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著者	Fujieda S., Fujita A., Fukamichi K., Yamazaki Y., Iijima Y.
journal or publication title	Applied Physics Letters
volume	79
number	5
page range	653-655
year	2001
URL	http://hdl.handle.net/10097/51791

doi: 10.1063/1.1388157

Giant isotropic magnetostriction of itinerant-electron metamagnetic $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_y$ compounds

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(Received 6 March 2001; accepted for publication 1 June 2001)

$\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ compounds exhibit an itinerant-electron metamagnetic (IEM) transition above Curie temperature T_C . The IEM transition in the compound with $x=0.88$ is accompanied by a giant volume change. From a practical viewpoint, T_C was controlled by hydrogen absorption in order to obtain such a giant volume magnetostriction at room temperature. For the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound, the IEM transition occurs above $T_C=278$ K, and a significant isotropic linear magnetostriction of about 0.3% at 7 T is induced in the vicinity of room temperature. This large magnetostriction is attributed to the giant volume magnetostriction of about 1% by the IEM transition. © 2001 American Institute of Physics. [DOI: 10.1063/1.1388157]

The itinerant-electron metamagnetic (IEM) transition, that is, the first-order field-induced transition from the paramagnetic (P) to the ferromagnetic (F) state was observed in Co-based Laves phase compounds such as YCo_2 , (Ref. 1) and LuCo_2 .² The origin of the IEM transition is associated with a special $3d$ band structure which exhibits a sharp peak of the density of state (DOS) just below the Fermi level.³ It has been confirmed experimentally⁴⁻⁶ and theoretically⁷ that Co-based Laves-phase quasibinary compounds exhibit a large magnetovolume effect.

Recently, it has been demonstrated that NaZn_{13} -type $\text{La}(\text{Fe}_x\text{Si}_{1-x})_{13}$ compounds composed of 112 atoms (space group $Fm\bar{3}c$) show an IEM transition.⁸ For the compound with $x=0.88$, a first-order phase transition occurs at the Curie temperature $T_C=195$ K. In the paramagnetic state, the IEM transition is induced by applying a magnetic field. At 200 K just above T_C , this field-induced transition results in a large volume magnetostriction of about 1.5%.⁸

The magnitude of the linear magnetostriction, namely, one third of the volume change for the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}$ compound is larger than that of TbFe_2 -based compounds, well known as large magnetostrictive materials. TbFe_2 -based compounds exhibit an anisotropic linear magnetostriction,⁹ whereas the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}$ compound exhibits an isotropic volume magnetostriction. The mechanism of the volume magnetostriction originates from the onset of the magnetic moment induced by the exchange splitting of the $3d$ -electron band. Therefore, the present compounds are attractive as a type of giant isotropic magnetostrictive material. However, it is not so easy to induce a large volume magnetostriction in the vicinity of room temperature, because the IEM transition field increases with increasing temperature.¹⁰ Alternatively, the IEM transition field becomes lower as the temperature comes close to T_C . Practically, therefore, it is necessary to adjust T_C around room temperature to obtain a giant volume magnetostriction. It has been confirmed that T_C is increased up to 250 K by substituting Co for Si.^{11,12} However, the volume magnetostriction becomes obscure, implying that the

DOS curve around the Fermi level is modified by the substitution.

Another useful method to increase T_C is to introduce interstitial atoms such as B, C, N, and H. Some hydrides have been synthesized and their magnetic properties have been investigated.¹³ In addition, a hydrogen absorption into NaZn_{13} -type $\text{La}(\text{Fe}_x\text{Al}_{1-x})_{13}$ compounds brings about a drastic change in the transition temperature.¹⁴ In order to obtain a giant volume magnetostriction at room temperature, therefore, the increase in T_C is examined by hydrogen absorption.

$\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}$ was prepared by arc melting in an Ar gas atmosphere. Heat treatment for homogenization was carried out in a vacuum quartz tube at 1323 K for 10 days.⁸ X-ray powder diffraction identified the phase in all the specimens as a NaZn_{13} -type single phase. Hydrogen absorption was carried out by differential scanning calorimeter (DSC) in a H_2 atmosphere of 5 MPa and Ar atmosphere of 0.1 MPa. After hydrogen absorption, the mechanical strength of the sample decreased, however, the form of the sample was kept without any trouble for measurement of the linear magnetostriction. The magnetization and magnetostriction were measured by an induction method and a three-terminal capacitance method, respectively.

Figure 1 shows the room-temperature lattice constant a against the Curie temperature T_C determined by thermomagnetoization measurements. It was confirmed that the lattice constant is increased by hydrogen absorption without a change in the NaZn_{13} -type structure. From the gas-chromatograph analysis for $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_y$, the maximum concentration of hydrogen y was determined to be 1.6. The value of T_C was increased up to 336 K, accompanied by a volume expansion of about 3%. This volume expansion is caused by not only the hydrogen absorption but also the magnetovolume effect, because $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.6}$ is ferromagnetic at room temperature. The lattice constant in the ferromagnetic range is apparently larger than that in the paramagnetic range. Therefore, a very large magnetovolume effect is preserved after hydrogen absorption.

For $y=1.0$ with $T_C=278$ K, a thermally induced phase transition occurs around room temperature. Figure 2 shows

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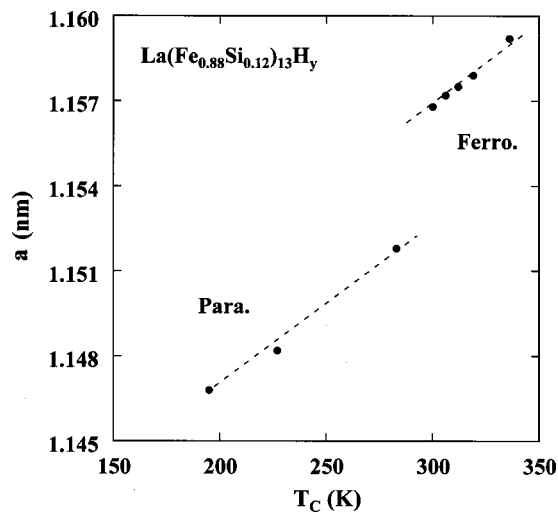


FIG. 1. Room-temperature lattice constant a against the Curie temperature T_C .

the x-ray diffraction patterns around T_C for $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$. The diffraction patterns in both the F and the P states exhibit the NaZn_{13} -type structure. The peak positions show a significant shift around T_C , and the coexistence of peaks in the F state with a large volume and the P state with a small volume is confirmed at 280 K. Such a discontinuous peak shift suggests that a thermally induced first-order phase transition also takes place after hydrogen absorption.

Figure 3 shows the temperature dependence of the relative volume change determined from x-ray diffraction patterns for the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound. Due to the first-order phase transition, the volume is changed discontinuously at T_C . There is a magnetic contribution to the thermal dependence of the volume change, which is called spontaneous volume magnetostriction ω_s . For itinerant electron magnets, ω_s is expressed by using the local magnetic moment $M(T)$ and the amplitude of thermal spin fluctuations $\xi(T)$ as¹⁵

$$\omega_s = \kappa C_{\text{mv}} \{M^2(T) + \xi^2(T)\}, \quad (1)$$

where κ and C_{mv} are the compressibility and the magneto-

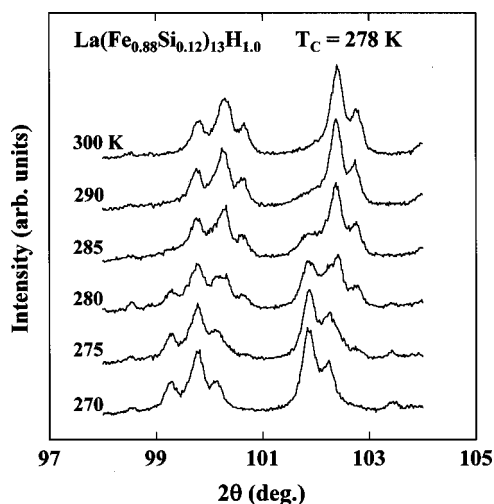


FIG. 2. X-ray diffraction patterns around the Curie temperature for the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound.

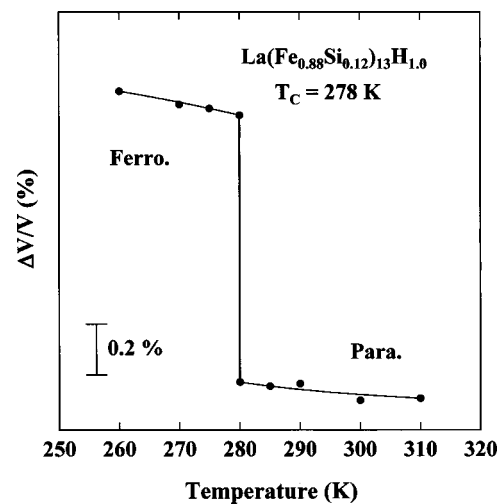


FIG. 3. Temperature dependence of the relative volume change for the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound.

volume coupling constant, respectively. In the F state, the contribution of the local magnetic moment to the volume is dominant, because the thermal spin fluctuations are suppressed below the Curie temperature. On the other hand, the local moment becomes zero in the P state. In the P state, the volume is dominated by thermal spin fluctuations. The difference in volume $\Delta\omega(T_C)$ is expressed by the following expression:⁷

$$\Delta\omega(T_C) = \kappa C_{\text{mv}} \{M_F^2(T_C) - \xi_P^2(T_C)\}. \quad (2)$$

For the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound, the difference in volume between the F and P states is about 1%. This result means that $M_F(T_C)$ becomes smaller and $\xi_P(T_C)$ becomes larger with increasing T_C , and hence, the value of $\Delta\omega(T_C)$ becomes smaller than that of the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}$ compound after hydrogen absorption.

Figure 4 shows the magnetization curves as a function of temperature for the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound around T_C . A characteristic S-shaped curve with a hysteresis is observed above T_C . This means that a field-induced first-order phase transition also takes place after hydrogen absorption.

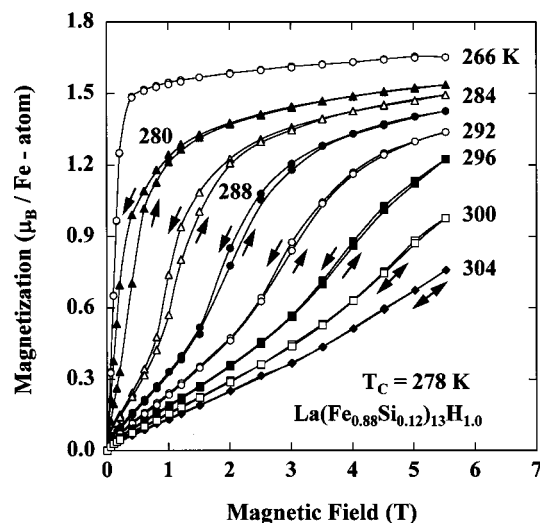


FIG. 4. Magnetization curves of the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound as a function of temperature in the vicinity of the Curie temperature.

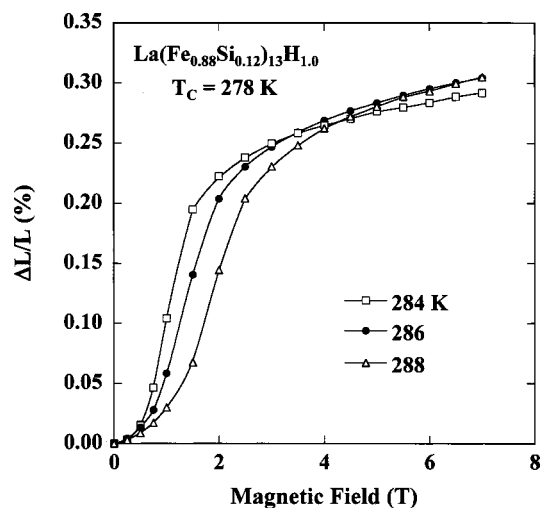


FIG. 5. Magnetic-field dependence of the linear magnetostriction parallel to the magnetic-field direction just above the Curie temperature for the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound.

In other words, the IEM transition at room temperature is realized by hydrogen absorption. This IEM transition is very sharp, different from the result reported for the substituted system.^{11,12}

The present result implies that the change of the DOS curve around the Fermi level is negligible after hydrogen absorption. From a practical viewpoint, therefore, hydrogen absorption is one of the useful methods to control T_C .

In order to elucidate the magnitude of the volume magnetostriction due to the IEM transition, the linear magnetostriction parallel to the magnetic-field direction was measured. The data of the $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})\text{H}_{1.0}$ compound are shown in Fig. 5. By applying the magnetic field, a significant change of the linear magnetostriction is observed around room temperature. The magnitude of the linear magnetostriction $\Delta L/L$ corresponds to the value of the discontinuous linear thermal expansion $\Delta\omega(T_C)/3$ due to the disappearance of the local magnetic moment shown in Fig. 3. This means that applying the magnetic field just above T_C restores the amplitude of the local magnetic moments. The temperature dependence of the linear magnetostriction is correlated to the temperature dependence of the IEM transition field. Accordingly, such a large linear magnetostriction is attributed to the giant volume magnetostriction of about 1% by the IEM transition around room temperature. The magnitude of a linear magnetostriction at 7 T is about 0.3%, larger than the room-temperature value of TbFe_2 -based compounds. With decrease

ing temperature, a large value is obtainable in lower magnetic fields, as seen from Fig. 5. Therefore, such a giant magnetostriction is expected between T_C and 284 K in much lower applied magnetic fields, because the critical field of the IEM transition tends to become zero at T_C .

In conclusion, in order to obtain a giant volume magnetostriction at room temperature, Curie temperature T_C was adjusted to around room temperature by hydrogen absorption. The $\text{La}(\text{Fe}_{0.88}\text{Si}_{0.12})_{13}\text{H}_{1.0}$ compound exhibits a thermally induced first-order transition at $T_C=278$ K. By applying a magnetic field above T_C , an IEM transition is caused, accompanied by a giant volume change of about 1%. This volume magnetostriction is isotropic, therefore, one third corresponds to the linear magnetostriction. Furthermore, it should be emphasized that no crystallographic structure controls are necessary in contrast to conventional magnetostrictive materials. Consequently, the present compound is a practical one for giant isotropic magnetostrictive materials.

One of the authors (K.F.) would like to acknowledge support by the Grant-in-Aid for Scientific Research on Priority Areas (A) (Area No. 299) from the Japanese Ministry of Education, Science and Culture. The present work has been partially supported by the Industrial Technology Research Grant Program in '00 (Grant No. 00A26019a) from the New Energy and Industrial Technology Development Organization (NEDO) of Japan.

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