IOPscience iopscience.iop.org

Home Search Collections Journals About Contact us My IOPscience

Magnetic properties and structure of the intermetallic compounds $La_2(Co_{1-x}Fe_x)_{16}Ti$

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1998 J. Phys.: Condens. Matter 10 179

(http://iopscience.iop.org/0953-8984/10/1/020)

View the table of contents for this issue, or go to the journal homepage for more

Download details:
IP Address: 171.66.16.151
The article was downloaded on 12/05/2010 at 23:17

Please note that terms and conditions apply.

J. Phys.: Condens. Matter **10** (1998) 179–184. Printed in the UK PII: S0953-8984(98)88064-8

Magnetic properties and structure of the intermetallic compounds $La_2(Co_{1-x}Fe_x)_{16}Ti$

Q L Liu†, J K Liang†‡, G H Rao†, J Xu†, X L Chen†, G Y Huo† and B G Shen§

† Institute of Physics and Centre for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

‡ International Centre for Materials Physics, Academia Sinica, Shenyang 110015, People's Republic of China

§ State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

Received 1 October 1997

Abstract. The effect of substitution of Fe for Co in the compounds $La_2(Co_{1-x}Fe_x)_{16}Ti$ ($x = 0.0$–0.5) on the crystal structure and magnetic properties has been investigated. The lattice parameters and the unit-cell volume of $La_2(Co_{1-x}Fe_x)_{16}Ti$ increase with x in the range $0 \leqslant x \leqslant 0.44$. The magnetic moment per formula unit in $La_2(Co_{1-x}Fe_x)_{16}Ti$ increases linearly as the Fe concentration increases. The Curie temperature is found to increase for $x \leqslant 0.1$ and then decrease for $x > 0.1$. The easy-magnetization direction changes from the easy-axis direction ($0 \leqslant x \leqslant 0.3$) to the easy-plane direction ($x = 0.4$), and the anisotropy field H_A decreases with the Fe substitution. Evidence for spin reorientation in the compounds $La_2(Co_{1-x}Fe_x)_{16}Ti$ for $0.3 \leqslant x \leqslant 0.44$ was observed in the M–T and χ_{ac}–T curves.

1. Introduction

Over the past thirty years, R_2Co_{17} (R = rare-earth element) compounds have been studied extensively. Although they have high saturation magnetizations and Curie temperatures, it is hard to find practical applications of the R_2Co_{17} series of compounds on their own due to their unfavourable anisotropy. The cobalt sublattice anisotropy in these compounds is proved to be easy-plane anisotropy [1]. The presence of 'dumb-bell' pairs of Co atoms in the compounds has an unfavourable influence on the magnetocrystalline anisotropy [2]. The overall anisotropy of R_2Co_{17} compounds results from a competition between the anisotropy of the R sublattice and the Co sublattice. Consequently, the R_2Co_{17} compounds are either weak uniaxial (e.g. Sm_2Co_{17}) or planar (e.g. Pr_2Co_{17}, Nd_2Co_{17}). However, it was found that for the R_2Co_{17} compounds the anisotropy can be increased or a planar anisotropy can be converted to a uniaxial one by replacing Co with an appropriate proportion of other 3d or 4d transition metals or Al [3–6]. Within the R_2Co_{17} series of compounds, La_2Co_{17} does not exist. So, little investigation of the isostructural 2:17 compound of La has been performed. In our previous paper [7], the new compounds $La_2Co_{17-x}Ti_x$, the isostructural 2:17 compounds, were reported on. These compounds exhibit favourable uniaxial anisotropy and so attracted our interest. In the present study, the influence of the substitution of Fe for Co in the compound $La_2Co_{16}Ti$ on the magnetic properties and crystal structure is investigated.

0953-8984/98/010179+06$19.50 © 1998 IOP Publishing Ltd

2. Experiment

$La_2(Co_{1-x}Fe_x)_{16}Ti$ ($x = 0$, 0.06, 0.1, 0.2, 0.3, 0.4, 0.44, 0.5) samples were prepared by argon arc melting of an appropriate amount of the starting metallic materials. The purities of the starting materials are better than 99.9%, To ensure the homogeneity of the samples, the ingots were turned and melted several times. The weight loss of the samples during melting was less than 1%. After arc melting, the samples were annealed in vacuum at 1273 K for two weeks. Phase identification of the samples was carried out by x-ray powder diffraction, using a four-layer monochromatic focusing Guinier–de Wolff camera and Co Kα radiation. The x-ray diffraction (XRD) data used to determine the lattice parameters were collected by a Rigaku Rint-2400 diffractometer with Cu Kα radiation and a graphite monochromator.

The magnetization (M–H) curves were measured at 1.5 K by use of an extraction sample magnetometer in a magnetic field ranging from 0 to 65 kOe. The saturation magnetization was derived using the law of approach to saturation. The temperature dependence of the magnetization of $La_2(Co_{1-x}Fe_x)_{16}Ti$ (M–T curves) in the temperature range 300–1123 K was measured by using a Faraday balance and a vibrating-sample magnetometer, and the Curie temperatures were determined from the M–T curves. The ac susceptibility (χ_{ac}–T curves) was measured in the temperature range from 77 K to 300 K with a frequency of 320 Hz. The spin-reorientation temperature was derived from the M–T and χ_{ac}–T curves. The easy-magnetization direction (EMD) was identified from the x-ray diffraction patterns of the magnetically aligned samples. The aligning was done by mixing fine particles of a sample with epoxy resin and allowing them to harden in a magnetic field of 1 T.

Table 1. The lattice parameters and magnetic data for $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds. T_C is the Curie temperature (in K), μ_s is the magnetic moment per formula unit (in μ_B/f.u.), H_A is the anisotropy field (in kOe).

x	a (Å)	c (Å)	V (Å^3)	T_C	μ_s	H_A
0.0	8.525	12.362	778.05	914	22.3	23
0.06	8.534	12.388	781.30	918	23.0	—
0.1	8.542	12.407	784.00	921	23.7	17
0.2	8.555	12.441	788.54	915	25.1	13
0.3	8.573	12.479	794.28	905	26.9	10
0.4	8.585	12.510	798.49	886	28.7	12
0.44	8.591	12.524	800.50	863	28.9	—
0.5	8.592	12.528	800.94	—	—	—

3. Results and discussion

For annealed $La_2(Co_{1-x}Fe_x)_{16}Ti$ ($x = 0$, 0.06, 0.1, 0.2, 0.3, 0.4, 0.44, 0.5) samples, x-ray diffraction patterns revealed that single-phase samples were obtained in the composition range $0 \leqslant x \leqslant 0.44$. The x-ray diffraction pattern was successfully indexed with a rhombohedral Th_2Zn_{17}-type structure using the program TREOR [8] and the lattice parameters were further least-squares refined. Figure 1 shows the dependence of the lattice parameters a and c as well as the unit-cell volume V of $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds on the concentration of Fe (see also table 1). A linear increase of a, c and V with x in the range $0 \leqslant x \leqslant 0.44$ is observed, which implies that the homogeneous range for the

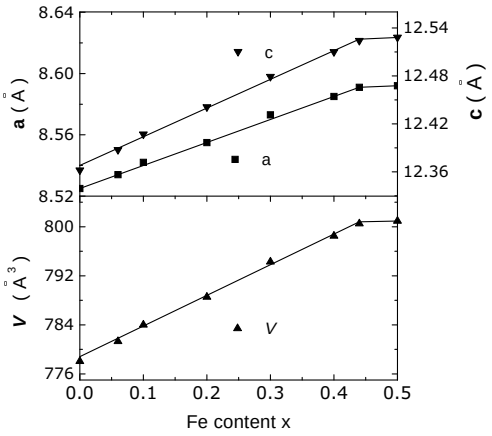

Figure 1. The dependence of the lattice parameters a, c, and the unit-cell volume V of $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds on the Fe concentration.

$La_2(Co_{1-x}Fe_x)_{16}Ti$ series is $0 \leqslant x \leqslant 0.44$. The increase of the lattice parameters and unit-cell volume with the Fe content in $La_2(Co_{1-x}Fe_x)_{16}Ti$ is consistent with the fact that the atomic radius of Fe is larger than that of Co. On the other hand, the enthalpy of alloying between La and Fe is positive, while that between La and Co is negative [9]; therefore, the substitution of Fe for Co in $La_2(Co_{1-x}Fe_x)_{16}Ti$ can also give rise to an increase of the unit-cell volume.

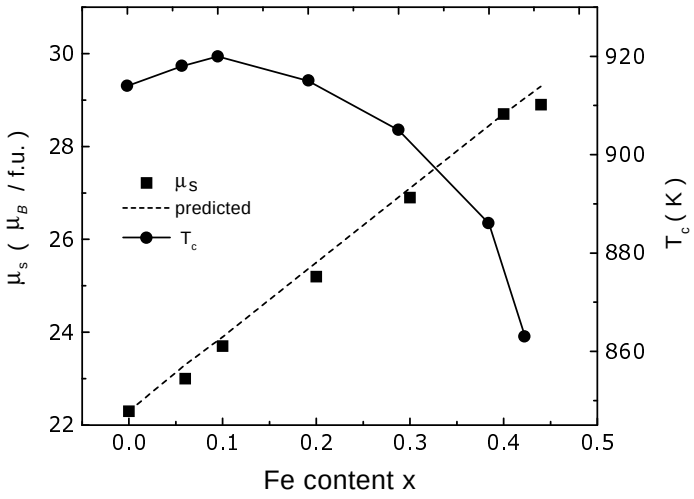

Figure 2. The dependence of the saturation magnetization μ_s and the Curie temperature T_C of $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds on the Fe concentration. The solid line is a guide to the eyes. The dashed line is the theoretical prediction based on the rigid-band model: $\mu_s = \mu_0 + 16x$.

The magnetization curves of the $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds were measured by an extraction sample magnetometer at 1.5 K in a magnetic field ranging from 0 to 65 kOe. The saturation moment was deduced from the M–H curves on the basis of the law of approach to saturation, and converted to the magnetic moment per formula unit; the results are listed

in table 1. The concentration dependence of the magnetic moment per formula unit is shown in figure 2. μ_s increases linearly with increasing x, the Fe content. This tendency is similar to that of $Y_2(Co_{1-x}Fe_x)_{17}$, and can be explained using the concept of rigid-band filling and emptying [10, 11]. A theoretical line, $\mu_s = \mu_0 + 16x$ $(0 \leqslant x \leqslant 0.44)$, based on the rigid-band model, are also shown in figure 2, where μ_0 is the saturation moment of $La_2Co_{16}Ti$ (in μ_B/f.u.). It can be seen that the experimental data coincide well with the theoretical prediction and imply that the magnetic moment per formula unit is increased by 1 μ_B upon substitution of an Fe atom for a Co atom.

The temperature dependence of the magnetization was measured in a low field, and the Curie temperature was determined from the M–T curves; the results are listed in table 1. The concentration dependence of the Curie temperature of the $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds is shown in figure 2. The Curie temperature first increases with the Fe concentration for $x \leqslant 0.1$, and then decreases for $x > 1$. This variation is similar to that occurring in $LaCo_{13-x-y}Fe_xSi_y$ compounds [12]. Within the mean-field approximation, the Curie temperature is proportional to the square of the magnetic moment and the exchange integral J. The exchange integral J decreases with increase of the unit-cell volume in Co-based 2:17 compounds [6]. The substitution of Fe for Co increases both the unit-cell volume and the magnetic moment. The subtle combination of the changes of the magnetic moment and J can give rise to a complex dependence of the Curie temperature on the Fe content in these compounds.

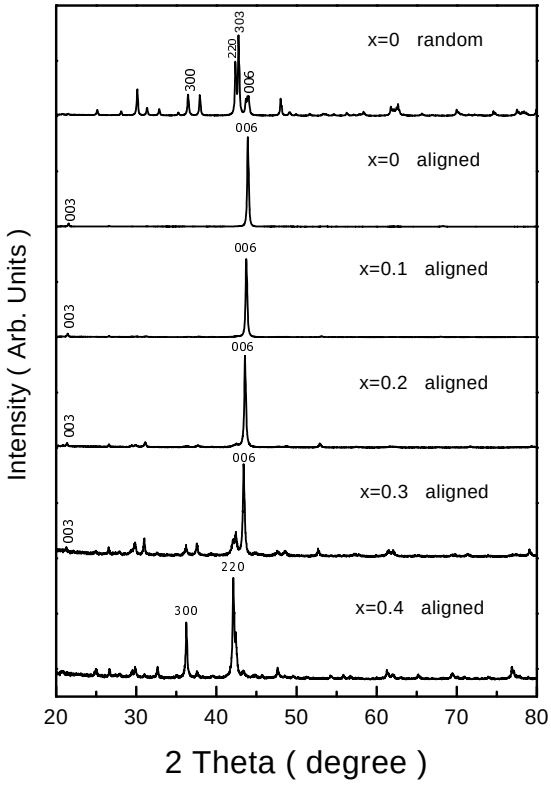

Figure 3. The XRD patterns of the random and magnetically aligned $La_2(Co_{1-x}Fe_x)_{16}Ti$ powders.

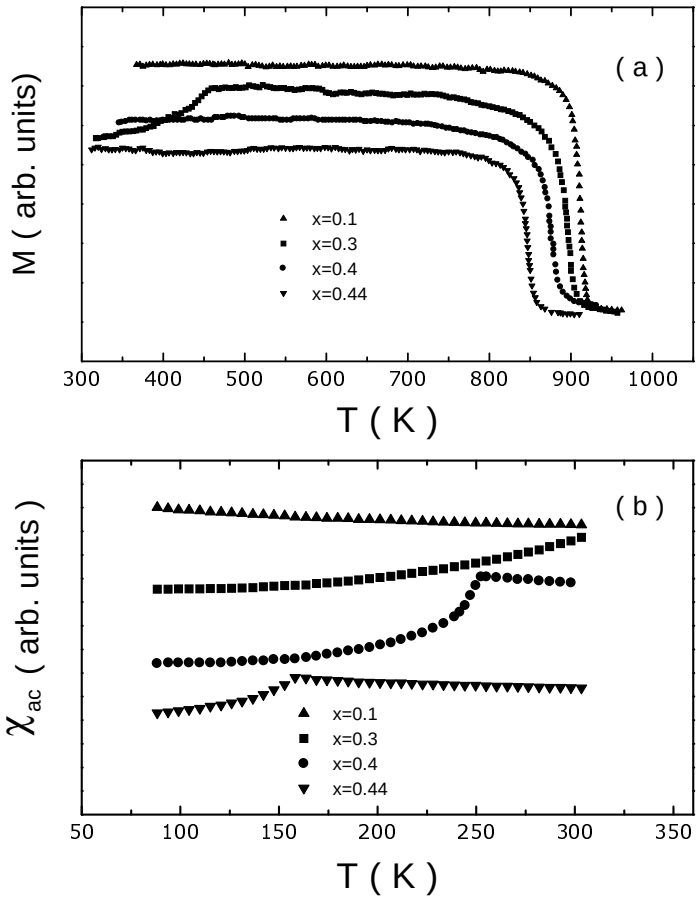

Figure 4. The temperature dependence of the magnetization (a) and the ac susceptibility (b) of $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds.

For the compound $La_2Co_{16}Ti$, the easy-magnetization direction (EMD) at room temperature is parallel to the *c*-axis [7]. In this paper, we present XRD patterns for magnetically oriented polycrystalline powders; these were obtained to investigate the change of the EMD with increasing Fe concentration. The XRD patterns indicate that the EMD changes from the *c*-axis to the basal plane with the increase of the Fe content (figure 3). The compounds retain easy-axis anisotropy for $x \leqslant 0.3$ and their anisotropy changes to an easy-basal-plane anisotropy for $x \geqslant 0.4$. The magnetization curves were measured in applied fields parallel or perpendicular to the EMD of the aligned powder sample by an extraction sample magnetometer at 1.5 K. By linearly extrapolating ΔM to zero on the ΔM–H curve ($\Delta M = M_{\parallel} - M_{\perp}$), the anisotropy fields H_A of the $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds were derived; these are listed in table 1. It can be seen that H_A for the $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds decreases with increasing Fe substitution ($x \leqslant 0.3$).

In general, the overall magnetocrystalline anisotropy of a rare-earth–transition metal intermetallic compound is the sum of the 4f-sublattice and 3d-sublattice anisotropies. For $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds, however, La atoms have no orbital magnetic moment and, consequently, make no contribution to the magnetocrystalline anisotropy. Thus, the total

anisotropy arises exclusively from the 3d sublattice. Because the relevant second-order Stevens factors of Fe and Co ions are of opposite sign, the contributions to the anisotropy energies of Fe and Co ions at all of the different crystallographic positions have opposite signs for most isostructural compounds [13]. Although both the Co sublattice anisotropy in Co-based 2:17-type compounds (e.g. Y_2Co_{17}) and the Fe sublattice anisotropy in Fe-based 2:17-type compounds (e.g. Y_2Fe_{17}) are planar, the anisotropy constant K_1 for the former is much smaller than that for the latter [14]. Thus, H_A for the $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds decreases with increasing Fe substitution ($x \leqslant 0.3$) and the EMD changes from the easy-axis direction to the easy-basal-plane direction for higher contents (e.g. $x = 0.4$).

Figure 4(a) shows the M–T curves of $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds. An anomaly at around 450 K for $x = 0.3$ is observable in figure 4(a). This anomaly could be due to a spin-reorientation transition, like that in $R_2Co_{17-x}Ti_x$ (R = Pr or Nd) [5]. Figure 4(b) shows the temperature dependence of the ac susceptibility measured for bulk samples between 77 K and 300 K. The χ_{ac}–T curves for $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds exhibit anomalies at around 250 K for $x = 0.4$ and at around 160 K for $x = 0.44$, also indicating a spin-reorientation transition in the compounds. Taking into consideration the fact that the magnetic anisotropy is uniaxial for $La_2(Co_{1-x}Fe_x)_{16}Ti$ with $x \leqslant 0.3$ and planar with $x = 0.4$ according to XRD experimental results obtained at room temperature, the anomaly in the M–T and χ_{ac}–T curves is indicative of a change of the magnetic anisotropy from uniaxial to planar anisotropy for $La_2(Co_{1-x}Fe_x)_{16}Ti$ for $x = 0.3$ at around 450 K, for $x = 0.4$ at around 250 K and for $x = 0.44$ at around 160 K as the temperature increases. These observations imply that the uniaxial anisotropy of $La_2(Co_{1-x}Fe_x)_{16}Ti$ compounds is weakened by the substitution of Fe for Co.

Acknowledgment

This work was supported by the National Natural Science Foundation of China.

References

[1] Buschow K H J 1991 *Rep. Prog. Phys.* **54** 1123
[2] Deportes J, Givord D, Lemaire R, Nagai H and Yang Y T 1976 *J. Less-Common Met.* **44** 273
[3] Schaller H J, Craig R S and Wallace W E 1972 *J. Appl. Phys.* **43** 3161
[4] Satyanarayana M V, Fujii H and Wallace W E 1984 *J. Magn. Magn. Mater.* **40** 241
[5] Tang N, Shen B G, Buschow K H J, de Boer F R, Wang Y Z and Qiao G W 1997 *J. Appl. Phys.* **81** 5127
[6] Chen Zhao-hua, Shen Bao-gen, Zhang Jun-xian, Liang Bing, Guo Hui-qun and Kronmuller H 1997 *Appl. Phys. Lett.* **70** 3467
[7] Liu Quanlin, Liang Jingkui, Rao Guanghui, Tang Weihua, Sun Jirong, Chen Xiaolong and Shen Baogen 1997 *Appl. Phys. Lett.* **71** 1869
[8] Werner P E 1964 *Z. Kristallogr.* **120** 375
Werner P E 1976 *J. Appl. Crystallogr.* **9** 216
[9] Rao G H, Wu S, Yan X H, Zhang Y L, Tang W H and Liang J K 1993 *J. Alloys Compounds* **202** 101
[10] Perkins R S and Nagel H 1975 *Physica* B **80** 143
[11] Perkins R S and Strassler S 1977 *Phys. Rev.* B **15** 490
[12] Rao G H, Liang J K, Zhang Y L, Tang W H and Cheng X R 1996 *J. Appl. Phys.* **80** 336
[13] Thuy N P, Franse J J M, Hong N M and Hien T D 1988 *J. Physique Coll.* **49** C8 499
[14] Thuy N P and Franse J J M 1986 *J. Magn. Magn. Mater.* **54–57** 915