Concentration Dependence of Pressure Effect in $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ Compounds

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Effects of pressure P on the magnetic moment M and the Curie temperature T_C have been investigated for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ above and below the magnetic phase boundary concentration $x = 0.86$, where the ferromagnetic-paramagnetic transition at T_C changes from the first-order ($x \geq 0.86$) to the second-order ($x < 0.86$). The pressure coefficient of M exhibits a sluggish variation against concentration and no anomaly was observed at $x = 0.86$, being consistent with the Landau expansion model. On the other hand, T_C for the second-order transition has a large negative pressure coefficient dT_C/dP and its magnitude increases with increasing x . Above $x = 0.86$, the magnitude of dT_C/dP for the first-order transition increases with x , contrary to the theoretical expectation. It has been revealed that the spin-wave dispersion coefficient becomes smaller when the first-order transition becomes clear by changing the concentration and also applying pressure. Consequently, it is plausible that dT_C/dP above $x = 0.86$ is enhanced by the increase of instability of the ferromagnetic state.

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1. Introduction

Itinerant-electron metamagnetic (IEM) transition, *i.e.*, the magnetic-field-induced first-order phase transition from the paramagnetic (P) to the ferromagnetic (F) state has been extensively investigated from both theoretical and experimental viewpoints.¹⁻⁵⁾

We have demonstrated that $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ compounds with $0.86 \leq x \leq 0.90$ exhibit a first-order magnetic phase transition at the Curie temperature T_C , and also the IEM transition above T_C .⁶⁻⁸⁾ Furthermore, large magnetovolume effects such as a large pressure coefficient of T_C are followed by the IEM transition.⁸⁻¹⁰⁾ The large magnetovolume effects of the IEM transition have theoretically been explained from both the steep change of the magnetization characterized by the first-order transition and the negative sign of the forth-order coefficient in the Landau expansion of magnetic free energy.^{4,11)} The negative forth-order Landau coefficient is also attributed to the large magnetovolume effects in so-called Invar-type alloys such as Fe-Pt alloys.¹¹⁾ Therefore, it is expected that a large pressure coefficient of T_C is also expected in the $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ with x smaller than the boundary concentration of $x = 0.86$ when the sign of the forth-order coefficient is negative.

In the present study, the pressure effects on magnetization M and T_C for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ have been investigated in both the concentration ranges above and below $x = 0.86$. The obtained results have been discussed in terms of spin fluctuations and stability of the F state.

2. Experimental

Alloying of $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ was carried out by arc-melting under an Ar gas atmosphere. The heat treatment for homogenizing of the specimens was made in an evacuated quartz tube at 1323 K for 10 d. The magnetization measurements under hydrostatic pressure were carried out by using an

extraction-type magnetometer equipped with a Cu-Ti pressure clamp cell.¹²⁾

3. Results and Discussion

Figures 1(a) and (b) show the pressure P dependence of the spontaneous magnetization M_s at 4.2 K and the Curie temperature T_C , respectively, for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ ($x = 0.83, 0.84$ and 0.85) compounds. Both M_s and T_C exhibit a negative linear dependence on P . The slope of the T_C - P line apparently increases with increasing x , while that of the M_s - P shows a weak concentration dependence. In the present compounds, a thermally induced ferromagnetic (F)-paramagnetic (P) transition is of the first-order in $0.86 \leq x \leq 0.90$ and an itinerant-electron metamagnetic (IEM) transition

Fig. 1 Pressure dependence of (a) the magnetization and (b) the Curie temperature for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ ($x = 0.83, 0.84$ and 0.85).

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appears just above T_C .⁸⁾ On the other hand, the transition at T_C becomes of the first-order in the concentration range $x < 0.86$ and no IEM transition is observed.

According to the spin fluctuation theory based on the Landau–Ginzburg expansion including influences of spin fluctuations and magnetovolume effect,^{4,5)} the change in the F–P transition from the second- to the first-order is explained by the balance among the Landau coefficients of the second, forth and sixth term of the magnetic free energy given as

$$F(M) = \frac{1}{2} \{ \tilde{a}(T) + 2\kappa C_{mv} P \} M^2 + \frac{1}{4} \tilde{b}(T) M^4 + \frac{1}{6} \tilde{c}(T) M^6. \quad (1)$$

where κ and C_{mv} are the compressibility and the magnetovolume coupling constant, respectively. The coefficients $\tilde{a}(T)$, $\tilde{b}(T)$ and $\tilde{c}(T)$ are defined as⁵⁾

$$\begin{aligned} \tilde{a}(T) &= a_0 + \left\{ \frac{5}{3} \tilde{b}(0) + \frac{4}{3} \kappa C_{mv}^2 \right\} \xi(T)^2 \\ &\quad + \frac{35}{9} \tilde{c}(0) \xi(T)^4, \\ \tilde{b}(T) &= (b_0 - 2\kappa C_{mv}^2) + \frac{14}{3} \tilde{c}(0) \xi(T)^2, \quad \tilde{c}(T) = c_0, \end{aligned} \quad (2)$$

where $\xi(T)^2$ is the mean-square amplitude of spin fluctuations. The parameters a_0 , b_0 and c_0 are related to the 3d band structures. The thermally induced first-order transition from the F to the P phase is observed under the conditions $\tilde{a}(T) > 0$, $\tilde{b}(T) < 0$, $\tilde{c}(T) > 0$ and $5/28 - \eta < \tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2 < 3/16$, where η is the parameter defined as

$$\eta = \frac{2}{7} \kappa C_{mv}^2 / |\tilde{b}(0)|. \quad (3)$$

In this region, T_C decreases with increasing the ratio of $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2$. By taking the phase diagram of the $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ into account, the relation $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2 \sim 5/28 - \eta$ is expected around the critical concentration of $x = 0.86$ for the appearance of the first-order transition at T_C . In the concentration region $x < 0.86$, where the relation $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2 < 5/28 - \eta$ is established, the pressure dependence of T_C is obtained from the pressure dependence of $\xi^2(T_C)$ calculated from eq. (1) as⁵⁾

$$\begin{aligned} \frac{d\xi(T_C)^2}{dP} &= \\ &= -\frac{3}{\sqrt{35}} \frac{\kappa C_{mv}}{\tilde{b}(0)} \left[\left(\frac{5}{28} - \eta \right) - \frac{\tilde{a}(0)\tilde{c}(0)}{\tilde{b}(0)^2} + \frac{7}{5} \eta^2 \right]^{-1/2}. \end{aligned} \quad (4)$$

Note that $\xi^2(T)$ proportional to T^2 and T at low and high temperatures respectively.⁴⁾ From eq. (4), it is demonstrated that the magnitude of pressure coefficient T_C for the second-order transition remarkably increases when $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2$ comes close to $5/28 - \eta$. Therefore, the increase in the slope of the T_C – P line with coming close to $x = 0.86$ is explained from eq. (4). On the other hand, the pressure coefficient of T_C for the first-order F–P transition is given by the following relation.⁵⁾

$$\begin{aligned} \frac{d\xi(T_C)^2}{dP} &= \\ &= -\frac{6}{\sqrt{7}} \frac{\kappa C_{mv}}{\tilde{b}(0)} \left[\frac{\tilde{a}(0)\tilde{c}(0)}{\tilde{b}(0)^2} - \left(\frac{5}{28} - \eta \right) + \frac{35}{38} \eta^2 \right]^{-1/2}. \end{aligned} \quad (5)$$

From eq. (5), the pressure dependence of $\xi(T_C)$ decreases with creasing $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2$. Accordingly, it is expected that the magnitude of dT_C/dP for the $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ decreases with increasing x above $x = 0.86$, and in conjunction with eq. (4), the concentration dependence of dT_C/dP is considered to have a negative maximum at around $x = 0.86$. Furthermore, the pressure dependence of M_s is expressed as

$$\begin{aligned} M_s^2 &= \\ &= -\frac{\tilde{b}(0)}{2\tilde{c}(0)} \left[1 + \sqrt{1 + 4\{\tilde{a}(0) + 2\kappa C_{mv} P\}\tilde{c}(0)/\tilde{b}(0)^2} \right]. \end{aligned} \quad (6)$$

From eq. (6), no anomaly is expected near the critical value $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2 = 5/28 - \eta$.

To verify whether anomaly in the pressure coefficients is induced by the change in the order of transition at T_C or not, the concentration dependences of dM_s/dP and dT_C/dP are displayed in Figs. 2(a) and (b), respectively. These coefficients in the concentration less than $x = 0.86$ are determined from the data in Fig. 1 in the ranges of $P < 0.5$ GPa, and those in the concentration $x \geq 0.86$ from the previous data⁸⁾ of the pressure dependence of M_s and T_C in the corresponding pressure ranges. As shown in Fig. 2(a), the concentration dependence of dM_s/dP is relatively small and no discontinuity is observed around $x = 0.86$, being consistent to the expected behavior from eq. (6).

The magnitude of dT_C/dP increases with increasing x above $x = 0.86$ in contrast to the variation expected from eqs. (4) and (5) in which the magnitude exhibits a maximum in the vicinity of the boundary concentration $x = 0.86$. What

Fig. 2 Concentration dependence of the pressure coefficient of (a) the magnetization dM_s/dP and (b) the Curie temperature dT_C/dP for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$.

Fig. 3 Concentration dependence of the spin-wave dispersion coefficient D_{sw} for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$.

has to be noticed is that theoretical variation on dT_C/dP mentioned above is obtained by changing the ratio $\tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2$ only. On the other hand, in the $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$, not only the Landau coefficients but also the thermal variation of spin fluctuations and/or thermal stability of the F state may be changed by changing Fe concentration. It is difficult to determine the thermal variation of spin fluctuations from static measurements and these parameters may be obtained from dynamical experiments such as neutron scattering up to high energy regions. However, the spin-wave dispersion coefficient D_{sw} obtained from the following $T^{3/2}$ relation¹³⁾ is regarded as one of the measures for the magnetic instability due to spin fluctuations.^{13,14)}

$$M = M_s - 5.26\mu_B \left[\frac{4\pi D}{k_B} \right]^{-3/2} T^{3/2} \quad (7)$$

For example, in Fe_3Pt invar-type alloy having a large magnitude of dT_C/dP , D_{sw} is evaluated to be $85 \text{ meV } \text{\AA}^2$ from magnetization measurement,¹⁴⁾ much smaller compared to the values of other itinerant-electron magnets such as $D_{sw} = 330 \text{ meV } \text{\AA}^2$ for Ni .¹⁵⁾ Shown in Fig. 3 is the concentration dependence of D_{sw} for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$. It should be noted that the values of D_{sw} in the whole concentration range also show small values. For more details, D_{sw} shows a slight increase with increasing x up to the vicinity of the boundary concentration $x = 0.86$, but a remarkable decrease takes place at this boundary concentration with a further increase of x . Strictly speaking, the value of D_{sw} obtained from the magnetizations does not coincide with that obtained from neutron scattering,^{13,14)} but such small D_{sw} means the fast decrease of magnetization against temperature due to the excitation of spin fluctuations, indicating instability of the F state. In the case of the second-order F-P transition, $F(M)$ in eq. (1) has a single minimum at $M = M_s$ in the F state.^{4,5)} On the other hand, $F(M)$ has double minimum at $M = 0$ in the P and $M = M_s$ in the F states.^{4,5)} The concentration dependence of D_{sw} implies that the appearance of the double minimum structure of $F(M)$ reduces the stability of the F state, resulting in the large magnitude of dT_C/dP mentioned above.

In order to confirm the relation between the double minimum structure and the instability of the F state, change in the temperature dependence of M for $x = 0.86$ has been

Fig. 4 Temperature T dependence of magnetization M plotted in the $M-T^{3/2}$ form under various external pressure P for $\text{La}(\text{Fe}_{0.86}\text{Si}_{0.12})_{13}$. The inset shows the pressure dependence of the spin-wave dispersion coefficient D_{sw} .

investigated by applying pressure. It has been reported that the transition at T_C for $x = 0.86$ becomes sharper with increasing applied pressure,^{8,10)} indicating that the double minimum structure becomes clear. Figure 4 shows the temperature dependence of magnetization plotted against $T^{3/2}$ for $x = 0.86$ under various external pressure. The linear variation of $M-T^{3/2}$ at low temperatures indicates the validity of eq. (7). With increasing P , the slope of the $M-T^{3/2}$ line becomes larger. From these results, the pressure dependence of D_{sw} is plotted in the inset of Fig. 4. It is clear that D_{sw} decreases with increasing P , revealing that the F state becomes unstable. The influence of the pressure does not correspond to the variation of Fe concentration for magnetic properties. However, it is obvious that the stability of the F state should be also taken into consideration in addition to the change in the Landau coefficients in eq. (1). In other words, the magnitude of dT_C/dP is enhanced due to the instability of the F state and therefore, the increase of dT_C/dP with x holds above the boundary concentration $x = 0.86$. Further detailed information on the relation between the double minimum structure and the magnetic excitation would be obtained from dynamical experiments such as neutron scattering and nuclear magnetic resonance.

4. Conclusion

Pressure effects on magnetization M and the Curie temperature T_C have been investigated for $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$. The pressure coefficient of M , dM/dP , exhibits a small negative value and slightly depends on the concentration. No anomalous behavior is observed around the boundary concentration $x = 0.86$ between the second-order and the first-order transitions, as expected from the spin fluctuation theory based on the Landau expansion. On the other hand, T_C is strongly decreased by pressure and the magnitude of dT_C/dP becomes larger with increasing x below $x = 0.86$. Furthermore, the magnitude of dT_C/dP keeps to increase even above

$x = 0.86$, in contrast to the theoretical model which shows a maximum of the magnitude of dT_C/dP at the boundary concentration $x = 0.86$. From the concentration dependence of the spin-wave dispersion coefficient D_{sw} in both $x < 0.86$ and $x \geq 0.86$, as well as the pressure dependence of D_{sw} for $x = 0.86$, it is concluded that the F state becomes more unstable with the appearance of the first-order transition at T_C , accompanied by the enhancement of dT_C/dP even above $x = 0.86$.

REFERENCES

- 1) T. Sakakibara, T. Goto, Y. Yoshimura, K. Murata and K. Fukamichi: J. Magn. Magn. Mater. **90&91** (1991) 131–134.
- 2) H. Saito, T. Yokoyama and K. Fukamichi: J. Phys.: Condens. Matter **9** (1997) 9333–9346.
- 3) T. Goto, K. Fukamichi and H. Yamada: Physica B **300** (2001) 167–185.
- 4) H. Yamada: Phys. Rev. B **47** (1993) 11211–11219.
- 5) H. Yamada, K. Fukamichi and T. Goto: Phys. Rev. B **65** (2002) 024413.
- 6) A. Fujita, Y. Akamatsu and K. Fukamichi: J. Appl. Phys. **85** (1999) 4756–4758.
- 7) A. Fujita and K. Fukamichi: IEEE Trans. Magn. **35** (1999) 3796–3798.
- 8) A. Fujita, S. Fujieda, K. Fukamichi, H. Mitamura and T. Goto: Phys. Rev. B **65** (2002) 014410.
- 9) A. Fujita, K. Fukamichi, J.-T. Wang and Y. Kawazoe: Phys. Rev. B **68** (2003) 104431.
- 10) A. Fujita, K. Fukamichi, M. Yamada and T. Goto: J. Appl. Phys. **93** (2003) 7268–7270.
- 11) H. Yamada and K. Terao: J. Phys.: Condens. Matter **6** (1994) 10805–10809.
- 12) T. Goto, Y. Shindo, H. Takahashi and S. Ogawa: Phys. Rev. B **56** (1997) 14019–14028.
- 13) H. A. Mook: *Spin Waves and Magnetic Excitations*; ed. A. S. Borovik-Romanov and S. K. Sinha, Modern Problems in Condensed Matter Science (North-Holland, Amsterdam, 1988) Vol. 22.1, pp. 425.
- 14) Y. Ishikawa, S. Onodera and K. Tajima: J. Magn. Magn. Mater. **10** (1979) 183–190.
- 15) P. W. Mitchell and D. Mck. Paul: Phys. Rev. B **32** (1985) 3272–3278.