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Pressure-induced anomalies in itinerant-electron metamagnetic properties around the critical end point in La($Fe_{0.89}Si_{0.11}$)$_{13}$

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Pressure-induced anomalies around the critical end point in an itinerant-electron metamagnetic La($Fe_{0.89}Si_{0.11}$)$_{13}$ compound have been investigated. The Curie temperature T_C is decreased from 189 to 0 K by applying a critical pressure $p_C=1.3$ GPa. The relation $T_C \propto (p_C-p)^{0.5}$ is explained by the spin-fluctuation theory based on the Landau expansion. A susceptibility maximum appears at $T_{\max} \sim 70$ K above $p=1.2$ GPa. On the other hand, the critical point $(T_0, \mu_0 H_0)$ of the transition is lowered to $T_0=14$ K and $\mu_0 H_0=2.5$ T at 1.4 GPa, showing no static ferromagnetic moment in the field-induced state below T_0.

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I. INTRODUCTION

A magnetic field-induced first-order transition from the paramagnetic (P) to the ferromagnetic (F) state in itinerant-electron systems has been referred to as the itinerant-electron metamagnetic (IEM) transition. During last two decades, various compounds, e.g., Co-based Laves phase and pyrite compounds,[1–3] and a UCoAl 5f-electron compound,[4] together with MnSi under the hydrostatic pressure,[5] have been found to exhibit the IEM transition.

In Fe-based compounds, La(Fe_xSi_{1-x})$_{13}$ compounds ($0.86 \leqslant x \leqslant 0.90$) are the sole examples exhibiting the IEM transition mentioned above,[6–11] apart from the order-order metamagnetic transition such as the antiferromagnetic-ferromagnetic transition in some Fe-based Laves phase compounds.[12,13] After the first observation of the IEM transition in La($Fe_{0.88}Si_{0.12}$)$_{13}$ just above the Curie temperature T_C in 1999,[7] much attention has been paid to the present compound system with high Fe concentrations, because of their large magnetovolume[7–11,14] and magnetocaloric effects[15,16] induced by the IEM transition.

In the ferromagnetic compounds having the IEM transition above T_C, the thermally induced first-order F-P transition is also observed at T_C, and a marked pressure dependence of T_C is common to them.[2–5] The pressure coefficient of T_C, $d \ln T_C/dp$, for La($Fe_{0.88}Si_{0.12}$)$_{13}$, is large, −0.46 GPa^{-1}.[9] Such significant pressure dependence of T_C has been explained by both spin-fluctuation effects based on the Landau expansion[17,18] and first-principles band calculations.[11] Furthermore, in recent studies on the magnetic properties of MnSi, the external pressure has been found not only to trigger the IEM transition, but also to settle the critical point $(T_0, \mu_0 H)$, as a termination condition of the first-order transition, down to 0 K, accompanied by an appearance of a novel magnetic state related to spin fluctuations.[19,20]

Although both MnSi and La(Fe_xSi_{1-x})$_{13}$ compounds are common in the appearance of the IEM transition, the former is weakly ferromagnetic with $T_C=29.5$ K and $M_s=0.4\mu_B$,[19] while the latter exhibits much stronger ferromagnetic properties with a high T_C of about 200 K and a large M_s of about $2\mu_B$. An electronic structure[11] and magnetic properties of the latter are very similar to those of Fe_3Pt invar alloy. The La($Fe_{0.88}Si_{0.12}$)$_{13}$ compound is in a strong ferromagnetic state, resulting in relatively small values of the electronic specific heat coefficient γ and the high-field magnetic susceptibility χ_{hf}, in analogy with the data for both the ordered and disordered $Fe_{78}Pt_{22}$ invar-type alloys.[21,22] Furthermore, for the La($Fe_{0.88}Si_{0.12}$)$_{13}$ compound,[11] and both the ordered and disordered $Fe_{78}Pt_{22}$ invar-type alloys,[23,24] $d \ln T_C/dP$ is one order larger in magnitude than $d \ln M_S/dP$. As a result, $d \ln T_C/d \ln M_S$ for the La($Fe_{0.88}Si_{0.12}$)$_{13}$ compound is almost the same as those for ordered and disordered $Fe_{72}Pt_{28}$ invar-type alloys. Accordingly, for the La(Fe_xSi_{1-x})$_{13}$ compounds, drastic changes of magnetic properties under high pressures are expected.

In the present study, we discuss the influence of pressure on the IEM transition in La($Fe_{0.89}Si_{0.11}$)$_{13}$. At high pressures, the F state is completely suppressed and the critical point of the IEM transition becomes close to 0 K, i.e., near the critical end point. From our previous results, the pressure effect becomes larger with increasing Fe concentration x,[9] while the preparation of a high-quality specimen becomes more difficult in higher x regions due to the immiscibility of Fe and La. Therefore, we tried to prepare the specimen with $x=0.89$ in the present study. The obtained results for the pressure effect have been discussed in terms of the spin fluctuations and the magnetovolume coupling.

II. EXPERIMENTS

The La($Fe_{0.89}Si_{0.11}$)$_{13}$ specimen was prepared by arc melting in an Ar gas atmosphere. The ingot was remelted several times, and the heat treatment for homogenization was carried out in an evacuated quartz tube at 1323 K for 10 days. The magnetization was measured with a superconducting quantum interference device (SQUID) magnetometer up to 5.5 T. Hydrostatic pressure was applied up to 1.4 GPa by using a nonmagnetic Cu-Ti clamp cell.[3,25] The value of susceptibility of the clamp is extremely small, that is, 5×10^{-8} emu/g at

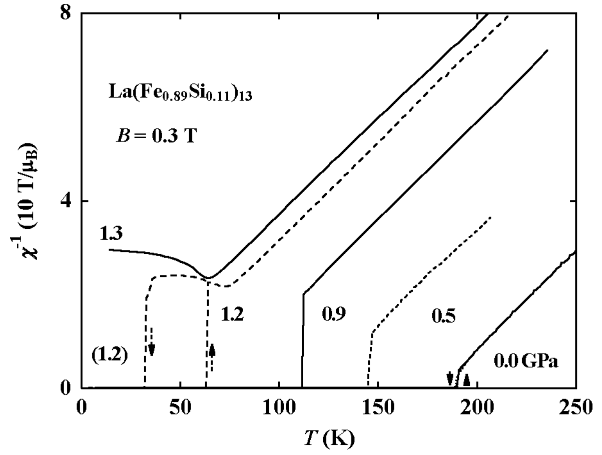

FIG. 1. Temperature dependence of the inverse magnetic susceptibility χ^{-1} for La(Fe$_{0.89}$Si$_{0.12}$)$_{13}$ in a magnetic field of 0.3 T under hydrostatic pressures p=0.0, 0.5, 0.9, 1.2, and 1.3 GPa. The thermal process is marked by the arrows.

1.8 K. Hydrostatic pressures were calibrated by measuring the shift of the superconducting transition temperature of Pb.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of inverse magnetic susceptibility χ^{-1} for the La(Fe$_{0.89}$Si$_{0.12}$)$_{13}$ obtained from thermomagnetization measurements in a magnetic field of 0.3 T by applying hydrostatic pressures p=0.0, 0.5, 0.9, 1.2, and 1.3 GPa. The $\chi^{-1}-T$ curve exhibits a discontinuous change $\Delta\chi$ at the Curie temperature T_C due to the first-order magnetic phase transition from the ferromagnetic (F) to the paramagnetic (P) state. With increasing pressure, T_C significantly decreases and $\Delta\chi$ at T_C becomes more significant. The sharp change of χ^{-1} at the transition temperature indicates that the specimen has no marked compositional inhomogeneity. The thermal hysteresis between heating and cooling processes indicated by the arrows is less than 3 K at ambient pressure, whereas it becomes 30 K at p=1.2 GPa. By applying p=1.3 GPa, no spontaneous magnetization is observed, resulting in the disappearance of the F state.

One of the characteristic behaviors related to the IEM transition is the appearance of a broad maximum in the temperature dependence of χ.[1–5,26] A Curie-Weiss behavior of χ^{-1} for the La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ appears just above T_C, and no maximum of χ (or minimum of χ^{-1}) is observed at ambient pressure. On the other hand, as shown in Fig. 2, the susceptibility maximum emerges around $T_{\max}$=72 K, just above T_C at 63 K, at p=1.2 GPa. By increasing pressure higher than 1.2 GPa, T_C is suppressed to 0 K, while $T_{\max}$ remains at 64 and 56 K for p=1.3 and 1.4 GPa, respectively. The Curie-Weiss behavior of χ^{-1} is still observed above $T_{\max}$ and the slope is almost independent of applying pressure. From the slope, the effective magnetic moment m_{eff} is estimated to be 3.20 μ_B/Fe/atom, being 1.6 times larger than M_s=2.07μ_B/Fe/atom at ambient pressure.

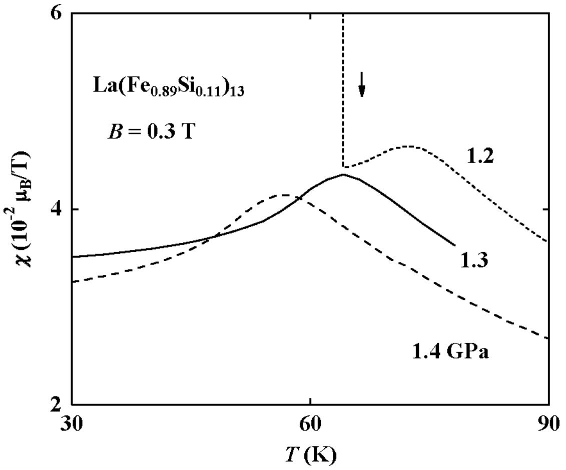

FIG. 2. Temperature dependence of susceptibility for La(Fe$_{0.89}$Si$_{0.12}$)$_{13}$ in a magnetic field of 0.3 T under p=1.2, 1.3, and 1.4 GPa.

From these results, together with the additional thermomagnetization data, the p-T phase diagram of the La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ is established in Fig. 3. The large pressure dependence of T_C for IEM compounds has been discussed by using the following Landau expansion of magnetic free energy $F(M)$:[18]

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

The Landau coefficients $\tilde{a}(T)$, $\tilde{b}(T)$, and $\tilde{c}(T)$ are defined as

$$\tilde{a}(T)=a_0+\left(\frac{5}{3}\tilde{b}(0)+\frac{4}{3}\kappa C_{mv}^2\right)\xi(T)^2+\frac{35}{9}\tilde{c}(0)\xi(T)^4,$$

$$\tilde{b}(T)=(b_0-2\kappa C_{mv}^2)+\frac{14}{3}\tilde{c}(0)\xi(T)^2, \quad \tilde{c}(T)=c_0, \tag{2}$$

where $\xi(T)^2$, κ, and C_{mv} are the mean-square amplitude of the spin fluctuations, the compressibility and the magneto-

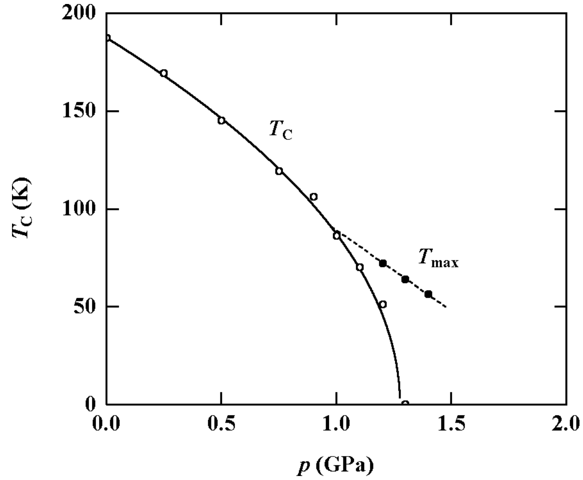

FIG. 3. Phase diagram in the temperature-pressure plane for La(Fe$_{0.89}$Si$_{0.12}$)$_{13}$. The temperatures T_C and $T_{\max}$ denote the first-order Curie temperature and the susceptibility maximum temperature, respectively.

volume coupling constant, respectively. The parameters a_0, b_0, and c_0 are related to the 3d band structures. By presuming $\xi(T)^2 = \delta T^2$,[26] the pressure dependence of the first-order T_C is given by

$$T_C = K_1\sqrt{K_2 - 4\sqrt{7}\sqrt{K_3 p - K_4}}, \tag{3}$$

with

$$K_1 = \sqrt{\frac{9}{7}\frac{|\tilde{b}(0)|}{\delta\tilde{c}(0)}}, \quad K_2 = 1 + 56\eta, \quad K_3 = \frac{2\kappa C_{mv}\tilde{c}(0)}{\tilde{b}(0)^2}, \tag{4}$$

and

$$K_4 = \left(\frac{5}{28} - \frac{\tilde{a}(0)\tilde{c}(0)}{\tilde{b}(0)^2}\right) - (\eta + 28\eta^2),$$

where $\eta = (2/7)\kappa C_{mv}^2/|\tilde{b}(0)|$.[18] The thermally induced first-order transition from the F to the P state is observed under the conditions of $\tilde{a}(0) > 0$, $\tilde{b}(0) < 0$, $\tilde{c}(0) > 0$ with $5/28 < \tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)2 < 3/16$. From the log-log plots of the p-T_C line, the exponent α in the following expression is determined to be 0.5:

$$T_C = \beta(p_C - p)^{\alpha}, \tag{5}$$

where p_C is the critical pressure where $T_C = 0$. Recently, it has been reported that the transition at T_C of MnSi changes from second to first order by applying hydrostatic pressure. It should be noted that the value of α also becomes 0.5 for the first-order T_C.[19,20] Furthermore, the square root p dependence of T_C has been discussed in terms of the Landau-Ginzburg-Wilson (LGW) model,[23] which adds nonanalytic terms to Eq. (1). However, the square root dependence in the present La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ persists up to 100 K, where the nonanalytic term is negligibly small;[27] therefore, the LGW model is inappropriate to apply to the present compound. To elucidate whether the square root dependence of the La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ is categorized as a different type from that of MnSi or not, we analyze these phenomena in terms of the spin fluctuation theory based on an analytic Landau expansion.[18] It is easy to denote that the asymptotic form of Eq. (3) around $T_C \sim 0$ becomes as follows:

$$T_C \sim 4\sqrt{\frac{7}{2}}K_1\sqrt{\frac{K_3}{K_2}}\sqrt{\frac{1}{K_3}\left(\frac{3}{16} - \frac{\tilde{a}(0)\tilde{c}(0)}{\tilde{b}(0)^2}\right) - p}. \tag{6}$$

Namely, the square root p dependence of T_C is also explained from the analytic Landau expansion model mentioned above. The coefficient β in Eq. (5) for the present compound is 160 K/GPa$^{1/2}$, being 7.3 times larger than that of 22 K/GPa$^{1/2}$ for MnSi.[28] From Eq. (6), β is related to K_1, K_2 and K_3, which are difficult to determine individually. The value of $\eta = (2/7)\kappa C_{mv}^2/|\tilde{b}(0)|$ is estimated to be 0.01 for a relatively small $|\tilde{b}(0)|$ of Co-based Laves phase compounds.[18] The magnitude of κC_{mv} is almost the same as transition metal-based compounds,[29,30] and consequently, it is expected that η rarely exceeds 0.01 and K_2 is close to unity. A set of parameters $K_1 K_3^{1/2}$ is proportional to $[\kappa C_{mv}/\{\tilde{b}(0)\delta\}]^{1/2}$. As mentioned in connection with Eq. (3), κC_{mv} of the La(Fe$_{0.88}$Si$_{0.12}$)$_{13}$ is $0.45\%/\mu_B^2$ in the F state[10] and that of MnSi is about $0.5\%/\mu_B^2$ in the F state.[31] Therefore, $|\tilde{b}(0)|\delta$ is expected to govern the magnitude of β.

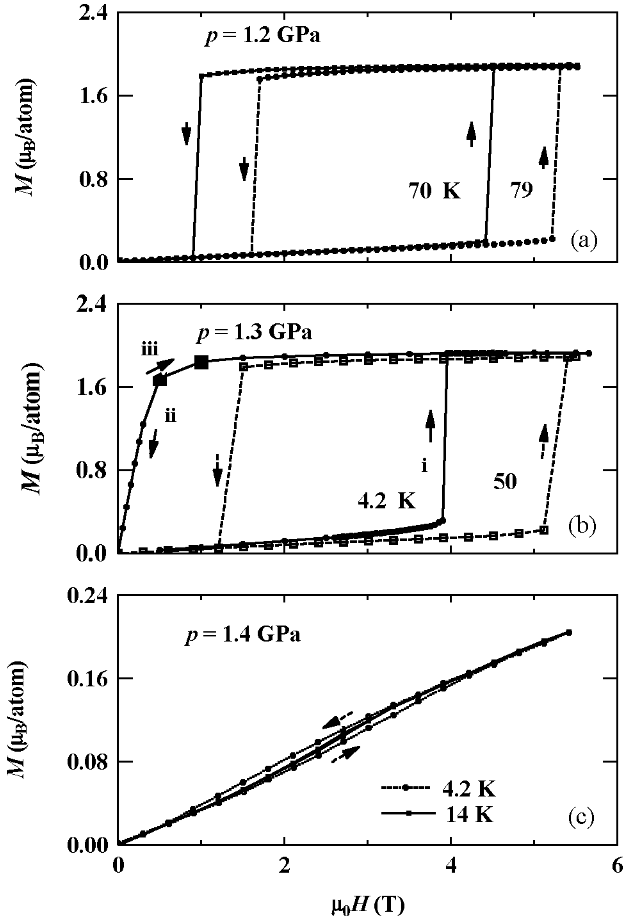

FIG. 4. Magnetization curves of La(Fe$_{0.89}$Si$_{0.12}$)$_{13}$ at representative temperatures at applied pressures of (a) 1.2 GPa, (b) 1.3 GPa, and (c) 1.4 GPa. The arrows indicate the direction of the applied magnetic field.

From Eq. (1), the following relation for the temperature dependence of the IEM transition field $\mu_0 H_C$ has been derived around $\tilde{a}(0)\tilde{b}(0)/\tilde{c}(0)^2 \sim 3/16$, where $T_C \sim 0$ K and $\mu_0 H_C \sim 0$ T,[18,26]

$$\mu_0 H_C = \mu_0 H_C^{T=0} + M_{ind}(|\tilde{b}(0)|\delta)T^2, \tag{7}$$

where M_{ind} is the induced magnetic moment at the IEM transition. By applying Eq. (7) to the data obtained from the isothermal magnetization curves under $p_c = 1.3$ GPa, the value of $(|\tilde{b}(0)|\delta)^{-1/2}$ is estimated to be about $44(\mu_B/T)^{1/2}$ K for the present compound. Note that the detail of thermal variation of the magnetization curve will be discussed in connection with Figs. 4(a)–4(c). The temperature dependence of $\mu_0 H_C$ in MnSi estimated from the literary data[28,32] results in $[|\tilde{b}(0)|\delta]^{-1/2} = 7 \sim 8(\mu_B/T)^{1/2}$ K. Therefore, the difference in $[|\tilde{b}(0)|\delta]^{-1/2}$ effectively explains the difference between β of La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ and MnSi. Accordingly, it is concluded that the square root p dependence of T_C for the $La(Fe_{0.89}Si_{0.11})_{13}$ as well as MnSi is common to the IEM compounds explained by the spin fluctuation theory based on the analytic Landau expansion.

The appearance of T_{max} in the P state for the IEM compounds is also derived from Eq. (1) as

$$T_{max} = \frac{3}{14\delta}\frac{|\tilde{b}(0)|}{\tilde{c}(0)}\left(1 - \frac{14}{5}\eta\right). \tag{8}$$

Equation (3) is not an explicit function of p, therefore, a weak pressure dependence of T_{max} is expected[18] and the weak pressure dependence of T_{max} in Fig. 1 is explained from the pressure dependence of b_0 and/or c_0, as discussed for $ZrZn_2$.[33] What has to be mentioned is that T_{max} is theoretically expected to be higher than the first-order T_C in an almost whole range of $5/28 < \tilde{a}(T)\tilde{b}(T)/\tilde{c}(T)^2 < 3/16$, and the relation $T_{max} < T_C$ appears in only a narrow range around $\tilde{a}(T)\tilde{b}(T) \sim 5/28$.[18] On the other hand, the relation $T_{max} > T_C$ is established when $p > 1.2$ GPa in the present compound, and the pressure range where $T_{max} > T_C$ is relatively wide. The theoretical discussion for the relation $T_{max} > T_C$ is based on the assumption that both the F and P states have the same value of κC_{mv}. However, it has been reported that $\kappa C_{mv} = 1.10\% / \mu_B^2$ in the P state is about two times larger than $0.50\% / \mu_B^2$ in the F state for $La(Fe_{0.88}Si_{0.12})_{13}$.[10] The magnitude of T_C is related to the local minimum of $F(M)$ in the F state described by Eq. (1), while T_{max} is related to that in the P state. As indicated by Eq. (8), the larger κC_{mv} makes $\tilde{b}(0)$ smaller and η larger, resulting in lower T_{max}. Therefore, the difference between the magnitude of κC_{mv} in the F and P states is one of the reasons why χ_{max} is hidden below T_C at ambient pressure, and T_{max} appears only when T_C is suppressed lower than T_C by applying pressure, as observed in the present study for the first time.

To evaluate the influence of pressure on the IEM transition, the magnetization measurements were carried out at various temperatures under $p = 1.2$, 1.3, and 1.4 GPa, and the results are displayed in Figs. 4(a)–4(c), respectively. The arrows indicate the direction of the applied magnetic field. As shown in Fig. 4(a), the IEM transition with a wide hysteresis is observed in the P state at 70 K under $p = 1.2$ GPa. With increasing temperature, both the onset and offset critical fields, $\mu_0 H_{on}$ and $\mu_0 H_{off}$, respectively, shift to a higher region, implying that the thermodynamic transition field $\mu_0 H_C = \mu_0 (H_{on} + H_{off})/2$ increases with temperature. Furthermore, the field-induced transition still occurs when temperature is elevated above T_{max}. Such variation of the magnetization curves against temperature under $p = p_c$ accords with the typical features of the IEM transition.[26] In Fig. 4(b), at $p = 1.3$ GPa, $\mu_0 H_{on}$ appears at around 4 T at 4.2 K in the magnetic-field increasing process (i), and M decreases toward 0 in the decreasing process (ii). By applying the magnetic field again in the process (iii), the values of M locate on the curve of the process (ii), instead of that of the process (i), as indicated by the solid square. Therefore, the decrease of M toward 0 in the process (ii) is not due to the IEM transition, but to the formation of the ferromagnetic domain. These results reveal that $\mu_0 H_C$ is close to 0 T at $T = 4.2$ K, and the reverse F-P transition is blocked by the energy barrier between the local minima of $F(M)$ in the F and P states. As depicted in Fig. 4(b), $\mu_0 H_C$ becomes larger and both the P-F and F-P transitions are observed at $T = 50$ K. A drastic change of the M-H curve is observed by applying $p = 1.4$ GPa, as depicted in Fig. 4(c). No static ferromagnetic moment is confirmed in the field-induced (FI) state up to $\mu_0 H = 5.5$ T at 4.2 K. Note that a scale of the ordinate for Fig. 4(c) is ten times as small as that for Figs. 4(a) and 4(c). However, a small hysteresis appears around 2.5 T at 4.2 K and disappears at the critical point $(T_0, \mu_0 H_0) = (14$ K, 2.5 T). This hysteresis is not attributed to the pressure gradient in the clamp cell, because there is no indication of increase in the transition field $\mu_0 H_C$, in contrast to the apparent increase of $\mu_0 H_C$ against temperature at pressures $p \leq 1.3$ GPa. To our knowledge, such a steep change of magnetization in the FI state by applying pressure has never been observed in the other IEM compounds. From Eq. (1), the M-p relation has a singular point that is prevented by the IEM transition.[18] However, this singularity would remain when influences of both the zero-point spin fluctuations and the difference between κC_{mv} in the F and P states are strong, which are neglected in Eq. (1).

IV. CONCLUSION

We have investigated the influence of high pressure on the itinerant-electron metamagnetic (IEM) transition around the critical end point in the $La(Fe_{0.89}Si_{0.11})_{13}$ compound. At ambient pressure, the thermally induced first-order ferromagnetic (F)—the paramagnetic (P) transition occurs at $T_C = 189$ K. By applying pressure, T_C is significantly decreased, especially above 1.0 GPa. The relation $T_C = \beta(p_c - p)^{0.5}$ is confirmed at high pressures. This square root dependence is explained by the spin fluctuation theory based on the analytic Landau expansion including the magnetovolume effect. Furthermore, it is clarified that β is clearly related to the temperature dependence of the IEM transition field, as expected from the theory mentioned above. The susceptibility maximum χ_{max} appears at $T_{max} = 72$ K, slightly higher than $T_C = 68$ K at $p = 1.2$ GPa. No spontaneous moment is observed in the ground state at 1.3 GPa, and the IEM transition field converges around 0 T at 4.2 K. Accordingly, the critical pressure p_c for the disappearance of the F state is close to 1.3 GPa. By increasing p up to 1.4 GPa, hysteresis of the field-induced transition is observed up to 14 K, however, no static ferromagnetic moment is confirmed in the field-induced state.

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