# Strong Pressure Effect on the Curie Temperature of Itinerant-Electron Metamagnetic $La(Fe_{0.88}Si_{0.12})_{13}H_y$ and $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_y$

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The initial pressure shift of the Curie temperature  $T_c$  for the first-order transition,  $dT_c/dP$ , for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> becomes small after hydrogen absorption. In addition, the hydrogen absorption reduces the ratio of the volume change at  $T_c$  to the magnetization squared at  $T_c$ ,  $\Delta\omega(T_c)/M(T_c)_F^2$ , suggesting the decrease of the magnetoelastic coupling constant  $\kappa C_{mv}$ . On the other hand,  $dT_c/dP$  is enhanced by the partial substitution of Ce for La in connection with the change of the band structure. Consequently,  $dT_c/dP$  of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> is larger than that of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> in a wide change of  $T_c$ . [doi:10.2320/matertrans.MBW200822]

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# 1. Introduction

La(Fe<sub>x</sub>Si<sub>1-x</sub>)<sub>13</sub> compounds with  $0.86 \le x \le 0.90$  exhibit the thermal-induced first-order transition between the paramagnetic (P) and the ferromagnetic (F) states at the Curie temperature  $T_c$ .<sup>1)</sup> By applying magnetic field, the itinerantelectron metamagnetic (IEM) transition, that is, the firstorder transition to the F state takes place above  $T_c$ .<sup>1)</sup> A cubic NaZn<sub>13</sub>-type structure of the  $Fm\bar{3}c$  space group is kept after undergoing the thermal-induced first-order transition<sup>2,3)</sup> and the IEM transition.<sup>4)</sup> In addition, the unit cell volume in the F state is larger than that of the P state because in a large spontaneous volume magnetostriction associated with the local magnetic moment.<sup>2–4)</sup> Therefore,  $T_c$  is significantly decreased by applying hydrostatic pressure.<sup>3)</sup> The critical temperature  $T_0$  of the disappearance of the IEM transition also decreases with increasing pressure.<sup>5)</sup> The pressure effect on  $T_0$  is smaller than that on  $T_c$ , and hence the temperature difference,  $T_c - T_0$ , increases with increasing pressure.<sup>5)</sup> A close interplay between magnetism and volume, namely, the large magnetovolume effects in La(Fe<sub>x</sub>Si<sub>1-x</sub>)<sub>13</sub> have been explained in terms of band structure and spin fluctuation based on the Landau-Ginzburg model for the IEM transition.<sup>3,5–7)</sup>

The unit cell volume of  $La(Fe_xSi_{1-x})_{13}$  is isotropically expanded, and  $T_c$  is increased up to about 330 K by hydrogen absorption.<sup>8,9)</sup> The both temperatures of  $T_c$  and  $T_0$  increase, while the value of  $T_0 - T_c$  decreases.<sup>10)</sup> In contrast, the unit cell volume is decreased by the partial substitution of Ce for La, because the ionic radius of Ce is smaller than that of La due to the lanthanide contraction.<sup>11,12)</sup> In this case, the temperatures of  $T_c$  and  $T_0$  are decreased and the value of  $T_0 - T_c$  is increased.<sup>13)</sup> Therefore, the volume dependence of  $T_c$  and  $T_0$  for La(Fe<sub>x</sub>Si\_{1-x})\_{13}H<sub>y</sub> and La<sub>1-z</sub>Ce<sub>z</sub>(Fe<sub>x</sub>Si\_{1-x})\_{13} is qualitatively consistent with that of La(Fe<sub>x</sub>Si\_{1-x})\_{13} under hydrostatic pressure. However, it has been pointed out that the IEM transition of La(Fe<sub>x</sub>Si\_{1-x})\_{13} under hydrostatic pressure is different from that of La<sub>1-z</sub>Ce<sub>z</sub>(Fe<sub>x</sub>Si\_{1-x})\_{13} having a similar value of  $T_c$ .<sup>14,15</sup>

In the present paper, to discuss the difference between the influence of the hydrogen absorption and the partial substitution of Ce, the pressure effect on  $T_c$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> and La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> having a similar value of  $T_c$  is investigated.

## 2. Experiments

The La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> and La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> compounds were prepared by arc-melting in an argon gas atmosphere and the heat-treatments were carried out in an evacuated quartz tube. The annealing temperatures of  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}$  and  $La(Fe_{0.88}Si_{0.12})_{13}$  were 1423 and 1323 K, respectively. The annealing duration was 10 days. The hydrogen absorption was carried out by annealing under a hydrogen gas atmosphere. In order to control the hydrogen content, the annealing temperature and the hydrogen gas pressure were changed. The powder x-ray diffractions were carried out by using Cu K $\alpha$  radiation. The magnetization was measured with a SQUID magnetometer. The hydrostatic pressure was applied by using a nonmagnetic Cu-Ti clamp cell. The applied pressures were calibrated by measuring the shift of the superconducting transition temperature of Pb.

## 3. Results and Discussion

Figure 1 depicts the thermomagnetization curve in the heating process under a magnetic field of 1T for  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_{1.7}$ . The Curie temperature  $T_c =$ 175 K of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> is increased up to about 320 K by hydrogen absorption. Since the magnetization decreases rapidly in the vicinity of  $T_c$  with increasing temperature, the thermal-induced first-order transition at  $T_{\rm c}$  remains after hydrogen absorption. The inset represents the powder x-ray diffraction patterns of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.7</sub> and La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> at room temperature. The several vertical short bars below the diffraction patterns are the positions of all the possible Bragg reflections from a cubic NaZn<sub>13</sub>-type structure with the Fm3c space group. The diffraction patterns of all the specimens are identified as the cubic NaZn<sub>13</sub>-type structure. The diffraction peaks shift to the lower angle region after hydrogen



Fig. 1 Thermomagnetization curve in the heating process under a magnetic field of 1 T for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.7</sub>. The inset shows powder x-ray diffraction patterns of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> with y = 0.0 and 1.7. The several vertical short bars below the diffraction patterns stand for the positions of all the possible Bragg reflections from a cubic NaZn<sub>13</sub>-type structure with the *Fm3c* space group.



Fig. 2 The Curie temperature  $T_c$  as a function of the unit cell volume for  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_y$  and  $La(Fe_{0.88}Si_{0.12})_{13}H_y$ , together with that of  $La_{1-z}Ce_z(Fe_{0.88}Si_{0.12})_{13}$ .<sup>12)</sup>

absorption, showing an isotropic expansion of the unit cell volume. Such volume expansion is caused by not only the invasion of hydrogen atom but also the spontaneous volume magnetostriction, because  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_{1.7}$  is in the ferromagnetic state at room temperature.

Figure 2 represents  $T_c$  as a function of the unit cell volume at room temperature for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> and La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub>, together with that of La<sub>1-z</sub>Ce<sub>z</sub>(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub>.<sup>12)</sup> To discuss the relation between the unit cell volume and  $T_c$  without the contribution from the large spontaneous volume magnetostriction, the data on the speci-



Fig. 3 Pressure shift of the Curie temperature  $T_c$  for  $La_{0.7}Ce_{0.3}$ -(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.7</sub>,  $La_{0.7}Ce_{0.3}$ (Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>,  $La(Fe_{0.88}Si_{0.12})_{13}H_{1.2}$  and  $La(Fe_{0.88}Si_{0.12})_{13}$ .<sup>3)</sup>



Fig. 4 Relationship between the Curie temperature  $T_c$  and the initial pressure shift  $dT_c/dP$  for  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_y$  and  $La(Fe_{0.88}Si_{0.12})_{13}H_y$ .

men with  $T_c < 273$  K are plotted. The value of  $T_c$  increases with increasing y and decreases with increasing z. The slope of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> is quite similar to that of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub>. However,  $T_c$  of the former is slightly higher than that of the latter at the same volume. In addition the volume dependence of  $T_c$  for the two compound systems mentioned above is stronger than that of La<sub>1-z</sub>Ce<sub>z</sub>(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub>.

Figure 3 depicts the pressure shift of  $T_c$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.7</sub> and La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>. For comparison, the data of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.2</sub> and La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub><sup>3)</sup> are also plotted in the same figure. Shown in Fig. 4 is the relationship between  $T_c$  and  $dT_c/dP$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> and La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub>. The value of  $dT_c/dP$  was estimated from the initial pressure shift of  $T_c$  shown in Fig. 3. The pressure shift of  $T_c$  for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> is increased by the partial substitution of Ce, whereas it is decreased by hydrogen absorption.



Fig. 5 Magnetization curves of (a)  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_{1.3}$  and (b)  $La(Fe_{0.88}Si_{0.12})_{13}H_{1.0}$  in the vicinity of the Curie temperature  $T_c$ . The increasing and decreasing processes of applied magnetic fields are indicated by the arrows.

Similarly, the pressure shift of  $T_c$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub> becomes small after hydrogen absorption. It should be noted that the pressure shift of  $T_c$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.7</sub> is larger than that for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.2</sub>, though  $T_c$  of the former is higher than that of the latter, showing that the enhancement of  $dT_c/dP$  caused by the partial substitution of Ce remains after hydrogen absorption. Accordingly,  $dT_c/dP$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> is larger than that for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> in a wide change of  $T_c$ .

The magnetization curves of (a)  $La_{0.7}Ce_{0.3}(Fe_{0.88}-Si_{0.12})_{13}H_{1.3}$  and (b)  $La(Fe_{0.88}Si_{0.12})_{13}H_{1.0}$  are shown in Fig. 5. The value of  $T_c$  for  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_{1.3}$  is about 280 K, being the same as that for  $La(Fe_{0.88}Si_{0.12})_{13}H_{1.0}$ . The increasing and decreasing processes of applied magnetic fields are indicated by the arrows. A characteristic S-shape magnetization curve with hysteresis is observed above  $T_c$  because of the itinerant-electron metamagnetic (IEM) transition. The hysteresis in the magnetization curve of  $La_{0.7}Ce_{0.3}(Fe_{0.88}Si_{0.12})_{13}H_{1.3}$  appears at higher temperatures, indicating that the critical temperature  $T_0$  of the disappearance of the IEM transition is much higher than that of  $La(Fe_{0.88}Si_{0.12})_{13}H_{1.0}$ . Such results make it clearer that the influence of the partial substitution of Ce cannot be explained by the volume contraction only.

The magnetovolume effects in itinerant-electron metamagnets have been discussed in terms of the Landau-type expansion of magnetic free energy.<sup>16</sup> In the theory, the magnetic free energy is given by the following expression by taking the effects of spin fluctuations, the magnetoelastic energy and the elastic energy into account.<sup>16</sup>

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

with

$$\tilde{a}(T) = \tilde{a}(0) + \left\{\frac{5}{3}\tilde{b}(0) + \frac{4}{3}\kappa C_{\rm mv}^2\right\}\xi(T)^2 + \frac{35}{9}\tilde{c}(0)\xi(T)^4,$$
$$\tilde{b}(T) = \tilde{b}(0) + \frac{14}{3}\tilde{c}(0)\xi(T)^2, \quad \tilde{c}(T) = \tilde{c}(0),$$
(2)

where  $\xi^2$  is the means-square amplitude of thermal spin fluctuations. The values of  $\tilde{a}(0)$ ,  $\tilde{b}(0)$  and  $\tilde{c}(0)$  are the Landau coefficients related to the 3*d* band structure, and  $\kappa C_{\rm mv}$  is the magnetoelastic coupling constant. When  $\tilde{a}(0) > 0$ ,  $\tilde{b}(0) < 0$ ,  $\tilde{c}(0) > 0$  and  $5/28 - \eta < \tilde{a}(0)\tilde{c}(0)/\tilde{b}(0)^2 < 3/16$  $(\eta = 2\kappa C_{\rm mv}^2/7|\tilde{b}(0)|)$ , the magnetic free energy has two minima at M = 0 in the paramagnetic state and at a finite value of M in the ferromagnetic state, resulting in the thermal-induced first-order transition at  $T_c$  and the IEM transition above  $T_c$ .<sup>16</sup> Under these conditions, the pressure shift of  $\xi^2$  at  $T_c$  for the first-order transition is given by<sup>16</sup>

$$\frac{d\xi(T_{\rm c})^2}{dP} = -\frac{6}{\sqrt{7}} \frac{\kappa C_{\rm mv}}{|\tilde{b}(0)|} \left[ \frac{\tilde{a}(0)\tilde{c}(0)}{\tilde{b}(0)^2} - \frac{5}{28} + \eta + 28\eta^2 \right]^{-1/2}.$$
 (3)

According to the theoretical discussion,  $\xi^2$  increases in proportion to  $T^2$  at low temperatures, and to T at high temperatures.<sup>17)</sup> It has been pointed out that the critical field  $B_c$  of the IEM transition for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> increases in line with  $T^{1.2}$  above  $T_c = 195$  K.<sup>10</sup> This result means that  $\xi^2$ for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> depends on  $T^{1.2}$  in the vicinity of 195 K, because  $B_c$  has a proportional relation with  $\xi^{2}$ .<sup>17)</sup> Thus  $d\xi(T_c)^2/dP$  of the present compound system can be approximately given by  $dT_c/dP$ . By adapting the thermodynamic relation:  $\partial F / \partial M = B$  to eq. (1), it reveals that the volume change causes not only the decrease of  $T_c$  but also the change of magnetization curve. However, it has been pointed out that the IEM transition of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.86</sub>Si<sub>0.14</sub>)<sub>13</sub> with the same  $T_{\rm c}$  is clearer than that of La(Fe<sub>0.86</sub>Si<sub>0.14</sub>)<sub>13</sub> under the hydrostatic pressure P = 0.25 GPa,<sup>14)</sup> resulting in the changes of  $\tilde{a}(0), \tilde{b}(0)$  and  $\tilde{c}(0)$ . Therefore, the enhancement of  $dT_c/dP$ due to the partial substitution of Ce is attributed to the change of the band structure as can be understood from eq. (3).

The value of  $dT_c/dP$  is also affected by the value of  $\kappa C_{mv}$  as seen from the same equation. Based on the theoretical discussion of itinerant-electron magnetism by taking the effect of spin fluctuations into account, the magnetic contribution to the thermal dependence of the volume, namely, the spontaneous volume magnetostriction  $\omega(T)$  is given by<sup>18)</sup>

$$\omega(T) = \kappa C_{\rm mv} \{ M(T)^2 + \xi(T)^2 \}.$$
(4)

Thus, the volume change  $\Delta \omega$  at  $T_c$  for the first-order transition is expressed as<sup>18)</sup>

$$\Delta\omega(T_{\rm c}) = \kappa C_{\rm mv} \{ M(T_{\rm c})_{\rm F}^{\ 2} - \xi(T_{\rm c})_{\rm P}^{\ 2} \}, \tag{5}$$

where  $M(T_c)_F$  is M at  $T_c$  in the F state and  $\xi(T_c)_P^2$  is  $\xi^2$  at  $T_c$ in the P state. Summarized in Table 1 are  $\Delta\omega(T_c)$ ,  $M(T_c)_F$ and  $\Delta\omega(T_c)/M(T_c)_F^2$  of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> and La(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.6</sub>. The values of  $\Delta\omega(T_c)$  and  $M(T_c)_F^2$  were

Table 1 The volume change  $\Delta\omega(T_c)$  at the Curie temperature  $T_c$  and magnetization  $M(T_c)$  at  $T_c$ , together with the ratio  $\Delta\omega(T_c)/M(T_c)^2$  of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> and La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.6</sub>.

	$\Delta\omega(T_{\rm c})(\%)$	$M(T_{\rm c})/\mu_{\rm B}$	$\Delta\omega(T_{\rm c})/M(T_{\rm c})^2/\mu_{\rm B}^{-2}$
La(Fe <sub>0.88</sub> Si <sub>0.12</sub> ) <sub>13</sub>	0.82	1.0	0.82
$La(Fe_{0.88}Si_{0.12})_{13}H_{1.6}$	0.54	0.9	0.67

evaluated from x-ray diffraction patterns<sup>3)</sup> and Mössbauer spectra,<sup>3,19)</sup> respectively. As seen from Table 1,  $\Delta \omega(T_c)$  of  $La(Fe_{0.88}Si_{0.12})_{13}H_{1.6}$  is about 65% of that before hydrogen absorption. On the other hand,  $M(T_c)_F^2$  for the former is about 80% of that for the latter. As a result,  $\Delta \omega(T_c)/M(T_c)_F^2$ of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.6</sub> corresponds to about 80% of that before hydrogen absorption. It is clear from eq. (5) that such behavior can be explained by the decrease of  $\kappa C_{\rm mv}$  and/ or the increase of  $\xi(T_c)_P^2$  due to the hydrogen absorption, although the values of  $\kappa C_{mv}$  and  $\xi(T_c)_P^2$  are not evaluated qualitatively at the present stage. It should be noted that the decrease of  $dT_c/dP$  for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>1.2</sub> is about 70% as shown in Fig. 4. This value is close to that of  $\Delta\omega(T_{\rm c})/M(T_{\rm c})_{\rm F}^2$ , suggesting that the decrease of  $dT_{\rm c}/dP$  for  $La(Fe_{0.88}Si_{0.12})_{13}H_y$  is mainly attributed to the decrease of  $\kappa C_{\rm mv}$  rather than the increase of  $\xi (T_{\rm c})_{\rm P}^2$ . Such influence of hydrogen absorption differs from that of the partial substitution of Ce. Consequently, it stands to reason that the influence of the partial substitution of Ce on  $dT_c/dP$  remains after hydrogen absorption, resulting in the stronger pressure effect on  $T_c$  for La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>v</sub>.

#### 4. Conclusion

The initial pressure shift of the Curie temperature  $T_c$ , namely,  $dT_c/dP$ , for La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub> becomes small after hydrogen absorption. In addition, the hydrogen absorption causes the reduction of the ratio of the volume change at  $T_c$  to the magnetization squared at  $T_c$ ,  $\Delta\omega(T_c)/M(T_c)F^2$ . Therefore, it is suggested that the decrease of  $dT_c/dP$  due to the hydrogen absorption is mainly attributed to the decrease of the magnetoelastic coupling constant  $\kappa C_{mv}$ . On the other hand,  $dT_c/dP$  is enhanced by the partial substitution of Ce for La in connection with the change of the band structure. As a result, the enhancement of  $dT_c/dP$  due to the partial substitution of Ce remains after hydrogen absorption. Consequently, the pressure effect on  $T_c$  of La<sub>0.7</sub>Ce<sub>0.3</sub>(Fe<sub>0.88</sub>-Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> is stronger than that of La(Fe<sub>0.88</sub>Si<sub>0.12</sub>)<sub>13</sub>H<sub>y</sub> in a wide change of  $T_c$ .

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