of thermally excited carriers across the pseudogap,\cite{15} being consistent with the NMR relaxation behavior.\cite{7} The strong doping effect of both Ge and Si in the Heusler Fe$_2$VAl system is reminiscent of the influence of Sb doping in half-Heusler MnNiSn system ($M$=Ti, Zr, and Hf).\cite{27,28} It is well known that the half-Heusler alloys exhibit a maximum of $|S|$ at a temperature higher than that for the present Fe$_2$VAl-based alloys, because of the possession of a greater band gap.\cite{29} We expect the Heusler system to have a good efficiency at around room temperature, while the half-Heusler system can be a potential candidate for intermediate-temperature thermoelectric applications.

In order to make clear the effect of the Ge substitution, the Seebeck coefficient $S$, measured typically at 100 and 300 K, is shown in Fig. 3(a) as a function of $x$ in Fe$_2$VAl$_{1-x}$Ge$_x$ alloys. Also shown in this figure is the Seebeck coefficient in Fe$_2$VAl$_{1-x}$Si$_x$ alloys: the data are taken from Ref. 20, the room-temperature values of which are in agreement with a recent report.\cite{24} The most spectacular feature is that, in spite of a positive value of $S$ for Fe$_2$VAl, a slight substitution of Ge and Si causes a rigid-bandlike shift of the Fermi level from the center of the pseudogap.

We expect a change in sign of the Seebeck coefficient most likely to occur concomitantly with the $x$-dependence of the Hall coefficient. For a valuable counterpoint to Fig. 3(a), the Hall coefficient measured at 100 and 300 K is shown in Fig. 3(b) as a function of the composition $x$ of Ge and Si. The Hall resistivity was found to increase almost linearly with increasing magnetic fields up to 5 T, so that we could determine the Hall coefficient $R_H$ by taking its slope at high fields. The magnitude of $R_H$ for the present samples is found to be of the order of $10^{-8}$ m$^2$/C, which is $10^2$–$10^3$ times larger than that for conventional metals and is nearly equal to that for elemental semimetals such as Sb. The order of magnitude of $R_H$ is determined by the number of carriers: on assuming only one kind of free carriers, we obtain the density of about $5 \times 10^{20}$ cm$^{-3}$ for Fe$_2$VAl. As we reported earlier,\cite{8} the value of $R_H$ for Fe$_2$VAl is positive and increases remarkably as the temperature decreases. This demonstrates that the hole-type carriers dominate in good agreement with the possession of a positive sign of the Seebeck coefficient for $x=0$. Both the Hall and Seebeck coefficient data strongly support that Fe$_2$VAl is a low-carrier density semimetal having slightly excess holes relative to electrons. When substituted by Ge and Si, however, the value of $R_H$ becomes negative, taking its minimum at $x=0.03$ as measured at 100 K. Therefore, the $x$ dependence of the Hall coefficient agrees well with that of the Seebeck coefficient shown in Fig. 3(a).

According to the band calculations of Fe$_2$VAl, the electron and hole pockets are nearly compensated:\cite{2,3} the hole pockets arise from the Fe 3$d$-dominant ($t_{2g}$ character) bands while the electron pocket is of mainly the V 3$d$-e$_g$ character. Since the DOS within the pseudogap is very small, a small change in the electron concentration due to doping would result in an appreciable shift of the Fermi level from the central region in the pseudogap. By considering that the substitution of Ge and Si increases the electron concentration in the system, one may naturally notice that the compositional variation of $S$ can be qualitatively accounted for by the pres-
In Fig. 4, and the low-temperature resistivity decreases significantly with increasing Ge composition $x$. In particular, the resistivity behavior for $x=0.03$ including the occurrence of a broad maximum near 250 K appears to be very similar to that for $\text{Fe}_{2}\text{V}_{0.98}\text{Al}_{0.02}$, which has been identified to be the least affected by magnetic antisite defects and to best represent a nonmagnetic semimetal as predicted by the band calculations of $\text{Fe}_2\text{VAI}$. We expect that doping of excess electrons could appreciably suppress the spin-fluctuation mechanism of magnetic defects. Meanwhile, as seen in Fig. 4, the resistivity for $x=0.10$ is reduced to only 75 $\mu$cm at 4.2 K, which is almost two orders of magnitude lower than that for $\text{Fe}_2\text{VAI}$ ($x=0$), and a positive slope in its temperature dependence appears in the region below 600 K. In spite of a significant decrease in the low-temperature resistivity, all the resistivity curves almost coincide with each other at higher temperatures above 800 K. A substantial reduction in the resistivity at around room temperature certainly acts in favor of the development of thermoelectric materials.

It is interesting to note here that the resistivity behavior for $\text{Fe}_2\text{VAI}_{1-x}\text{Ge}$ is very similar to that for $\text{Fe}_2\text{VAI}_{1-x}\text{Si}$, as reported previously. In order to compare the doping effect, the electrical resistivity $\rho$ measured at 300 K is plotted in the inset of Fig. 4 as a function of the composition $x$ of Ge and Si: the data on $\text{Fe}_2\text{VAI}_{1-x}\text{Si}$ are taken from Ref. 20. It is seen that the Ge and Si substitution results in a sharp decrease in $\rho$ down to 200 $\mu$cm, which is as low as one-third of that for $\text{Fe}_2\text{VAI}$. Moreover, the compositional variation coincides with each other regardless of the doping elements, which is also a positive evidence for a rigid-bandlike shift of the Fermi level due to doping of isoelectronic elements. It is remarked that the value of $\rho$ varies most significantly in the vicinity of the composition of $x=0.05$, where the largest value of $S$ can be obtained as shown in Fig. 3(a). In other word, a smaller $\rho$, which is preferable for thermoelectric materials, is obtained when $E_F$ is located not at the bottom of the pseudogap but at a rather higher or lower energy, when a large value of $S$ with a negative or positive sign, respectively, is also observed. We conclude that one can control both the Seebeck coefficient and the electrical resistivity in the pseudogap systems by tuning the Fermi level to an energy position at which Eq. (1) can be maximized without a substantial modification of the electronic structure.

### B. Electrical resistivity

Figure 4 shows the temperature dependence of the electrical resistivity in $\text{Fe}_2\text{VAI}_{1-x}\text{Ge}$ alloys with $0 \leq x \leq 0.20$. As we reported in Ref. 1 and further discussed in Ref. 12, the stoichiometric $\text{Fe}_2\text{VAI}$ ($x=0$) exhibits a semiconductorlike behavior with the resistivity $\rho$ reaching approximately 2600 $\mu$cm at 4.2 K. The $\ln \rho$ versus $1/T$ plots for the data on $x=0$ become almost linear in the temperature interval 400–800 K so that, on assuming an ordinary $\exp(-\Delta/2k_BT)$-type equation, an energy gap $\Delta$ of approximately 0.1 eV is deduced from its slope. A similar thermal excitation behavior has been observed by NMR experiments, which yield an energy gap of about 0.2 eV. We consider the thermal excitation to originate from the presence of the pseudogap in $\text{Fe}_2\text{VAI}$, as predicted by the band calculations and experimentally confirmed by optical conductivity and photoelectron spectroscopy measurements.

As soon as Al is partially substituted by Ge, the semiconductorlike resistivity behavior disappears rapidly, as shown in Fig. 4, and the low-temperature resistivity decreases significantly with increasing Ge composition $x$. In particular, the resistivity behavior for $x=0.03$ including the occurrence of a broad maximum near 250 K appears to be very similar to that for $\text{Fe}_{1.98}\text{V}_{0.02}\text{Al}$, which has been identified to be the least affected by magnetic antisite defects and to best represent a nonmagnetic semimetal as predicted by the band calculations of $\text{Fe}_2\text{VAI}$. We expect that doping of excess electrons could appreciably suppress the spin-fluctuation mechanism of magnetic defects.
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FIG. 5. Thermal conductivity $\kappa$ at 300 K as a function of composition $x$ in Fe$_2$VAl$_{1-x}$Ge$_x$ and Fe$_2$VAl$_{1-x}$Si$_x$. $\kappa$ is given by the sum of the electronic $\kappa_e$ and lattice contributions $\kappa_{ph}$; the calculated values of $\kappa_e$ are also plotted by the closed symbols.

We have shown that a slight substitution of Ge for Al in Fe$_2$VAl leads to a large enhancement in the Seebeck coefficient $S$, reaching $S = -130 \mu$V/K at 300 K, and a significant reduction in the electrical resistivity $\rho$. While the power factor, $P = S^2/\rho$, for Fe$_2$VAl is only of the order of $10^{-5}$ W/m K$^2$, the value of $P$ rapidly increases with the Ge substitution up to $P = 5.9 \times 10^{-3}$ W/m K$^2$ at 300 K for Fe$_2$VAl$_{0.9}$Ge$_{0.1}$, which is as large as that for Fe$_2$VAl$_{0.9}$Si$_{0.1}$ but is higher than that for conventional thermoelectric materials such as Bi$_2$Te$_3$. We conclude that a rigid-bandlike shift

FIG. 6. Lattice thermal resistivity $1/\kappa_{ph}$ at 300 K as a function of the dimensionless strength of phonon scattering, $\Gamma = c(\Delta M/M)^2$, in Fe$_2$VAl$_{1-x}$Ge$_x$ and Fe$_2$VAl$_{1-x}$Si$_x$. 

$$\tau^{-1} = \Omega \Gamma \omega^4/4\pi \nu^3,$$ (2)

where $\Omega$ is the average atomic volume, $\omega$ is the phonon frequency, and $\nu$ refers to the average phonon velocity. Also $\Gamma$ is a measure of the scattering strength and is given by

$$\Gamma = \sum_i c_i (\Delta M_i M)^2,$$ (3)

where $c_i$ is the fractional concentration of the impurity, whose atomic mass $M_i$ differs from the average mass $M$ by $\Delta M_i = M_i - M$. Obviously this method of reducing $\kappa_{ph}$ is useful in such alloys that the mass of the substitutional atom differs considerably. In Fig. 6, we plot the lattice thermal resistivity, i.e., the inverse of the lattice thermal conductivity, $1/\kappa_{ph}$ for Fe$_2$VAl$_{1-x}$Ge$_x$ and Fe$_2$VAl$_{1-x}$Si$_x$ alloys against the dimensionless strength of phonon scattering, $\Gamma$, evaluated by using Eq. (3) with $c = 0.25x$. As expected from Eq. (2), there is a linear relation between $1/\kappa_{ph}$ and $\Gamma$, and the data on both the alloys are found to be in line with each other. Therefore the lattice thermal resistivity can be mainly interpreted in terms of the mass-difference scattering, so that doping of heavier elements reduces more effectively the phonon mean-free-path in Fe$_2$VAl-based alloys. It is concluded that the substitution of heavy atoms like Ge certainly acts in favor of the development of thermoelectric materials because of a substantial reduction of $\kappa_{ph}$ while retaining the low value of $\rho$ and the large value of $|S|$.

IV. CONCLUSIONS

We have shown that a slight substitution of Ge for Al in Fe$_2$VAl leads to a large enhancement in the Seebeck coefficient $S$, reaching $S = -130 \mu$V/K at 300 K, and a significant reduction in the electrical resistivity $\rho$. While the power factor, $P = S^2/\rho$, for Fe$_2$VAl is only of the order of $10^{-5}$ W/m K$^2$, the value of $P$ rapidly increases with the Ge substitution up to $P = 5.9 \times 10^{-3}$ W/m K$^2$ at 300 K for Fe$_2$VAl$_{0.9}$Ge$_{0.1}$, which is as large as that for Fe$_2$VAl$_{0.9}$Si$_{0.1}$ but is higher than that for conventional thermoelectric materials such as Bi$_2$Te$_3$. We conclude that a rigid-bandlike shift

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of $E_g$ from the center of the pseudogap due to doping can account for a large enhancement in the Seebeck coefficient as well as a significant reduction in the low-temperature resistivity.

Thermoelectric performance is often characterized by the figure of merit, $Z = P/\kappa$ ($\kappa$, thermal conductivity). Although the value of $\kappa$ is relatively large, approximately 28 W/m K for Fe$_2$VAI, it decreases significantly with the Ge substitution, reaching $\kappa = 10-15$ W/m K. Comparison between the Ge and Si substitution demonstrates that heavier Ge doping is more effective in reducing the lattice thermal conductivity, while the electronic contribution is almost the same as each other because Ge and Si are isoelectronic elements. Nevertheless the value of $\kappa$ is still an order of magnitude higher than that for conventional thermoelectric materials, so that we obtain $Z = 4.3 \times 10^{-4}$ K$^{-1}$ at room temperature for Fe$_2$VAI$_{0.9}$Ge$_{0.1}$. Since semimetals with heavy band mass are expected to have a large Seebeck coefficient, the pseudogap system Fe$_2$VAI deserves further studies as an intriguing candidate for low- and intermediate-temperature thermoelectric applications.

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