Large magnetovolume effects and band structure of itinerant-electron metamagnetic $La(Fe_xSi_{1-x})_{13}$ compounds

A. Fujita* and K. Fukamichi
Department of Materials Science, Graduate School of Engineering, Tohoku University, Aoba-yama 02, Sendai 980-8579, Japan

J.-T. Wang† and Y. Kawazoe
Institute for Materials Research, Tohoku University, Katahira 2-1-1, Sendai, 980-8577, Japan

(Received 1 April 2003; published 29 September 2003)

Among Fe-based ferromagnets, $La(Fe_xSi_{1-x})_{13}$ compounds are the first example realizing the itinerant-electron metamagnetic transition from the paramagnetic to the ferromagnetic state, accompanied by a marked magnetovolume effects. The effect of pressure on the Curie temperature is one order larger than that on the spontaneous magnetization in analogy with the results for Fe-Pt invar-type alloys. The large pressure coefficient of T_C, $d \ln T_C/dP$, is attributed to the renormalization effect caused by spin fluctuations for free energy. The metamagnetic transition and the marked magnetovolume effects in the present compounds are related to the characteristic $3d$ band structures in analogy with Fe-Pt alloys in both the ferromagnetic and paramagnetic states.

DOI: 10.1103/PhysRevB.68.104431 PACS number(s): 71.20.Lp, 75.30.Kz

I. INTRODUCTION

It is well known that several kinds of Fe-based alloys, such as Fe-Ni and Fe-Pt, show a significantly large magnetovolume effect.[1,2] In these alloys, the temperature dependence of the spontaneous magnetostriction compensates the lattice expansion due to anharmonic atomic vibration around a certain concentration, resulting in almost zero thermal expansion, i.e., invar effect. The origin of such a large magnetovolume effect in these alloys was investigated during last few decades and is still discussed as one of the important phenomena related to itinerant-electron ferromagnetism.[3,4] Theoretical researches of itinerant-electron ferromagnetism have been a great help in promoting fundamental understandings of the large magnetovolume effect in the invar-type alloys.[5–11] Recent first-principles calculations based on the fixed-spin moment method reveal that the large magnetovolume effect is closely related to the degeneracy of more than two magnetic states,[5–11] which are accompanied by the first-order phase transition. According to the calculations, the transition from the magnetic ordered to disordered state is classified into mainly three types.[11] Namely, type I is a conventional second-order transition, and type II is the first-order transition between the magnetic ordered and the disordered state. Type III involves intermediate ordered states in evolution from the ordered state in the ground state to the disordered state. It has been pointed out that the magnetic phase transition in Fe-Pt invar-type alloy lies close to type II and the first-order transition between the ferromagnetic (F) and the paramagnetic (P) state is expected from theoretical calculations.[5–11] However, to our knowledge, no experimental evidence for such a transition at the Curie temperature T_C has been observed in conventional invar-type alloys mentioned above. A martensitic transformation in these alloys[1,2] may hinder the first-order magnetic phase transition.

The magnetic first-order transition in the itinerant-electron ferromagnets was discussed by Wohlfarth and Rhodes[12] and the magnetic-field-induced first-order transition from the paramagnetic (P) to the ferromagnetic (F) state is predicted from the Stoner model. The field-induced first-order P-F transition in itinerant-electron magnets, which is the so-called itinerant-electron metamagnetic (IEM) transition, has been investigated theoretically[13–18] and experimentally[19–35] by taking spin fluctuations and magnetovolume effects into consideration. The experimental candidates of the IEM transition have been limited to Co-based Laves[19–26] and pyrite[27,28] compounds until recent reports on the IEM transition in UCoAl,[29] and MnSi under hydrostatic pressure.[30] Among Fe-based compounds, the IEM transition defined above has never been observed until we found in $La(Fe_xSi_{1-x})_{13}$ compounds,[31–35] although the following magnetic order-order metamagnetic (MT) transitions have been reported for Fe-based intermetalic compounds such as the ferromagnetic-antiferromagnetic transition in $Hf_{1-x}Ta_xFe_2$,[36] $Sc_{1-x}Ti_xFe_2$,[37] $Ce(Fe_xAl_{1-x})_2$ (Ref. 38) and $La(Fe_xAl_{1-x})_{13}$ (Refs. 39 and 40) compounds and the ferromagnetic-ferromagnetic transition in $Sc_{1-x}Ti_xFe_2$ ($0.5 \leqslant x \leqslant 0.7$) compounds.[41] Very recently, the unique features of the IEM transition observed in Co-based Laves and pyrite compounds, MnSi and $La(Fe_xSi_{1-x})_{13}$ compounds are theoretically weighed against that of the order-order MT's.[18] The present $La(Fe_xSi_{1-x})_{13}$ compounds with $0.84 \leqslant x \leqslant 0.90$ show the thermal-induced first-order F-P phase transition at T_C and the field-induced IEM transition above T_C is caused by applying magnetic field.[31–35] These characteristics are consistent with the theoretical model of the IEM transition based on the Landau expansion including the influence of spin fluctuations.[14–18] According to this theory, the fourth-order Landau coefficient, which is the mode-mode coupling coefficient of spin fluctuations, becomes negative in the itinerant-electron metamagnets, resulting in a large magnetovolume effect.[15,16] It should be noted that the negative fourth-order Landau coefficient in Fe-Pt and Fe-Ni invar-type alloys is also obtained by the fixed spin moment calculations.[6,9,42,43] Therefore it is expected that the electronic structures of the present compounds are very close to the condition for occurrence of the large magnetovolume ef-

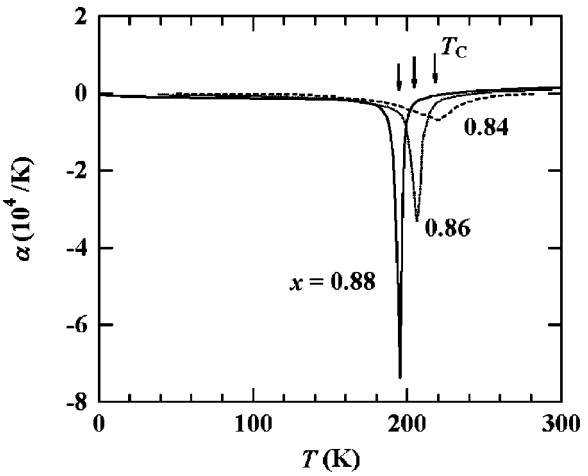

FIG. 1. Temperature dependence of the linear thermal-expansion coefficient α for $La(Fe_xSi_{1-x})_{13}$ ($x=0.84$, 0.86, and 0.88) compounds.

fects related to the first-order magnetic transition discussed on Fe-Pt alloys.[5–11] In the present study, the characteristics of the magnetovolume effects in the $La(Fe_xSi_{1-x})_{13}$ compounds are discussed by using the calculated band structures and compared with those in Fe-Pt and Fe-Ni invar-type alloys.

II. EXPERIMENTS AND BAND CALCULATION

The $La(Fe_xSi_{1-x})_{13}$ compounds were prepared by arc melting in an Ar atmosphere. To homogenize these specimens, the heat treatment was made in a vacuum quartz tube at 1323 K for 10 days. The thermal-expansion measurement was carried out by a three-terminal capacitance method and the linear thermal-expansion coefficient was obtained by a numerical differentiation. The high-field magnetic susceptibility was measured with a superconducting quantum interference device magnetometer. The electronic heat-capacity coefficient was measured by a relaxation method.

The band calculations were carried out by means of the self-consistent full-potential linearized augmented-plane-wave method[44] with the generalized gradient approximation.[45] To simulate the random distribution of Si, a $2\times2\times2$ supercell (including 8 atoms of La, 92 of Fe, and 12 of Si) based on $NaZn_{13}$ unit cell were used with the lattice parameter of 1.1468 nm and 24 of symmetry operations. Therefore the concentration of the calculated compound corresponds to $La(Fe_{0.885}Si_{0.115})_{13}$. The radii of the muffin-tin spheres were set to 0.1482 nm for La, 0.1164 nm for Fe, and 0.1217 for Si, respectively. The Brillouin-zone sampling is performed using four special k points in the irreducible Brillouin zone for such a large supercell. Convergences of the total energy and the charge were carefully checked throughout the present calculations.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the linear thermal-expansion coefficient α obtained from the experimental thermal-expansion curves for the compounds with $x=0.84$, 0.86, and 0.88. The Curie temperature T_C given by the arrow increases with decreasing x.[34] For $x<0.84$, the transition from the ferromagnetic (F) to the paramagnetic (P) state at T_C is of the second order, whereas it becomes the first order for $0.84\leqslant x\leqslant 0.90$ and the itinerant-electron metamagnetic (IEM) transition is observed above T_C.[31–35] According to the results for the pressure effects on magnetic properties of $x=0.86$ with broad transition at T_C, the Curie temperature is decreased by hydrostatic pressure, and both the thermal-induced transition at T_C and the IEM transition above T_C become sharper with increasing pressure,[35] as expected from the theoretical model based on Landau expansion for the magnetic free energy renormalized by spin fluctuations.[17] Therefore the change in order of transition from the first to the second with decreasing x in the present compounds is not extrinsic phenomena such as broadening of the first-order transition due to the concentration distribution, but an intrinsic thermodynamical property of the itinerant-electron metamagnets. The detail relations between the order of transition and the band structure is discussed in connection with Fig. 3. From the temperature T and the magnetic field H dependences of magnetization M around T_C for $x=0.84$, the critical indexes β and δ defined by $M\propto(1-T/T_C)^{\beta}$ and $M\propto H^{1/\delta}$ are evaluated to be 0.27 and 5.3, respectively. These values are very close to $\beta=0.25$ and $\delta=5.0$ for the tricritical point,[46] therefore the tricritical point among these transitions is located around $x=0.84$.

The spontaneous magnetostriction $\omega_m(T)$ is expressed by[15,47]

$$\omega_m(T)=3\int \alpha_m(T)dT=kC_{mv}\{M(T)^2+\xi(T)^2\}, \quad (1)$$

where $\alpha_m(T)$ is the magnetic contribution of the linear thermal-expansion coefficient at T, κ and C_{mv} are the compressibility and the magnetovolume coupling, and $M(T)$ and $\xi(T)$ are the amplitudes of local magnetic moment and spin fluctuations, respectively. For the second-order transition, $M(T)$ continuously decreases and $\xi(T)$ gradually increases with temperature, and therefore the temperature dependence of $\omega_m(T)$ shows no drastic change at T_C. On the other hand, $M(T)$ discontinuously disappears at T_C because of the first-order transition. With increasing Fe concentration x, the discontinuous change of magnetization at T_C takes place. Accordingly, a negative broad peak of the thermal-expansion coefficient α around T_C for $x=0.84$ is attributed to the spontaneous magnetostriction. The negative peak of α for $x=0.86$ becomes shaper, and eventually α for $x=0.88$ shows a divergent behavior at T_C due to the first-order transition. By x-ray diffraction at low temperatures, the $NaZn_{13}$-type cubic symmetry ($Fm\bar{3}c$) is confirmed below and above T_C, therefore the present first-order transition is unrelated to the structural phase transition and a significant change in volume at T_C is caused by the change of $M(T)$ given in Eq. (1). It should be noticed that the coefficient κC_{mv} of $8\times10^{-3}/\mu_B^2$ for $x=0.88$ is comparable to that of $5\sim10\times10^{-3}/\mu_B^2$ for itinerant-electron metamagnetic $Lu(Co_xGa_{1-x})_2$ and $Y(Co_xAl_{1-x})_2$ compounds.[22–26] In the present compounds,

TABLE I. Spontaneous magnetization M_S, the Curie temperature T_C, pressure coefficients of the spontaneous magnetization $d \ln M_S/dP$, and the Curie temperature $d \ln T_C/dP$, and the ratio $d \ln T_C/d \ln M_S$ for the $La(Fe_{0.88}Si_{0.12})_{13}$ (Ref. 34), together with those for ordered and disordered $Fe_{78}Pt_{22}$, and $Fe_{65}Ni_{35}$ alloys (Refs. 48 and 49).

	$La(Fe_xSi_{1-x})_{13}$ [a]	$Fe_{78}Pt_{22}$ [b] (ordered)	$Fe_{78}Pt_{22}$ [c] (disordered)	$Fe_{65}Ni_{35}$ [c]
$M_S(\mu_B)$	2.05	2.15	2.13	1.77
T_C(K)	195	510	380	500
$d \ln M_S/dP$(1/GPa)	−0.019	−0.004	−0.007	−0.050
$d \ln T_C/dP$(1/GPa)	−0.46	−0.09	−0.09	−0.08
$d \ln T_C/d \ln M_S$	24	23	13	1.6

[a]Reference 34.
[b]Reference 48.
[c]Reference 49.

the IEM transition above T_C is also followed by a significant volume change. The critical magnetic field B_C of the IEM transition linearly increases with temperature and the phase transition at T_C corresponds to the IEM transition with $B_C = 0$. Therefore the volume change by the thermal-induced transition is due to the onset of magnetic moment in analogy with the IEM transition.

The IEM transition has been discussed by using the phenomenological theory in terms of the Landau-Ginzburg (LG) expansion including the renormalization effect associated with spin fluctuations for free energy.[15–17] The temperature dependence of the magnetic state is expressed by the following equation for the magnetic free energy:[15–17]

$$F(M) = \frac{1}{2}A(T)M(T)^2 + \frac{1}{4}B(T)M(T)^4 + \frac{1}{6}C(T)M(T)^6 \tag{2}$$

with

$$A(T) = a + \frac{5}{3}b\xi(T)^2 + \frac{35}{9}c\xi(T)^4,$$

$$B(T) = b + \frac{14}{3}c\xi(T)^2,$$

$$C(T) = c.$$

The parameters a, b, and c are correlated to the density of states and its derivative around the Fermi energy. A thermal-induced first-order F-P transition takes place in the range of $a>0$, $b<0$, $c>0$, and $5/28<ac/b^2<3/16$. By considering the magnetovolume coupling energy, the significant magnetovolume effects are expected to appear around the tricritical point of $ac/b^2=5/28$. It should be emphasized that the magnetic phase transition is of the second-order in the range $ac/b^2<5/28$, but the large magnetovolume effects also appear when ac/b^2 is close to 5/28. Actually, the value of α for $x=0.84$, which is very close to the tricritical point, exhibits a negative value due to a large spontaneous volume magnetostriction as seen from Fig. 1. Similar model based on the LG expansion has been applied to calculate the magnetovolume effects in Fe-Pt and Fe-Ni invar-type alloys.[6,9,42,43] According to these results, the magnetic state is also close to the tricritical point.[42] Furthermore, a thermal induced first-order transition from the F state with a large volume to the P state with a small volume is derived from the fixed-spin-moment (FSM) band calculation for an ordered Fe_3Pt invar-type alloy.[42] Unfortunately, the first-order transition is depressed by a martensitic transformation[1,2] and hence there are no experimental reports on the first-order magnetic phase transition at T_C for Fe-Pt alloys. It should be noted that the recent FSM band calculation for disordered $Fe_{75}Pt_{25}$ invar-type alloy also indicates a similar energy barrier between the F and the P states.[43] On the other hand, the calculated phase transition for Fe-Ni invar-type alloys is the second order due to an unclear energy barrier in the magnetic free energy.[6,42] Accordingly, it is expected that the feature of the magnetovolume effect in the $La(Fe_xSi_{1-x})_{13}$ compounds is similar to that of Fe-Pt rather than that of Fe-Ni invar-type alloys.

One of the most important features of the magnetovolume effects related to the IEM transition is that the pressure dependence of the Curie temperature T_C is significant, compared with that of the spontaneous magnetic moment M_S.[14,17] To compare the pressure effects of the present compounds and those of invar-type alloys, the values of M_S, T_C, pressure coefficients of M_S and T_C and the ratio of pressure coefficient of T_C to that of M_S for the $La(Fe_{0.88}Si_{0.12})_{13}$ compound,[34] ordered and disordered $Fe_{78}Pt_{22}$, and $Fe_{65}Ni_{35}$ invar-type alloys[48,49] are listed in Table I. As shown in Table I, for both the ordered and disordered $Fe_{78}Pt_{22}$ invar-type alloys, $d \ln T_C/dP$ is one order larger in magnitude than $d \ln M_S/dP$, while both the pressure coefficients are the same order for $Fe_{65}Ni_{35}$ invar-type alloy. Accordingly, the ratio of the pressure coefficient of T_C to that of M_S, $d \ln T_C/d \ln M_S$, for ordered and disordered $Fe_{72}Pt_{28}$ invar-type alloys is one order larger than that for $Fe_{65}Ni_{35}$ alloy. It should be noted that the ratio of the $La(Fe_{0.88}Si_{0.12})_{13}$ compound is almost the same as that for ordered $Fe_{72}Pt_{28}$ alloy as seen from the last line in the table, although the former exhibits the first-order and the latter the second-order phase transition. The effect of pressure on M_S is mainly attributed to the bandwidth narrowing caused by a smaller volume un-

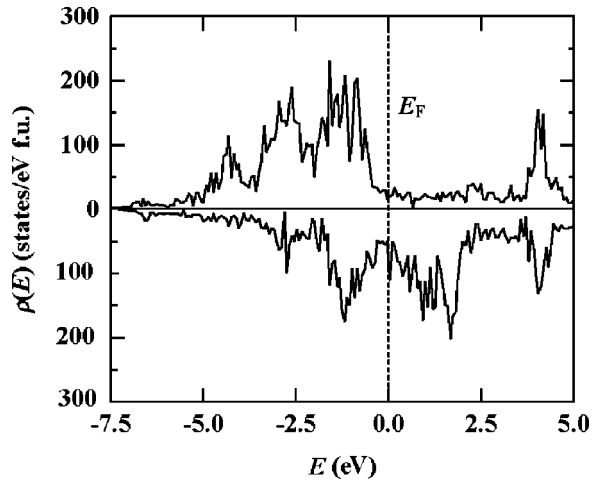

FIG. 2. Density of states of the $La(Fe_{0.885}Si_{0.115})_{13}$ compound in the ferromagnetic state.

der applied pressure.[2] On the other hand, the Curie temperature strongly depends on the temperature dependence of the magnetic free energy due to the renormalization effect of spin fluctuations given by Eq. (2).[6,15,42] Therefore the effect of pressure on also involves the influence of spin fluctuations.

By adding the elastic energy, $\omega(T)^2/2\ \kappa$, for the volume change, $\omega(T)$, and the magnetovolume energy,$-\omega(T)C_{mv}\{M(T)^2+\xi(T)^2\}$, to Eq. (2), the following relations are derived:[15]

$$-\omega(T)=\kappa P-\kappa C_{mv}\{M(T)^2+\xi(T)^2\} \tag{3}$$

and

$$\mu_0 H=A(T)M(T)+B(T)M(T)^3+C(T)M(T)^5 -2\omega(T)C_{mv}M(T). \tag{4}$$

For the first-order F-P transition, the Curie temperature T_C is obtained from Eqs. (3) and (4) under the condition that the energy level in the F state is the same as that in the P state. For the second-order transition, on the other hand, T_C is determined as the temperature where $dH/dM=0$.[15,16] Equations (3) and (4) describe that the renormalization effect related to spin fluctuations for free energy indicates the pressure dependence through the magnetovolume effect. Namely, the spontaneous volume magnetostriction discussed in Eq. (1) reduces the variation of the free energy by the renormalization effect of spin fluctuations. On the other hand, the decrease of volume by hydrostatic pressure enhances the renormalization effect, resulting in the decrease in the Curie temperature. Therefore the significant difference between the magnitude of $d\ln Ms/dP$ and $d\ln T_C/dP$ implies a strong contribution from the renormalization effect of spin fluctuations, and the influence of spin fluctuations on the magnetovolume effects in the present compound is similar to that in the order and disorder Fe-Pt invar-type alloys, though the direct comparison of the renormalization effect is difficult because of the dependence of $\xi(T)$ on the microscopic parameters such as a cutoff wave vector of spin fluctuations.

TABLE II. Electronic specific-heat coefficient γ and high-field magnetic susceptibility χ_{hf} of the $La(Fe_{0.88}Si_{0.12})_{13}$, together with those of ordered and disordered $Fe_{78}Pt_{22}$ and $Fe_{65}Ni_{35}$ alloys (Refs. 50–52).

	$La(Fe_xSi_{1-x})_{13}$	$Fe_{78}Pt_{22}$ (ordered)	$Fe_{78}Pt_{22}$ (disordered)	$Fe_{65}Ni_{35}$
γ (mJ/mol K^2)	7.5	9.5 [a]	7.5 [a]	11.3 [b]
χ_{hf} ($10^{-3}\mu_B/T$)	1.2	1.0 [c]	0.8 [c]	2.0 [c]

[a]Reference 50.
[b]Reference 51.
[c]Reference 52.

Next, the $3d$ band calculation has been applied to examine the relation between the magnetovolume effect and the metamagnetic transition.[5–10] The projected density of state (DOS) curves in the F state is presented in Fig. 2. The energy scale is shifted so as to the Fermi level $E_F=0$. The La$5d$ and Fe$3d$ bands are well hybridized, but the main part of the partial DOS of the La$5d$ is still recognized around $+4$ eV. The main part of the $3d$ DOS in the majority spin band is located below E_F. Furthermore, E_F lies just at the dip between maxima around $+2.0$ eV and around -1.8 eV in the minority spin band. It should be noticed that a similar dip structure of DOS around E_F located between large maxima for Fe-Pt system is connected with the small $d\ln M_S/dP$.[7]

In order to confirm the low density of states at the Fermi level, $\rho(E_F)$, the coefficient of the electronic specific-heat coefficient γ and the high-field magnetic susceptibility χ_{hf} have been investigated. The values of γ and χ_{hf} for the $La(Fe_{0.88}Si_{0.12})_{13}$ together with those for ordered and disordered $Fe_{78}Pt_{22}$, and $Ni_{65}Fe_{35}$ invar-type alloys[50–52] are presented in Table II. As seen in Fig. 2, E_F of the $La(Fe_{0.885}Si_{0.115})_{13}$ is located well above the $3d$ majority spin band. According to the first-principles band calculations, E_F of ordered Fe_3Ni and disordered $Fe_{75}Ni_{25}$ alloys crosses the upper part of majority spin $3d$ band,[3,9] while those of ordered Fe_3Pt and disordered $Fe_{75}Pt_{25}$ alloys are situated the upper edge.[7,53] In Table II, the value of γ proportional to $\rho(E_F)$ in the free-electron model exhibits the difference in magnitude, consistent with the calculated results of $\rho(E_F)$ regardless of the difference in composition and atomic order between the calculations and the experiments for Fe-Pt and Fe-Ni invar-type alloys. The value of χ_{hf} is also related to $\rho(E_F)$ as[54]

$$\chi_{hf}=\chi_{hf}^0\left\{1+(\chi_{hf}^0)^2\left[(g\mu_B)^{-3}\left(\frac{\rho_+(E_F)'}{\rho_+(E_F)^3}-\frac{\rho_-(E_F)'}{\rho_-(E_F)^3}\right)-\frac{\partial I}{\partial M}\bigg|_{M=M_0}\right]H\right\}, \tag{5}$$

with

$$\chi_{hf}^0=(g\mu_B)^2\left(\frac{1}{\rho_+(E_F)}+\frac{1}{\rho_-(E_F)}-I\right)^{-1},$$

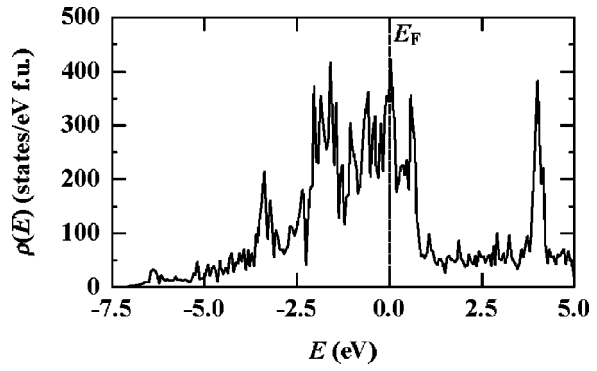

FIG. 3. Density of states of the $La(Fe_{0.885}Si_{0.115})_{13}$ compound in the paramagnetic phase.

where I is the effective exchange integral. The subscripts $+$ and $-$ of ρ indicate the majority and the minority spin bands, respectively. Although this relation is not so straightforward as γ, the value of $\rho_+(E_F)$ is a leading term for strong ferromagnets in which the majority spin band is situated below E_F. Accordingly, χ_{hf} of the strong ferromagnetic $La(Fe_{0.88}Si_{0.12})_{13}$ compound and the ordered and disordered $Fe_{78}Pt_{22}$ invar-type alloys is smaller than that of the weak ferromagnetic $Fe_{65}Ni_{35}$ invar-type alloy.

From the results of the fixed-spin-moment band calculations, it has been pointed out that the feature of paramagnetic DOS is also important to discuss the large volume effect in Fe-Pt and Fe-Ni invar-type alloys.[7,9] Figure 3 shows the DOS curve for the paramagnetic $La(Fe_{0.885}Si_{0.115})_{13}$ compound. The general feature of the $3d$ DOS is relatively similar to that of fcc Fe, ordered Fe_3Pt, and Fe_3Ni,[7,9,55] though the bandwidth is different from one another. Especially, a high DOS in the upper part of the $3d$ band close to the Fermi level is considered as one of the triggers of the magnetic instability in Fe-Pt and Fe-Ni invar-type alloys.[7,9] Furthermore, it has been pointed out that the peak of DOS just above E_F, which goes down below E_F by the exchange splitting in the F state, is the origin of the energy barrier between the P and the F states.[9] It should be noticed that the peak of DOS just above E_F is consistent with the negative sign of the mode-mode coupling coefficient among spin fluctuations discussed in connection with $d \ln T_C/dP$. Namely, the mode-mode coupling coefficient b in Eq. (2) is related to the $3d$ band structure as follows:[56]

$$b = \frac{1}{16\rho(E_F)^3}\left\{\left(\frac{\rho(E_F)'}{\rho(E_F)}\right)^2 - \frac{\rho(E_F)''}{3\rho(E_F)}\right\}. \quad (6)$$

The peak of the DOS just above E_F tends to make the sign of b negative.[56] Therefore it is concluded these features for the large magnetovolume effects and the first-order magnetic phase transition are confirmed in the band structures of the $La(Fe_{0.885}Si_{0.115})_{13}$, though a quantitative uncertainty of the peak height of the calculated DOS is involved due to a large number of atoms in the formula unit cell.

IV. CONCLUSION

In order to discuss the correlation between the itinerant-electron metamagnetic (IEM) transition and the large magnetovolume effects in the $La(Fe_xSi_{1-x})_{13}$ compounds, calculated $3d$ band structures of the $La(Fe_{0.885}Si_{0.115})_{13}$ compound in the ferromagnetic and the paramagnetic states are compared with those of Fe-Pt and Fe-Ni invar-type alloys. A significant magnetovolume effect is observed in $La(Fe_xSi_{1-x})_{13}$ not only in the range $x \geqslant 0.84$ at the first-order ferromagnetic-paramagnetic transition at T_C, but also in the range $x < 0.84$, where the transition at T_C is of the second order. A large pressure dependence of the Curie temperature T_C compared to that of the spontaneous magnetization M_S is observed in $x = 0.88$, which is similar to the magnetovolume effects in the $Fe_{78}Pt_{22}$ invar-type alloy. The band calculations reveal that the $La(Fe_{0.88}Si_{0.12})_{13}$ compound is in a strong ferromagnetic state, consistent with the experimental results of the electronic specific-heat coefficient γ and the high-field magnetic susceptibility χ_{hf}. Furthermore, the feature of majority spin band in the ferromagnetic state is similar to that of ordered and disordered Fe-Pt invar-type alloys, and the feature of the $3d$ band in the paramagnetic state is close to the condition for the inducement of magnetic instability, accompanied by the first-order magnetic transition.

ACKNOWLEDGMENT

The present work was supported by a Grant-in Aid for Scientific Research (B2), No. 13555168, from the Japan Society for the Promotion of Science.

*Electronic address: afujita@material.tohoku.ac.jp
†Present address: Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China.

[1] E.F. Wassermann, *Ferromagnetic Materials*, edited by K.H.J. Buschow and E.P. Wohlfarth (North-Holland, Amsterdam, 1990), Vol. 5, p. 237.
[2] M. Shiga, in *Electronic and Magnetic Properties of Metals and Ceramics II*, Materials Science and Technology Vol. 3B, edited by R.W. Cahn, P. Haasen, and E.J. Kramer (Wiley-VCH, Weinheim, 1993), p. 159.
[3] M. Schlifgaarde, I.A. Abrikosov, and B. Johansson, Nature (London) **400**, 46 (1999).
[4] L. Dubrovinsky, N. Dubrovinskaia, I.A. Abrikosov, M. Venström, F. Westman, S. Carlson, M. van Schilfgaarde, and B. Johansson, Phys. Rev. Lett. **86**, 4851 (2001).
[5] V.L. Morruzi, Phys. Rev. B **41**, 6939 (1990).
[6] P. Mohn, K. Schwarz, and D. Wagner, Phys. Rev. B **43**, 3318 (1991).
[7] M. Podgórny, Phys. Rev. B **43**, 11 300 (1991).
[8] M. Podgórny, Phys. Rev. B **46**, 6293 (1992).
[9] P. Entel, E. Hoffmann, P. Mohn, K. Schwarz, and V.L. Moruzzi, Phys. Rev. B **47**, 8706 (1993).
[10] B. Sanyal and S.K. Bose, Phys. Rev. B **62**, 12 730 (2000).
[11] V.L. Moruzzi, Phys. Rev. Lett. **57**, 2211 (1986).
[12] E.P. Wohlfarth and P. Rhodes, Philos. Mag. **7**, 1817 (1962).
[13] M. Shimizu, J. Phys. (Paris) **43**, 155 (1982).

[14] H. Yamada, J. Phys.: Condens. Matter **3**, 4115 (1991).
[15] H. Yamada, J. Magn. Magn. Mater. **139**, 162 (1995).
[16] H. Yamada, Phys. Rev. B **47**, 11 211 (1993).
[17] H. Yamada, K. Fukamichi, and T. Goto, Phys. Rev. B **65**, 024413 (2002).
[18] H. Yamada, K. Fukamichi, and T. Goto, Physica B **327**, 148 (2003).
[19] T. Sakakibara, T. Goto, Y. Yoshimura, K. Murata, and K. Fukamichi, J. Magn. Magn. Mater. **90&91**, 131 (1991).
[20] I.L. Gabelko, R.Z. Levitin, A.S. Markosyan, V.I. Silant'ev, and V.V. Snegirev, J. Magn. Magn. Mater. **94**, 287 (1991).
[21] H. Saito, T. Yokoyama, and K. Fukamichi, J. Phys.: Condens. Matter **9**, 9333 (1997).
[22] H. Saito, T. Yokoyama, Y. Terada, K. Fukamichi, H. Mitamura, and T. Goto, Solid State Commun. **113**, 447 (2000).
[23] T. Yokoyama, H. Saito, K. Fukamichi, K. Kamishima, T. Goto, and H. Yamada, J. Phys.: Condens. Matter **13**, 928 (2001).
[24] K. Fukamichi, T. Yokoyama, H. Saito, T. Goto, and H. Yamada, Phys. Rev. B **64**, 134401 (2001).
[25] H. Saito, T. Yokoyama, K. Fukamichi, K. Kamishima, and T. Goto, Phys. Rev. B **59**, 8725 (1999).
[26] T. Goto and M.I. Bartashevich, J. Phys.: Condens. Matter **10**, 3625 (1998).
[27] K. Adachi, M. Matsui, and M. Kawai, J. Phys. Soc. Jpn. **46**, 1474 (1979).
[28] T. Goto, Y. Shindo, S. Ogawa, and T. Harada, Physica B **237**, 482 (1997).
[29] N.V. Mushnikov, T. Goto, K. Kamishima, H. Yamada, A.V. Andreev, Y. Shiokawa, A. Iwao, and V. Sechovsky, Phys. Rev. B **59**, 6677 (1999).
[30] C. Thessieu, J. Flouquet, G. Lapertot, A.N. Stepanov, and D. Jaccard, Solid State Commun. **95**, 707 (1995).
[31] A. Fujita, K. Akamatsu, and K. Fukamichi, J. Appl. Phys. **85**, 4756 (1999).
[32] A. Fujita and K. Fukamichi, IEEE Trans. Magn. **35**, 3796 (1999).
[33] K. Fukamichi and A. Fujita, J. Mater. Sci. Technol. **16**, 167 (2000).
[34] A. Fujita, S. Fujieda, K. Fukamichi, H. Mitamura, and T. Goto, Phys. Rev. B **65**, 014410 (2002).
[35] A. Fujita, K. Fukamichi, M. Yamada, and T. Goto, J. Appl. Phys. **93**, 7268 (2003).
[36] Y. Nishihara and Y. Yamaguchi, J. Phys. Soc. Jpn. **51**, 1333 (1982).
[37] Y. Nishihara and Y. Yamaguchi, J. Phys. Soc. Jpn. **53**, 2201 (1984).
[38] D.F. Franceschini and S.F. da Cunha, J. Magn. Magn. Mater. **51**, 280 (1985).
[39] T.T.M. Palstra, G.J. Nieuwenhuys, J.A. Mydosh, and K.H.J. Buschow, Phys. Rev. B **31**, 4622 (1985).
[40] K. Irisawa, A. Fujita, and K. Fukamichi, J. Alloys Compd. **305**, 17 (2000).
[41] Y. Nishihara and Y. Yamaguchi, J. Phys. Soc. Jpn. **54**, 1689 (1985).
[42] P. Entel and M. Schröter, Physica B **161**, 160 (1989).
[43] R. Hayn and V. Drchal, Phys. Rev. B **58**, 4341 (1998).
[44] P. Blaha, K. Schwarz, and J. Luitz, *Wien97* (Tech. Univ. Wien, Austria, 1999).
[45] J.P. Pardew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
[46] A. Aharony, *Critical Phenomena*, Lecture Notes in Physics Vol. 186, edited by F.J.W. Hahne (Springer, Berlin, 1983), p. 209.
[47] T. Moriya and K. Usami, Solid State Commun. **34**, 95 (1980).
[48] M.M. Abd-Elmeguid and H. Micklitz, Phys. Rev. B **40**, 7395 (1989).
[49] K. Hayashi and N. Mori, Solid State Commun. **38**, 1057 (1981).
[50] K. Sumiyama, M. Shiga, and Y. Nakamura, J. Phys. Soc. Jpn. **40**, 996 (1976).
[51] R. Caudron, J. Meunier, and P. Costa, Solid State Commun. **14**, 975 (1974).
[52] O. Yamada, H. Maruyama, and R. Pauthenet, *High Field Magnetism*: *Proceedings of the International Symposium on High Field Magnetism*, edited by M. Date (North-Holland, Amsterdam, 1983), p. 97.
[53] J. Inoue and M. Shimizu, J. Phys. F: Met. Phys. **13**, 2677 (1983).
[54] M. Shimizu, Rep. Prog. Phys. **44**, 21 (1981).
[55] P.M. Marcus and V.L. Moruzzi, Phys. Rev. B **38**, 6949 (1988).
[56] M. Shimizu and J. Inoue, J. Phys. F: Met. Phys. **17**, 1221 (1987).