Vol 13 No 8, August 2004
1009-1963/2004/13(08)/1344-04

Chinese Physics

© 2004 Chin. Phys. Soc.
and IOP Publishing Ltd

Change in the magnetic ground state of $LaFe_{11.4}Al_{1.6}$ compound by the substitution of Mn for Fe*

Wang Fang(王 芳)†, Chen Yuan-Fu(陈远富), Wang Guang-Jun(王光军), Sun Ji-Rong(孙继荣), and Shen Bao-Gen(沈保根)

State Key Laboratory of Magnetism, Institute of Physics and Centre of Condensed Matter Physics, Chinese Academy of Sciences, Beijng 100080, *China*

(Received 9 January 2004; revised manuscript received 13 April 2004)

The structure and magnetic properties of $La(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ ($0 \leq x \leq 0.25$) compounds have been studied. The $NaZn_{13}$-type structure is preserved and the lattice parameter increases linearly with increasing the Mn concentration. The magnetic ground state changes from the antiferromagnetic to the spin-glass or the cluster-glass state by the substitution of Mn for Fe. Furthermore, a field-induced transition from cluster glass to ferromagnet is found for the samples with x=0.05 and 0.10.

Keywords: $La(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ compounds, magnetic ground state, spin glass, Arrott plot
PACC: 7550E, 7550L

1. Introduction

$La(Fe_xAl_{1-x})_{13}$ compounds in the concentration range $0.46 \leq x \leq 0.92$ crystallize in the cubic $NaZn_{13}$-type structure with the space group of $Fm3c$, in which Fe and Al atoms form the icosahedron.[1] Previous studies have shown that the changes in the lattice parameter and the coordination number of Fe by Al can bring about a change in the magnetic ground state.[2,3] The micromagnetic state is observed in the concentration range $0.46 \leq x \leq 0.62$ due to the competition between a nearest-neighbour Fe–Fe ferromagnetic exchange and a next nearest neighbour Fe–Al–Fe antiferromagnetic exchange. Ferromagnetic state is observed in the range $0.62 < x < 0.86$. For higher iron concentration range $0.86 \leq x \leq 0.92$ the antiferromagnetic state is found because of the short distance between Fe–Fe atoms.

Recent studies have confirmed that the magnetic ground state of $La(Fe_xAl_{1-x})_{13}$ compounds can also be changed by the substitution of Co for Fe or by introducing interstitial H or N atoms.[4–7] Very recently, we observed that the substitution of Mn for Fe can obviously change the magnetic properties, especially the magnetic ground state of $La(Fe_{1-x}Mn_x)_{11.4}Si_{1.6}$ compounds.[8] In the present work, antiferromagnetic state $LaFe_{11.4}Al_{1.6}$ is selected as a parent compound and the effects of the substitution of Mn for Fe on the structure and the magnetic properties, especially the magnetic ground state of $La(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ compounds are systematically investigated.

2. Experimental procedure

A series of $La(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ compounds ($0 \leq x \leq 0.25$) were prepared by arc melting under a high-purity argon atmosphere. The purity of the raw materials La, Fe, Mn and Al are better than 99.9%. An excess 10at% of La and Mn over the stoichiometric composition was added to compensate for the losses during melting. The arc-melted ingots were homogenized at 950^0C for 10 days and quenched quickly into the liquid nitrogen. X-ray diffraction (XRD) measurements on powder samples using Cu$K\alpha$ radiation were performed to identify the single phase and to determine the crystallographic structure. The magnetic measurements were performed on a superconducting quantum interference device (SQUID) magne-

*Project supported by the State Key Development Programme for Basic Research of China (Grant No G1998061303), by the National High Technology Development Program of China (Grant No 2002AA324010), and by the National Natural Science Foundation of China (Grant No 10174094).

†Corresponding author. E-mail: sdfang@g203.iphy.ac.cn

tometer and a physical property measurement system (PPMS).

3. Results and discussion

The XRD results show that all the samples crystallize in a single phase with the cubic $NaZn_{13}$-type structure ($Fm3c$) at room temperature. As shown in Fig.1, the increase in the Mn concentration results in a volume expansion due to the larger atomic size of the Mn atoms in comparison to that of the Fe ones. In the $NaZn_{13}$ structure Fe^{I} atoms are surrounded by an icosahedron of 12 Fe^{II} atoms and possess a local symmetry peculiar to FCC-like lattice. Previous studies have shown that the Fe–Fe exchange interaction is sensitive to the Fe–Fe bond length d.[9,10] It is antiferromagnetic interaction for $d \leq 0.244$nm and ferromagnetic for $d > 0.244$nm. Furthermore, there is an optimal d for the Fe–Fe exchange interaction. The value of d in the $LaFe_{11.4}Al_{1.6}$ compound is slightly smaller than 0.244nm, which results in the antiferromagnetic ground state. The temperature dependence of magnetization exhibits a paramagnetic-to-antiferromagnetic transition at the Néel temperature T_N of 217K as shown in the inset of Fig.2. The substitution of Mn for Fe brings about a volume expansion, accordingly the Fe–Fe bond length d will increase and the magnetic state of the Fe atoms may undergo strong change. In this case a ferromagnetic ground state is expected. However, a spin-glass-like (SGL) behaviour, instead of the ferromagnetic ground state, is found, as shown in Fig.2 (M_{ZFC} andM_{FC} correspond to the magnetizations measured in zero field cooling and field cooling modes, respectively). M_{FC} for the sample with x=0.05 shows a monotonic increase with decreasing the temperature, whereas M_{ZFC} exhibits a broad maximum around ~106K, below which a significant deviation from M_{FC} occurs. For the samples with $x \geq 0.10$, the M_{FC} curves show a cusp at temperature T_f=46K (x=0.10), 31K (x=0.15), 26K (x=0.20) and 21K (x=0.25). T_f monotonically decreases with increasing Mn concentration. These behaviours are also usually observed in the spin-glass or the cluster-glass systems.[11,12]

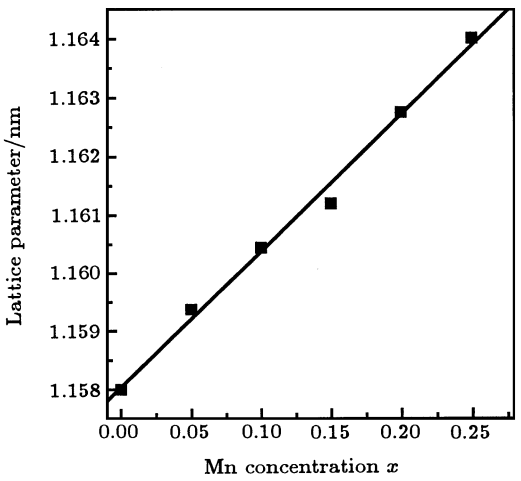


Fig.1. Mn concentration x dependence of the lattice parameter for $La(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ ($0 \leq x \leq 0.25$) compounds.

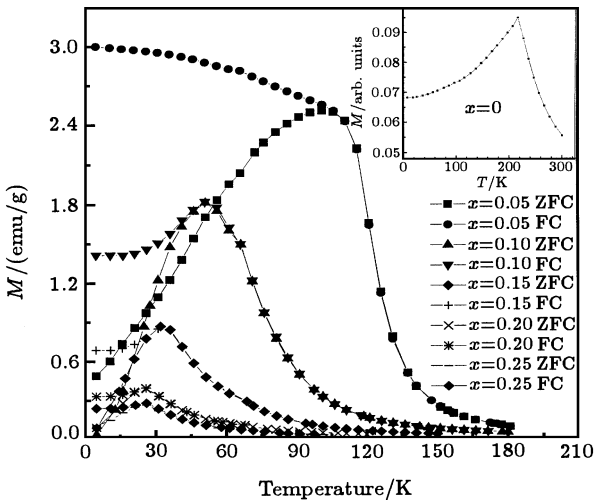


Fig.2. Temperature dependence of magnetization measured in zero field cooling and field cooling modes for La $(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ ($0.00 \leq x \leq 0.25$) compounds under a low magnetic field of 0.01T.

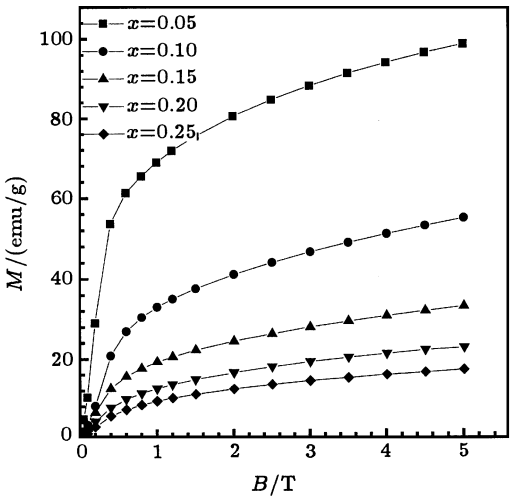
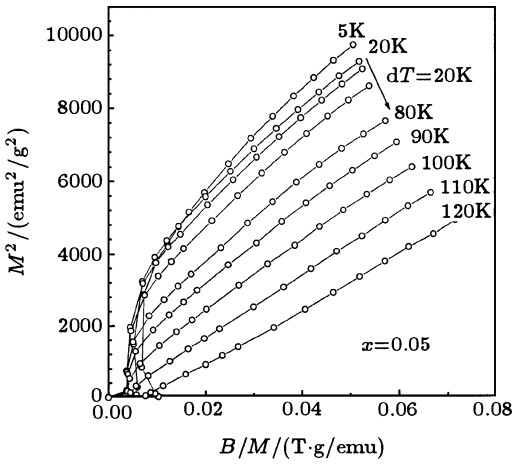


Fig.3. Magnetic field dependence of magnetization for La $(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ ($0.05 \leq x \leq 0.25$) compounds at 5K.

Magnetization curves $M(H)$ for all the samples are measured at 5K and the results are shown in Fig.3. It shows an obvious decrease of the magnetization with the increase of x, and the full saturation state is not

achieved even under the applied magnetic field of 5T for all the samples. It is also one of the characteristic features in spin glasses.

To further verify the magnetic state, $M(H)$ under the ZFC condition for all the samples was also measured up to 5T at different temperatures; the Arrott plots of the results for x=0.05 and x=0.15 are shown in Fig.4. The curves for x=0.05 and 0.10 display a negative slope below 106K and 45K, which suggests a field-induced transition from a disordered state to a long-range ordered state. The M^2-H/M curve approaches a linear line, and its interception with the M^2-axis varies from a positive value to a negative one at 106K and 45K for the samples with x=0.05 and 0.10, respectively. There is no positive interceptions at any temperature for the sample with $x \geq$0.15, which confirms the absence of long-range ferromagnetic order.[13]



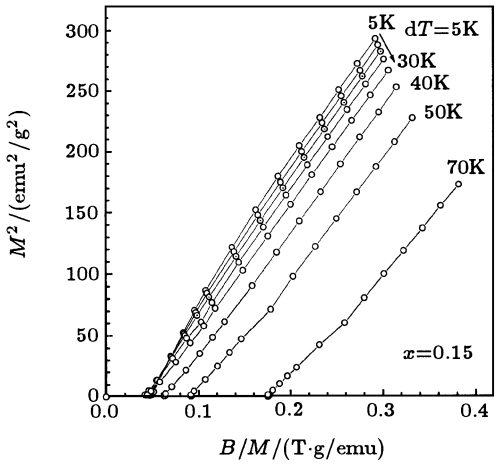


Fig.4. Arrott plots of magnetization at different temperatures for compounds with x=0.05 and x=0.15.

A time-dependent magnetization is one of the key features of spin glasses. We measured the magnetic relaxation of the sample x=0.05 at 5K under the applied fields of 0.01, 0.1 and 0.5T. From Fig.5 one can see that the magnetization increases with waiting time at the applied field of 0.01 and 0.1T. However, the magnetization was time independent under the field of 0.5T, which means that the applied field of 0.5T is sufficient to destroy the cluster glass behaviour for the sample with x=0.05. This is also confirmed by the thermomagnetic curves under different applied fields as shown in Fig.6. Combined all the results obtained, one can see that samples for x=0.05 and 0.10 show a cluster-glass behaviour with a finite ferromagnetic correlation. And samples in the Mn concentration region $0.25 \geq x \geq 0.15$ exhibit a spin-glass behaviour with no long-rang ordering.

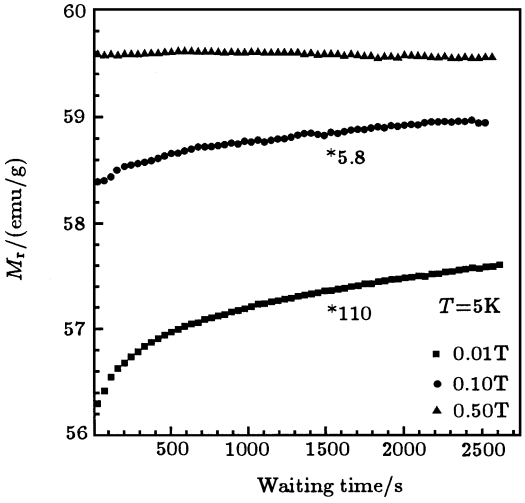


Fig.5. Magnetic relaxation of the sample x=0.05 at 5K under the applied fields of 0.01, 0.1 and 0.5T.

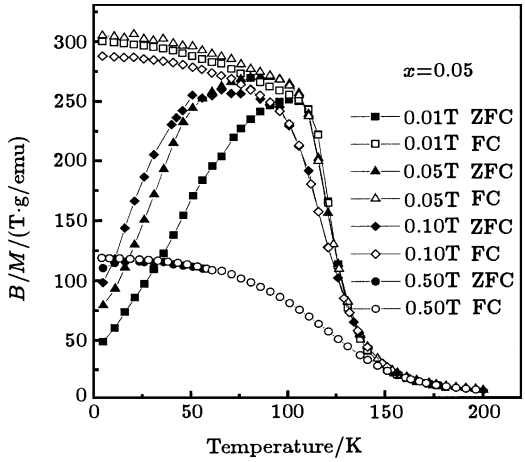


Fig.6. The thermomagnetic curves under different applied fields 0.01, 0.05, 0.1 and 0.5T for compound with x=0.05.

Hereafter, we take the sample with x=0.15 as the example to discuss the dynamical properties of the spin glass systems.

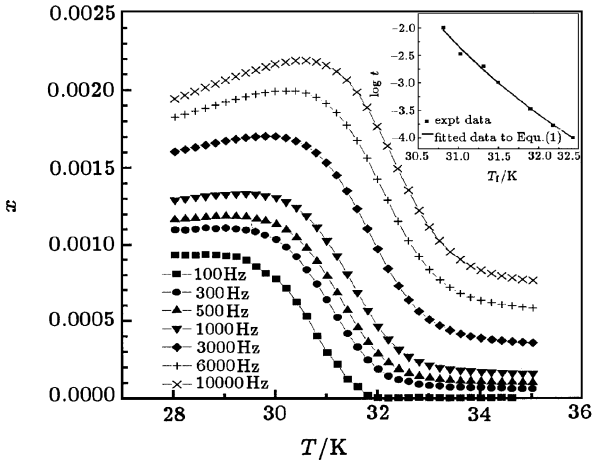


Fig.7. The temperature dependence of the out-of-phase Ac magnetic susceptibility χ'' around the freezing temperature at different frequencies (f) for compound with x=0.15.

Figure 7 shows the temperature dependence of the out-of-phase Ac magnetic susceptibility χ'' around the freezing temperature at different frequencies (f). The inflection point of $\chi''(\omega, T)$ show a variance with the frequency. It is about 30K for frequency f=100Hz and shifts towards higher temperature with the increase of the frequency. The divergence of the maximum relaxation time $\tau_{\max}$, occurring at the spin-glass transition temperature, can be investigated by using the conventional critical slowing down law:

$$\frac{\tau_{\max}}{\tau_0} = \xi^{-zv} = \left(\frac{T_{\rm f}(\omega) - T_{\rm SG}}{T_{\rm SG}}\right)^{-zv}. \qquad (1)$$

Here, zv is the dynamic exponent and τ_0 is a microscopic relaxation time, $T_{\rm f}(\omega)$ is defined by the temperature corresponding to the inflection of the $\chi''(\omega, T)$, $T_{\rm SG}$ is the spin glass transition temperature.[14] The best fit of the measured data to Eq.(1) is shown in the inset of Fig.7, and yields the values τ_0=$10^{-11.3}$s, zv=8.4, and $T_{\rm SG}$=28.6K. Both values of τ_0 and zv are very similar to those found in ordinary PM to SG transitions.[14−16] The sensitivity of $T_{\rm f}(\omega)$ to frequency, defined as $\Delta T_{\rm f}/(T_{\rm f}(\omega)\Delta \log_{10}\omega)$, which has been used as a criterion to assert the presence of a spin-glass phase, is 0.03. It is very close to that of the conventional spin glass system.[17]

Based on the results above, one can find that with the incorporation of Mn, which produces a volume expansion, the Fe–Fe exchange experiences an antiferromagnetic to ferromagnetic transition. Meanwhile, a Fe–Mn antiferromagnetic coupling is induced. The competition between ferromagnetic and antiferromagnetic exchange interactions results in the interesting magnetic properties in $La(Fe_{1-x}Mn_x)_{11.4}Al_{1.6}$ compounds.

4. Summary

In conclusion, the substitution of Mn for Fe brings about obvious change in $LaFe_{11.4}Al_{1.6}$ compound. The lattice parameter increases almost linearly with increasing Mn concentration. A cusp in the ZFC susceptibility curves, a distinctive separation of the FC and ZFC curves below freezing temperatures, the frequency dependence of the Ac susceptibility, and the time dependence of magnetization confirm the occurrence of the spin-glass or cluster-glass state. Furthermore, for the samples with $x \leq$0.10, a field above 0.5T can induce a ferromagnetic state.

References

[1] van der Kraan A M, Buschow K H J and Palstra T T M 1983 *Hyperfine Int.* **15-16** 717

[2] Palstra T T M, Werij H G C, Nieuwenhuys G J, Mydosh J A, de Boer FR and Buschow K H J 1984 *J. Phys.* F **14** 1961

[3] Palstra T T M, Nieuwenhuys G J, Mydosh JA and Buschow K H J 1984 *J. Appl. Phys.* **55** 2367

[4] Hu F X, Shen B G, Sun J R and Cheng Z H 2001 *Phys. Rev.* B **64** 132412

[5] Irisawa K, Fujita A, Fukamichi K, Yamazaki Y, Iijima Y and Matsubara E 2001 *J. Alloys Compounds* **316** 70

[6] Liu J P, Tang N, de Boer F R, de Chatel P F and Buschow K H J 1995 *J. Magn. Magn. Mater.* **140-144** 1035

[7] Ye Shcherbakova V, Korolyov A V and Podgornykh S M 2001 *J. Magn. Magn. Mater.* **237** 147

[8] Wang F, Zhang J, Chen Y F, Wang G J, Sun J R and Shen B G 2004 *Phys. Rev.* B **69** 094424

[9] Givord D and Lemaire R 1974 *IEEE Trans. Magn. MAG.* **10** 109

[10] Wang F W, Zhang P L, Wang J Y, Sun X D, Yan Q W, Shen B G and Zhang L G 1999 *J. Magn. Magn. Mater.* **192** 485

[11] Joonghoe Dho, Kim W S and Hur N H 2002 *Phys. Rev. Lett.* **89** 027202

[12] Nimori S and Tsai A P 2002 *J. Magn. Magn. Matter.* **241** 11

[13] Arrott A 1957 *Phys. Rev.* **108** 1394

[14] Gunnarsson K, Svedlindh P, Nordblad P and Lundgren L 1988 *Phys. Rev. Lett.* **61** 754

[15] Mydosh J A 1993 *Spin glasses: an Experimental Introduction* (London·Washington, DC: Taylor & Francis) p72

[16] Jonason K, Mattsson J and Nordblad P 1996 *Phys. Rev.* B **53** 6507

[17] Mydosh J A 1993 *Spin glasses: an Experimental Introduction* (London·Washington, DC: Taylor & Francis) p68