Magnetoresistance in $LaFe_{11.2}Co_{0.7}Si_{1.1}$ Compound

F. X. Hu, J. Gao, X. L. Qian, Y. X. Li, J. Du, J. R. Sun, and B. G. Shen

***Abstract*—Magnetoresistance has been studied in $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound. The ferromagnetic ordering at Curie temperature T_C of 274 K was found being accompanied by a drastic negative lattice expansion due to the strong structural and magnetic interplay. Such a simultaneous magnetic and lattice change would cause changes in transport properties. The measured transport properties indicate that the transition can be induced by temperature or applied magnetic field. The sample shows a metallic character below T_C, whereas the electrical resistance decreases dramatically and then recovers the metal-like behavior above T_C. Application of a magnetic field retains the transitions via increasing the ferromagnetic ordering temperature. An isothermal increase of field leads to an increase of resistance at temperatures near but above T_C, which is a result of the field-induced metamagnetic transition from paramagnetic to ferromagnetic state.**

***Index Terms*—Magnetocaloric effect, magnetoresistance, $NaZn_{13}$-type compounds.**

I. Introduction

COMPOUNDS with cubic $NaZn_{13}$-type structure have been suggested to be appropriate materials for exploring efficient magnetic refrigerants [1]–[6]. The great magnetic entropy change recently reported in this class of ferromagnetic materials [3]–[6] warrants further experimental and theoretical studies. The achieved giant magnetic entropy change has been demonstrated to be due to a transition from the low-temperature ferromagnetic (FM) phase to the high-temperature paramagnetic (PM) phase, which is accompanied by a drastic negative lattice expansion [3]–[6]. The simultaneous magnetic and lattice change should cause changes in transport properties. However, there is little information on the electrical-transport properties of this class of materials.

The $NaZn_{13}$-type compounds $LaFe_{13-x}Si_x$ with low Si content show an itinerant electron metamagnetic transition above Curie temperature T_C and a negative lattice expansion at T_C. With decreasing Si content the nature of the phase transition at T_C evolves from second-order to first-order and the first-order nature is strengthened by further reducing the Si content. Furthermore, incorporation of Co weakens the nature of the first-order transition and drives T_C to higher temperature. A proper combination of Si and Co could make T_C near room temperature and remain first-order nature of the transition at T_C, characterized by a sharp change of lattice parameter. Therefore, we synthesized a $NaZn_{13}$-type compound $LaFe_{11.2}Co_{0.7}Si_{1.1}$ with a drastic negative lattice expansion at T_C of 274 K and achieved a great magnetic entropy change near room temperature [4]. It is known that both magnetic and structural transformations can lead to the changes in the charge carrier concentration and in the scattering mechanisms, which can be detected from the temperature and magnetic-field dependencies of the electrical resistance. In this paper, we report on the experimental results of the temperature and magnetic-field dependencies of the electrical resistance in such a compound $LaFe_{11.2}Co_{0.7}Si_{1.1}$.

Manuscript received October 15, 2003; revised January 5, 2004. This work was supported by the Beijing Nature Science Foundation of China, the State Key Project of Fundamental Research, the National Natural Science Foundation of China, and a grant from the Research Grant Council of Hong Kong, China.

F. X. Hu is with the Department of Physics, Capital Normal University, Beijing 100037, China, the State Key Laboratory of Magnetism, Institute of Physics and Center of Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China, and the Department of Physics, The University of Hong Kong, Hong Kong, China (e-mail: fxhu2002@hku.hk).

J. Gao is with the Department of Physics, The University of Hong Kong, Hong Kong, China (e-mail: jugao@hku.hk).

X. L. Qian is with the Department of Physics, Capital Normal University, Beijing 100037, China (e-mail: xlqian1953@sohu.com).

Y. X. Li and J. Du are with the School of Material Science and Engineering, Hebei University of Technology, Tianjin 300130, China (e-mail: yxli@hebut.edu.cn).

J. R. Sun and B. G. Shen are with the State Key Laboratory of Magnetism, Institute of Physics and Center of Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, China (e-mail: jrsun@g203.iphy.ac.cn; bgshen@cashq.ac.cn).

Digital Object Identifier 10.1109/TMAG.2004.832500

II. Experiments

The samples employed in the present investigation were prepared by repeatedly arc-melting appropriate amounts of the starting materials with purity of 99.9 wt%. The ingots were wrapped with Ta foil and subsequently homogenized in a vacuum-sealed quartz tube for 30 days at 1323 K, then quenched in liquid nitrogen. Quenching is important to obtain a stable compound with low Si content for this class of alloys. A single phase of the $NaZn_{13}$-type structure was identified by X-ray diffraction analysis. A minor phase α-Fe was observed. The amount of the impurity a-Fe phase is estimated to be 6 wt% based on the chemical analysis by inductively coupled plasma-atomic emission spectrometry (ICP-AES). The sample used for electrical-resistance measurements was cut from the main sample by spark erosion and had the dimensions $2 \times 2 \times 5$ mm^3. The electrical resistance measurements were performed using a superconducting quantum interference device (SQUID) magnetometer equipped with a probe for making four-point electrical resistance measurements. The measurements were carried out using a current of 10 mA in the temperature range from 5 to 380 K and in magnetic fields from 0 to 4 T. The magnetic field vector was oriented parallel to the direction of the electrical current. Curie temperature T_C of present sample was determined to be $\sim$274 K from the temperature dependence of magnetization measured under a field of 0.01 T by SQUID.

III. Results and Discussion

X-ray powder diffraction (XRD) at various temperatures was performed in order to study the structure change on

0018-9464/04$20.00 © 2004 IEEE

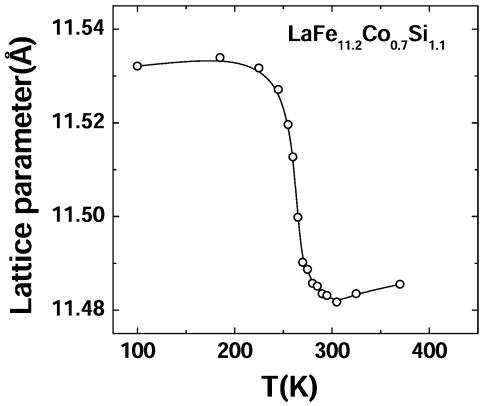

Fig. 1. Temperature dependence of lattice parameter of $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound.

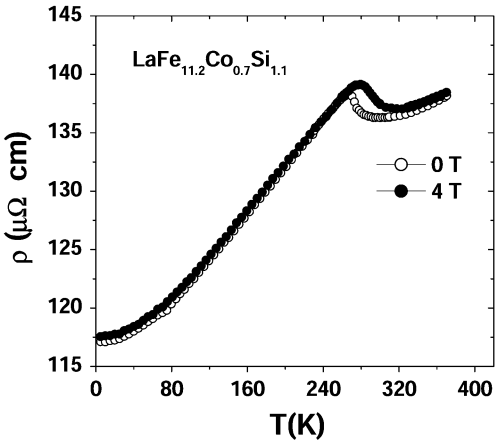

Fig. 2. Temperature dependence of electrical resistance measured in zero and 4T field on cooling for $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound.

altering magnetic state with temperature. It is found that the crystal structure remains cubic $NaZn_{13}$-type, but the lattice parameter changes dramatically at T_C, implying a first-order transition. Fig. 1 shows the temperature dependence of lattice parameter obtained from the XRD spectrum, noting the large negative thermal expansion near T_C. The lattice parameter at ferromagnetic state is bigger than that at paramagnetic state by $\sim$0.44% in the vicinity of T_C. Careful measurements of ac susceptibility on heating and cooling indicate temperature hysteresis of the transition is small, $\sim$1.5 K.

The temperature dependence of electrical resistance of $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound measured in zero and 4 T field in cooling process is presented in Fig. 2. It can be seen that there is an anomaly in the temperature dependence of the electrical resistance at T_C in the absence of field. Below T_C, the electrical resistance increases with increasing temperature and has metallic character, but above T_C, it decreases dramatically in a narrow temperature range and then recovers the metal-like dependence on temperature. It is noteworthy that in our sample the total contribution from both the electron–phonon scattering and the electron–magnon scattering in the PM phase is smaller than that in the FM phase, which is contrary to normal FM metallic materials and also much different from other systems showing giant magnetocaloric effect, such as $Gd_5Si_2Ge_2$ alloy [7]. The giant magnetocaloric effect in $Gd_5Si_2Ge_2$ comes from the simultaneous magnetic and structure transition from low-temperature FM orthorhombic to high-temperature PM monoclinic phase [7]. The monoclinic PM phase in $Gd_5Si_2Ge_2$ shows a pronounced higher resistance than the orthorhombic FM phase [8]. It has been argued that the change in the electron-phonon scattering during the transition is the primary mechanism responsible for the increased resistance in the PM monoclinic phase. The monoclinic lattice of PM phase is more disordered compared to the orthorhombic lattice of FM phase, resulting in the increased electron-phonon scattering [8]. It is generally believed that the most important contributions to the electrical resistivity during the first-order magnetic and crystallographic phase transition are the electron-phonon scattering and the electron-magnon scattering. In most FM metallic materials, the electron-magnon component usually approaches maximum at T_C and becomes weakly dependent on temperature above T_C. In present $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound, magnetic phase transition at T_C is accompanied by a drastic change of lattice parameter remaining $NaZn_{13}$-type structure. It is naturally considered that the change of electron–phonon scattering upon phase transition might be responsible for the reduction of resistance in the PM phase of $LaFe_{11.2}Co_{0.7}Si_{1.1}$. The drastic large change of the lattice parameter may influence the density of states (DOS) around the Fermi level and affect the electron-phonon scattering. A quantitative explanation for the decreased resistance in PM phase with shrunk lattice requires the detailed information of band structure.

From Fig. 2, it can be also found that the application of magnetic field retains the transition via increasing the ferromagnetic ordering temperature. The transition temperature increases from 274 to 294 K upon applying a field of 4 T. The resistance at temperatures away from T_C keeps nearly unchanged upon the application of 4T field. However, in the temperature range from 274 to 294 K, application of magnetic field does result in an increase of resistance.

The isothermal magnetic-field-dependent resistance measured upon field increase and decrease at temperatures near T_C is shown in Fig. 3. It can be found that the isothermal increase of the magnetic field leads to an increase of electrical resistance at temperatures close but above T_C. This is a result of the field-induced itinerant-electron metamagnetic (IEM) transition from paramagnetic to ferromagnetic state [3]. Generally, the origin of the IEM transition is associated with a special 3-D band structure which exhibits a sharp peak of the DOS just below the Fermi level [9]. The field hysteresis of the transition in the field cycles is very small in present system, which is consistent with the small temperature hysteresis as aforementioned.

The phase transition in present compound is first-order in nature. In real materials, a first-order transition takes place generally not at a temperature point but in a temperature interval [10]. The Curie temperature T_C of present compound is found at $\sim$274 K, which means the FM and PM phases may coexist in a small temperature region around 274 K. Those R–H curves measured at different temperatures near T_C reflect the competition of the FM and PM phases (Fig. 3). For the R–H curve measured at 270 K, close but below T_C, the small increase of R with

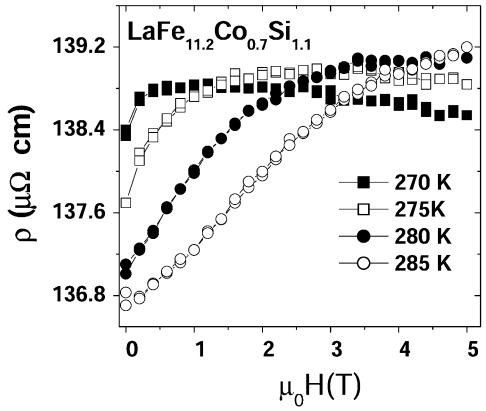

Fig. 3. Isothermal magnetic-field dependence of resistance of $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound with increasing and decreasing fields at temperatures 270, 275, 280, and 285 K.

increasing H might be related to the existence of a small amount of PM phase. Further increasing H results in a slow decrease of R, which is an effect of general electron-magnon scattering in FM phase. The content of PM phase gradually increases with increasing temperature. The result of competition between FM and PM phases makes the R at 275 K firstly increase and then nearly keep unchanged with increasing H. At 280 and 285 K, R increases monotonously with H because the most contents are PM phase. On the other hand, anisotropic magnetoresistance (AMR) phenomena in present compound should be very small due to its high symmetric structure of cubic $NaZn_{13}$-type in both FM and PM states.

In summary, the magnetic phase transition from PM to FM in $LaFe_{11.2}Co_{0.7}Si_{1.1}$ compound is accompanied by a drastic large increase of lattice parameter remaining the $NaZn_{13}$-type structure. The temperature and field dependence of the electrical resistance indicates that the transition can be induced both by temperature and by magnetic field. The transition is of first-order in nature with a small temperature and field hysteresis. The change of electron-phonon scattering during the transition may be responsible for the decreased resistance in PM phase. The magnetic-field-induced IEM transition from PM to FM state leads to a positive magnetoresistance at temperatures close but above T_C.

REFERENCES

[1] F. X. Hu, B. G. Shen, J. R. Sun, Z. H. Cheng, and X. X. Zhang, "Magnetic entropy change in $La(Fe_{0.98}Co_{0.02})_{11.7}Al_{1.3}$," *J. Phys.: Condens. Matter.*, vol. 12, p. L691, 2000.

[2] F. X. Hu, B. G. Shen, J. R. Sun, and Z. H. Cheng, "Large magnetic entropy change in $La(Fe,Co)_{11.83}Al_{1.17}$," *Phys. Rev. B*, vol. 64, p. 012 409, 2001.

[3] F. X. Hu, B. G. Shen, J. R. Sun, Z. H. Cheng, G. H. Rao, and X. X. Zhang, "Influence of lattice negative expansion and metamagnetic transition on magnetic entropy change in the compound $LaFe_{11.4}Si_{1.6}$," *Appl. Phys. Lett.*, vol. 78, p. 3675, 2001.

[4] F. X. Hu, B. G. Shen, J. R. Sun, G. J.Wang, and Z. H. Cheng, "Very large magnetic entropy change near room temperature in $LaFe_{11.2}Co_{0.7}Si_{1.1}$," *Appl. Phys. Lett.*, vol. 80, p. 826, 2002.

[5] F. X. Hu, M. Ilyn, A. M. Tishin, J. R. Sun, G. J. Wang, Y. F. Chen, F. Wang, Z. H. Cheng, and B. G. Shen, "Direct measurements of magnetocaloric effect in first-order system $LaFe_{11.7}Si_{1.3}$," *J. Appl. Phys.*, vol. 93, p. 5503, 2003.

[6] A. Fujita, S. Fujieda, Y. Hasegawa, and K. Fukamichi, "Itinerant-electron metamagnetic transition and large magnetocaloric effects in $La(Fe_xSi_{1-x})_{13}$ compounds and their hydrides," *Phys. Rev. B*, vol. 67, p. 104 416, 2003.

[7] V. K. Pecharsky and K. A. Gschneidner, Jr., "Giant magnetocaloric effect in $Gd_5Si_2Ge_2$," *Phys. Rev. Lett.*, vol. 78, p. 4494, 1997.

[8] E. M. Levin, V. K. Pecharsky, and K. A. Gschneidner, Jr., "Magnetic-field and temperature dependencies of the electrical resistance near the magnetic and crystallographic first-order phase transition of $Gd_5(Si_2Ge_2)$," *Phys. Rev. B*, vol. 60, p. 7993, 1999.

[9] H. Yamada, "Metamagnetic transition and susceptibility maximum in an itinerant-electron system," *Phys. Rev. B*, vol. 47, p. 11 211, 1993.

[10] J. R. Sun, F. X. Hu, and B. G. Shen, "Comment on "Direct measurement of the 'giant' adiabatic temperature change in $Gd_5Si_2Ge_2$"," *Phys. Rev. Lett.*, vol. 85, p. 4191, 2000.