2012

Anisotropic and excellent magnetocaloric properties of La 0.7Ca0.3MnO3 single crystal with anomalous magnetization

J.C Debnath
ISEM, University of Wollongong

Rong Zeng
University of Wollongong, rzeng@uow.edu.au

Jung Ho Kim
University of Wollongong, jhk@uow.edu.au

Dapeng Chen
University of Wollongong, dapeng@uow.edu.au

S X. Dou
University of Wollongong, shi@uow.edu.au

Anisotropic and excellent magnetocaloric properties of La 0.7Ca0.3MnO3 single crystal with anomalous magnetization

Abstract

Magnetic properties and the magnetocaloric effect (MCE) have been investigated in La0.7Ca0.3MnO3 single crystal with applied field along both the ab-plane and the c-direction. Due to the magnetocrystalline anisotropy, the crystal exhibits anisotropy in the MCE. Upon application of a 5 T field, the magnetic entropy changes (SM), reaching values of 7.668 J/(kg K) and 6.412 J/(kg K) for both the ab-plane and the c-direction, respectively. A magnetic entropy change of 3.3 J/(kg K) was achieved for a magnetic field change of 1.5 T at the Curie temperature, TC = 245 K. Due to the absence of grains in the single crystal, the SM distribution here is much more uniform than for gadolinium (Gd) and other polycrystalline manganites, which is desirable for an Ericsson-cycle magnetic refrigerator. For a field change of 5 T, the relative cooling power, RCP, reached 358.17 J/kg, while the maximum adiabatic temperature change of 5.33 K and a magnetoresistance (MR) ratio of 507.88% at TC were observed. We analysed the magnetization of La0.7Ca0.3MnO3 single crystal at TC and estimated several parameters of spin fluctuation on the basis of a self-consistent renormalization theory of spin fluctuations, with reciprocal susceptibility above TC. We found that the magnetic property of La0.7Ca0.3MnO3 is weakly itinerant ferromagnetic. A large reversible MCE and no hysteresis loss with a considerable value of refrigerant capacity indicate that La0.7Ca0.3MnO3 single crystal is a potential candidate as a magnetic refrigerant.

Keywords

7ca0, magnetization, crystal, la, single, properties, magnetocaloric, excellent, anisotropic, 3mno3, anomalous

Disciplines

Engineering | Physical Sciences and Mathematics

Publication Details

Debnath, J, Zeng, R, Kim, J, Chen, D & Dou, SX (2012), Anisotropic and excellent magnetocaloric properties of La 0.7Ca0.3MnO3 single crystal with anomalous magnetization, Materials Science and Engineering B: Advanced Functional Solid-state Materials, 177(1), pp 48-53.

Anisotropic and excellent magnetocaloric properties of $La_{0.7}Ca_{0.3}MnO_3$ single crystal with anomalous magnetization

J. C. Debnath*, R. Zeng, J. H. Kim, D.Chen, S. X. Dou

Institute for Superconducting and Electronic Materials, University of Wollongong, Squaire Way, North Wollongong, Wollongong, NSW 2500, Australia

* Corresponding author, E-mail: jcd341@uowmail.edu.au, jcdebnath@yahoo.com
Fax: +61 2 4221 5731

Abstract

Magnetic properties and the magnetocaloric effect (MCE) have been investigated in $La_{0.7}Ca_{0.3}MnO_3$ single crystal with applied field along both the *ab*-plane and the *c*-direction. Due to the magnetocrystalline anisotropy, the crystal exhibits anisotropy in the MCE. Upon application of a 5 T field, the magnetic entropy changes (ΔS_M), reaching values of 7.668 J/kg K and 6.412 J/kgK for both the *ab*-plane and the *c*-direction, respectively. A magnetic entropy change of 3.3 J/kgK was achieved for a magnetic field change of 1.5 T at the Curie temperature, T_C = 245 K. Due to the absence of grains in the single crystal, the ΔS_M distribution here is much more uniform than for gadolinium (Gd) and other polycrystalline manganites, which is desirable for an Ericsson-cycle magnetic refrigerator. For a field change of 5 T, the relative cooling power, RCP, reached 358.17 J/kg, while the maximum adiabatic temperature change of 5.33 K and a magnetoresistance (MR) ratio of 507.88% at T_C were observed. We analysed the magnetization of $La_{0.7}Ca_{0.3}MnO_3$ single crystal at T_C and estimated several parameters of spin fluctuation on the basis of a self-consistent renormalization theory of spin fluctuations, with reciprocal susceptibility above T_C. We found that the magnetic property of $La_{0.7}Ca_{0.3}MnO_3$ is weakly itinerant ferromagnetic. A large reversible MCE and no hysteresis loss with a considerable value of refrigerant capacity indicate that $La_{0.7}Ca_{0.3}MnO_3$ single crystal is a potential candidate as a magnetic refrigerant.

Keywords: spin fluctuation; Hysteresis; Anisotropy; Magnetocaloric effect; Weakly itinerant ferromagnetic; Reciprocal susceptibility;

1. Introduction

The magnetocaloric effect (MCE) is a basic characteristic of magnetic materials and is understood as an isothermal magnetic entropy change or an adiabatic temperature change of a magnetic material upon the application of a magnetic field. It has been shown that the heating and cooling in the magnetic refrigeration process are proportional to the size of the magnetic moments and to the applied magnetic field. That is why research on magnetic refrigeration has been exclusively conducted on heavy rare-earth elements and their compounds [1, 2]. Among the rare-earth metals, gadolinium was found to show the highest MCE [2]. Nonetheless, the cost of gadolinium as a magnetic refrigerant is quite expensive (~$4000/kg), and this actually limits the usage of it as an active magnetic refrigerant (AMR) in magnetic refrigerators. Since the discovery of the giant MCE phenomenon, a giant MCE has been found in a large variety of magnetic materials, including $Gd_5(Si_{1-x}Ge_x)_4$, $MnAs_{1-x}Sb_x$, $MnFe(P_{1-x}As_x)$, $La(Fe_{13-x}Si_x)$, and RM_2 (where R = rare earth, M = Al, Co, Ni) [3-8]. Further efforts to discover new materials, especially materials without rare-earth elements and exhibiting large MCE in response to low applied field, are of significant importance. Among them, perovskite-type manganese oxide materials [9-13] having large MCEs are believed to be good candidates for magnetic refrigeration at various temperatures.

In view of the AMR requirements, note that besides the requirement that an AMR material should have a large magnetic entropy change (ΔS_M) induced by a low magnetic field change, the ΔS_M distribution also plays an important role in achieving magnetic cooling efficiency [14, 5]. Unfortunately, a non-uniform ΔS_M distribution, which is not beneficial for an Ericsson-cycle magnetic refrigerator, has been found for several AMR materials, such as gadolinium [1] and polycrystalline perovskite manganites, due to structural inhomogeneity [11, 12].

There are two key requirements for a magnetic material to possess a large MCE. One is a large enough spontaneous magnetization, which is characteristic of a class of heavy rare-earth metals. The second is that it exhibit sharp drop in magnetization, which is associated with the ferromagnetic-paramagnetic transition at the Curie temperature, as was found in perovskite manganites. The magnetic properties of manganites, the Curie temperature, and the saturation magnetization are strongly doping-dependent. So, these materials may be good candidates for MR at various temperatures. Manganite magnetocaloric materials can be promising

candidates for active magnetic regenerative (AMR) refrigeration, because they show large MCEs that are comparable to those of Gd and other magnetic refrigerant candidate materials. It is interesting to note that, when compared with Gd and other candidate materials, $La_{0.7}Ca_{0.3}MnO_3$ single crystal perovskite manganite is more convenient to prepare and exhibit higher chemical stability, as well as the higher resistivity that is favorable for lowering eddy current heating. In addition, the manganites possess much smaller thermal and field hysteresis than any rare earth and 3d-transition metal based alloy. In addition, this material is the cheapest among the existing magnetic refrigerants. These superior features may make it more promising for future MR technology.

In this context, study of the MCE in lanthanum manganite single crystals can be of great interest, because the absence of grains in these materials would be expected to allow a uniform ΔS_M distribution, which is desirable for an Ericsson-cycle magnetic refrigerator. Here, we report on the excellent magnetocaloric properties of $La_{0.7}Ca_{0.3}MnO_3$ single crystal, with which the requirements for an AMR material can be fulfilled.

Moriya and his co-workers self-consistently fed back the effects of spin fluctuations to the magnetic susceptibility, and succeeded in theoretically reproducing the Curie-Weiss behaviour of ferromagnetic materials originated in the linear growing of spin fluctuations, e.g., square of local spin density $S^2_{L,}$ with T. This self-consistent renormalization (SCR) theory of spin fluctuations succeeded in clarifying the nature of nearly and weakly itinerant ferromagnetic metals. Furthermore, Takahashi, one of Moriya's co-workers, developed the SCR theory by assuming that the sum of zero-point and thermal spin fluctuations is conserved against temperature [15-18]. We estimated the spin fluctuation parameters by adopting Takahashi's developed spin-fluctuation theory and calculated $1/\chi$. We show that the magnetic property of $La_{0.7}Ca_{0.3}MnO_3$ is weakly itinerant ferromagnetic (WIF).

2. Experimental Details

A high-quality single crystal of $La_{0.7}Ca_{0.3}MnO_3$ was prepared by the floating zone method, using an infrared radiation convergence-type image furnace that consists of four mirrors and a halogen lamp. The starting ceramic rods were synthesized by the standard methods of solid-state reaction of a stoichiometric mixture of La_2O_3, $CaCO_3$, and MnO_2. X-ray diffraction data and electron-probe microanalysis confirmed the quality of the crystal. The magnetic measurements were performed using a physical properties measurement system 14 T (PPMS-14T) magnetometer.

3. Results and Discussion

The phase composition and crystal structure of the sample was characterized by X-ray diffraction (XRD), and high purity and perovskite structure were confirmed. The XRD pattern of the sample is shown in Fig. 1. A single-phase diffraction pattern was obtained, yielding an orthorhombic structure with the P n m a space group.

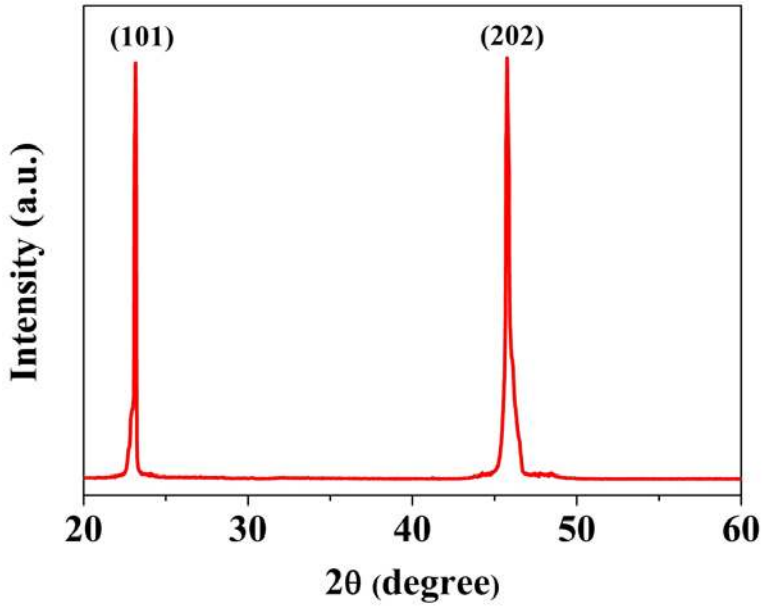

Fig. 1. XRD pattern of the $La_{0.7}Ca_{0.3}MnO_3$ single crystal.

Fig. 2 shows the temperature dependence of the magnetization (M-T curves) in a 1.59×10^4 A/m field with the direction of the magnetic field along both the *ab*-plane and the *c*-direction of the $La_{0.7}Ca_{0.3}MnO_3$ single crystal. The Curie temperature (T_C), defined by the maximum in the absolute value of *dM/dT*, has been determined from the M-T curve to be about 245 K. The magnetization in the *ab*-plane is much bigger than that in the *c*-direction, suggesting that the magnetic easy axis lies in the *ab*-plane of the crystal. A steep change in the magnetization occurs at T_C, implying a first-order transition, leading to expectations that the material will show a large MCE near the Curie temperature.

In order to confirm this, the isothermal magnetization of the sample was measured in the range of 0-5 T for a range of temperatures around T_C. To ensure the readability of the figure, only some of isothermals are presented in Fig. 3. It can be seen from Fig. 3 that there is a drastic change in the magnetization around T_C, indicating a large magnetic entropy change.

In order to evaluate the MCE of the present material, we calculated the changes in the magnetic entropy ΔS_M caused by the application of external magnetic fields from the isothermal curves of magnetization versus applied field.

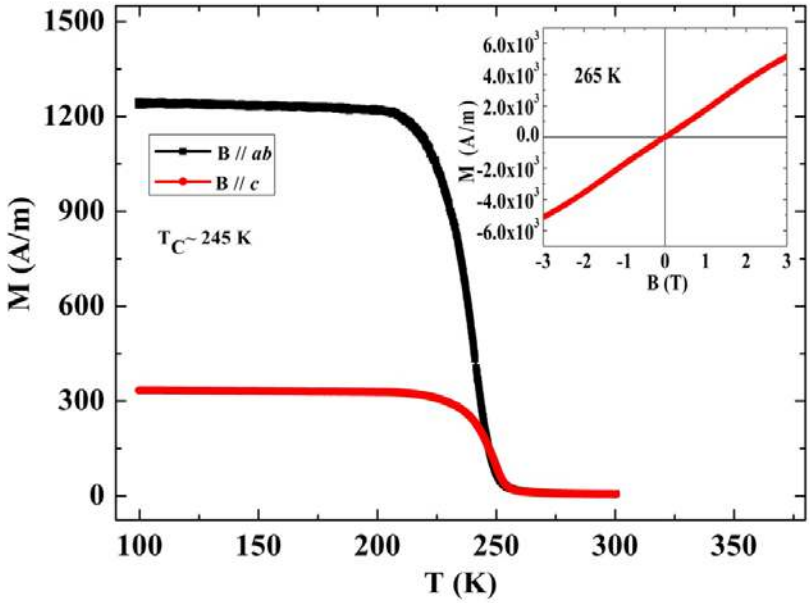

Fig. 2. Temperature dependence of magnetization along the *ab*-plane and the *c*-direction of $La_{0.7}Ca_{0.3}MnO_3$ single crystal. Inset: hysteresis curve of the sample at 265 K.

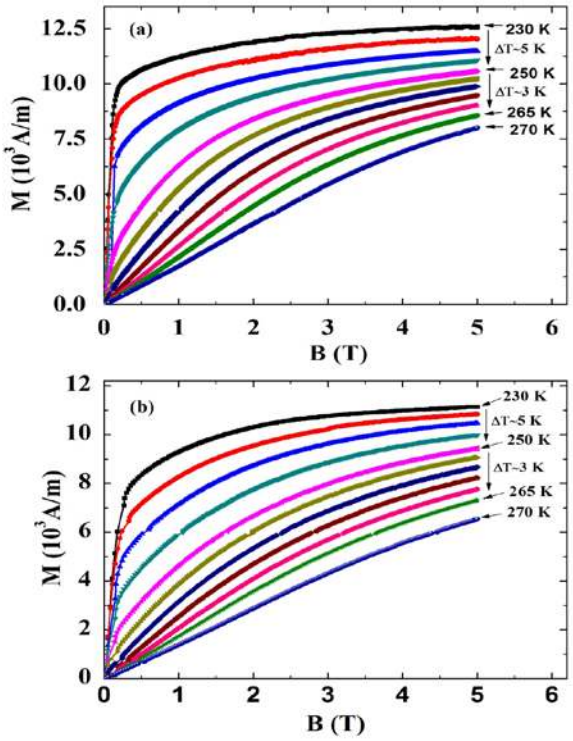

Fig. 3. Magnetic field dependence of magnetization measured at various temperatures around T_C for $La_{0.7}Ca_{0.3}MnO_3$ single crystal along (a) the *ab*-plane and (b) the *c*-direction.

Fig. 4 shows the magnetic entropy change as a function of temperature in different magnetic fields ranging from 0 to 5 T for (a) the *ab*-plane and (b) the *c*-direction. The maximum value of ΔS_M, corresponding to external field changes of 1.5 T and 5 T for both the *ab*-plane and the *c*-direction reaches about 3.33 and 2.364 J/kg K, and 7.668 and 6.412 J/kg K, respectively. These values are higher than the result found by Phan et. al. [19] and Tian et. al. [20] for the same single crystal $La_{0.7}Ca_{0.3}MnO_3$. We have compared the peak values of ΔS_M between the *ab*-plane and *c*-direction as a function of magnetic field. In the whole field range from 0 to 5 T, ΔS_M in the *ab*-plane is larger than along the *c*-direction. The maximum differences at 1.5 T and 5 T are about 1 and 1.37 J/kg K, respectively. The large anisotropic magnetic entropy change in $La_{0.7}Ca_{0.3}MnO_3$ single crystal is ascribed to the strong magnetocrystalline anisotropy.

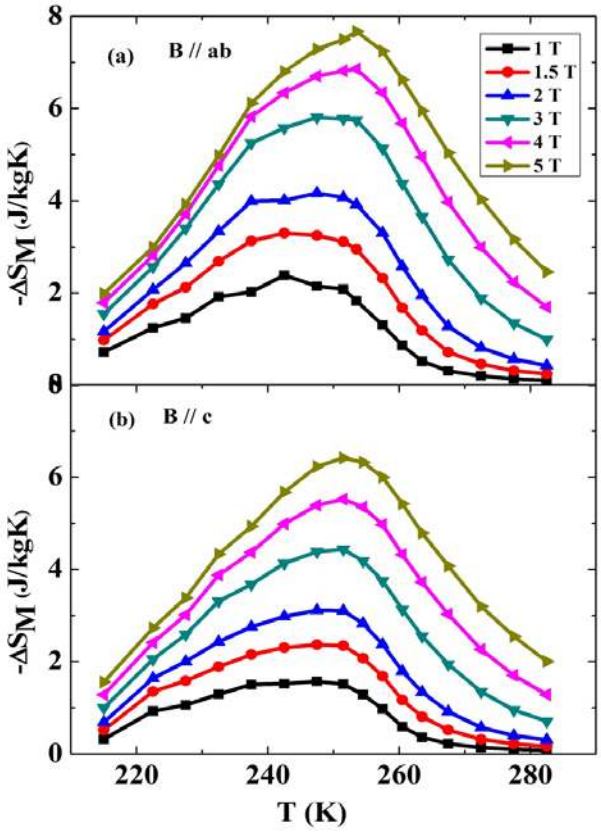

Fig. 4. Magnetic entropy change ΔS_M as a function of temperature for the magnetic field along (a) the *ab*-plane and (b) the *c*-direction of the $La_{0.7}Ca_{0.3}MnO_3$ single crystal.

Another interesting result is that the magnitude of the large low-field magnetic entropy change along the *ab*- plane of the single crystal is 3.3 J/kg K for a magnetic field change of 1.5 T. This value of ΔS_M is of the same magnitude as that of Gd [2] and also is larger than for many cubic perovskite manganites [21, 22]. This low-field MCE is beneficial for the practical

application of MCE because the low field can be directly supplied by $Nd_2Fe_{14}B$ or other permanent magnets. This low-field large magnetic entropy change in the $La_{0.7}Ca_{0.3}MnO_3$ single crystal is due to the rapid change in the magnetization near the Curie temperature in the easy magnetizing plane.

The symmetrical and uniform distribution of ΔS_M is observed for single crystal, even in high fields, and is much more uniform than for gadolinium and polycrystalline manganites [12, 23-26]. This is ascribed to the absence of grains in such a single-crystalline material, where there was a symmetric and uniform distribution of the temperature dependence of magnetoresistance [27, 28]. In contrast to the single crystalline manganite, considerable and asymmetrical variations in the ΔS_M curves with external magnetic field, particularly under high magnetic fields, for the polycrystalline manganites were observed and are likely due to the grain boundary effects [27-29]. The nonuniform distribution of ΔS_M in polycrystalline perovskite manganites is also believe to be attributable to a spread of the ferromagnetic clusters caused by the inhomogeneity of structure and stoichiometry [24, 25].

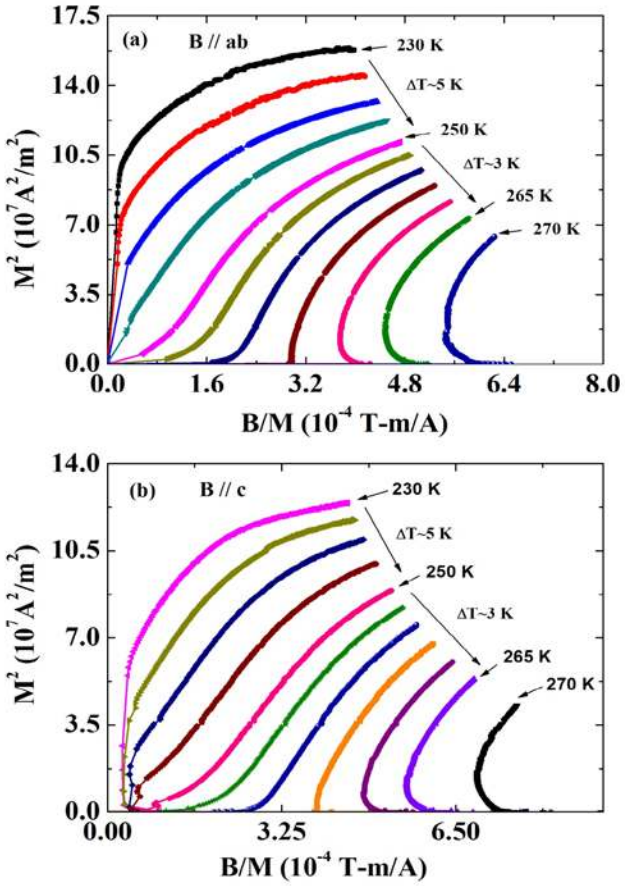

Fig. 5. Arrott plots of the $La_{0.7}Ca_{0.3}MnO_3$ single crystal with the direction of the magnetic field along (a) the *ab*-plane and (b) the *c*-direction.

A first-order magnetic transition is of particular importance for giant MCE. The first order nature of the phase transition of the single crystal can be confirmed by the Banerjee criterion [30]. According to the Banerjee criterion, a negative slope in a relatively small M^2 region above T_C indicates that the magnetic phase transition is first order, as is shown in Fig. 5. The curves have negative slopes or inflection points above T_C, also indicating a metamagnetic transition from a paramagnetic to a ferromagnetic state [31].

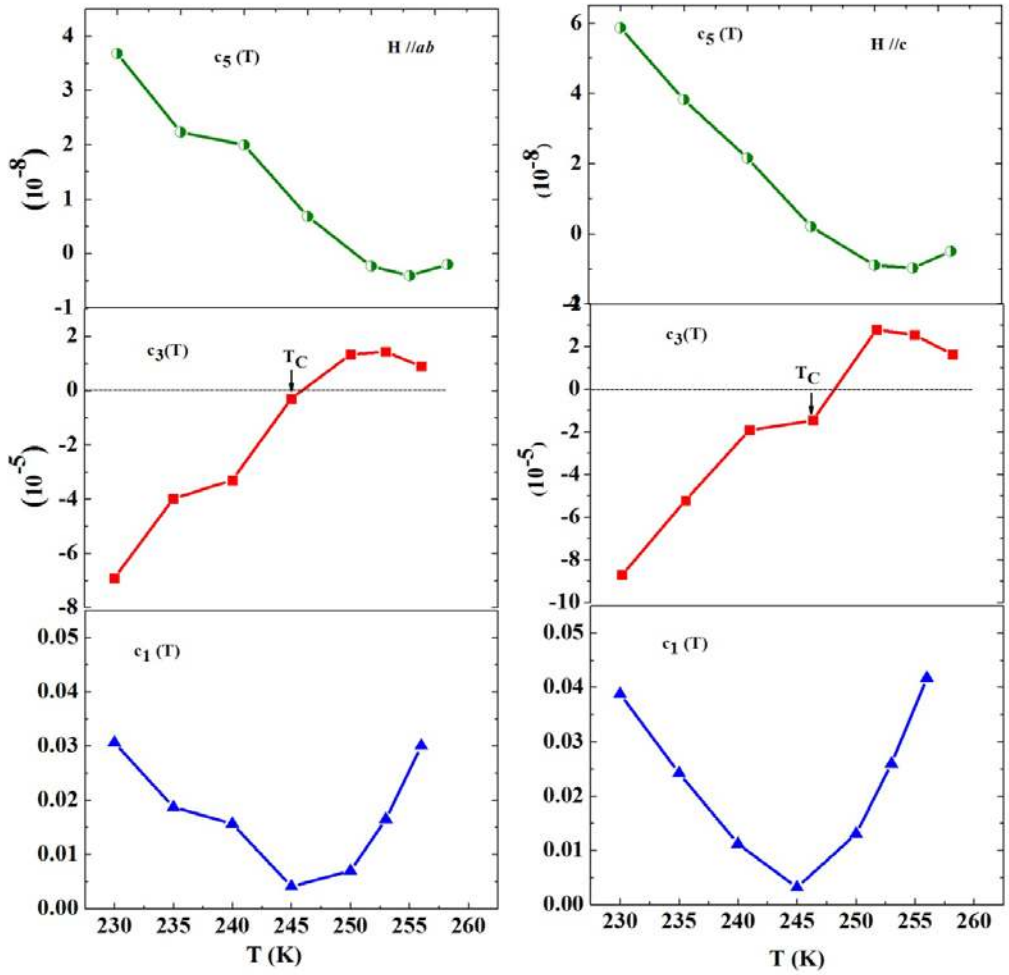

Fig. 6. Temperature dependence of Landau coefficients for $La_{0.7}Ca_{0.3}MnO_3$ single crystal along (left) the *ab*-plane and (right) the *c*-direction. The units of $c_1(T)$, $c_3(T)$, and $c_5(T)$ are T^2 m/A, T^4 m^3/A^3, and T^6 m^5/A^5, respectively.

To gain more insight into the magnetic transition, the Landau coefficients $c_1(T_C)$, $c_3(T_C)$, and $c_5(T_C)$ were determined by fitting the magnetic field against magnetization using the equation [32]

$$\mu_o H = c_1(T)M + c_3(T)M^3 + c_5(T)M^5 \qquad (1)$$

where H is the magnetic field strength and unit is A/m. This magnetic field strength can be expressed by the equation $B = \mu_o H$, where B is the magnetic field and μ_o is the magnetic permeability of the space. The details of the fitting method have been reported elsewhere [33]. The temperature dependence of the Landau coefficients derived from these fitting results is shown in Fig. 6. The value of $c_3(T_C)$ at T_C is negative for both field directions, again confirming that the magnetic phase transition is first-order.

In Fig. 5, M^2 does not show a linear relation, but does have a convex curvature against B/M for every temperature. The convex curvature of M^2 against B/M reminds us of the typical weakly itinerant ferromagnet (WIF) property of the magnetization which is found in MnSi [34]. To the best of our knowledge, the convex curvature of M^2 against B/M has been reproduced only by Takahashi's developed spin-fluctuation theory so far [18]. In this theory, it is assumed that the sum of the zero point and thermal spin fluctuations is conserved against T, and as a consequence of this assumption, he argued the importance of the sixth coefficient of the free energy when the system has a relatively large η [$= (T_C/T_0)^{1/3}$] value. Here, T_0 characterizes the energy width of the dynamical spin-fluctuation spectrum. In such a case, the magnetization obeys the following relation at T_C because the fourth coefficient becomes zero [35]:

$$H^* = [T_A/3(2+\sqrt{5})\, T_C]^2 p^5 \qquad (2)$$

where $H^* = B/2\mu_B$ and p are the magnetic field strength and the magnetization in μ_B unit respectively. The parameter T_A characterizes the dispersion of the static magnetic susceptibility in wave vector (q) space.

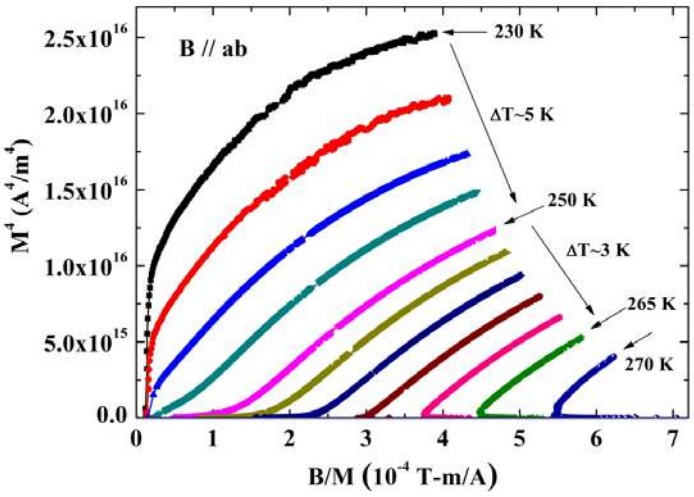

Fig. 7. M^4 versus B/M for $La_{0.7}Ca_{0.3}MnO_3$ single crystal along the *ab*-plane at various temperatures. Around T_C, M^4 is almost linear against B/M.

We plotted the isothermal magnetization curves in the form of M^4 vs. B/M, as shown in Fig. 7. Here, M^4 is almost linear against B/M around $T_C \approx 245$ K. Strictly speaking, M^4 is not completely linear with respect to B/M around T_C, as seen in Figs. 7 and 8(b). However, most itinerant ferromagnets show concave behaviour when plotted in the form of M^4 vs. B/M [36]. Furthermore, M^4 of the typical itinerant ferromagnetic compound MnSi is also not exactly linear with respect to B/M [34]. According to Takahashi's theory, in the case of the coefficient of M^4 in the Landau expansion of free energy being zero, M^4 becomes linear against B/M. Thus, complete linearity is only obtained in a delicate condition. $La_{0.7}Ca_{0.3}MnO_3$ is not just on this condition, but seems to be very near it, as well as having a quantum critical point due to spin fluctuations.

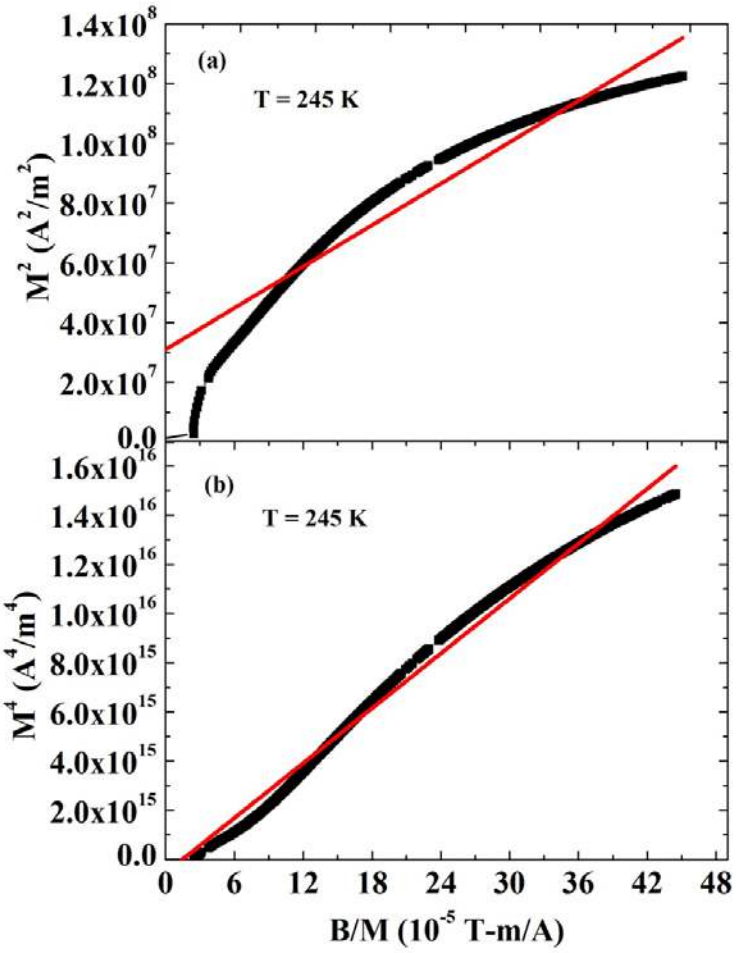

Fig. 8. Linear fitting for (a) M^2 versus B/M and (b) M^4 versus B/M for $La_{0.7}Ca_{0.3}MnO_3$ single crystal along the *ab*-plane at T = 245 K. Straight lines are the results of linear fitting to the data.

We show the results of linear fitting to M^2 and M^4 plotted against B/M at $T_C \approx 245$ K in Fig. 8. This figure clearly shows that at $T_C \approx 245$ K, the M^4 curve shows higher linearity against B/M than the M^2 curve against B/M. Equation (2) can be transformed to the relation of M^4 vs. B/M as [35]

$$M^4 = 1.17\times10^{19}(T^2_C/\ T^3_A)(B/M) \qquad (3)$$

where M and B are in units of A/m and T, respectively. By putting the value of the slope at $T_C \approx 245$ K into Eq. (3), we obtained T_A as 2.5×10^6 K.

According to the SCR theory for WIF materials [15-17], T_C is related to P_s, T_A, and T_0 by the following relation [35]:

$$T_C = (60c)^{-3/4}\, P_s^{3/2}\, T_A^{3/4}\, T_0^{1/4} \qquad (4)$$

where c and P_s are constants equals to 0.335 and 2.67. By putting the value of T_A determined by Takahashi's theory into Eq. (4), we obtained T_0 as 6×10^{-4} K.

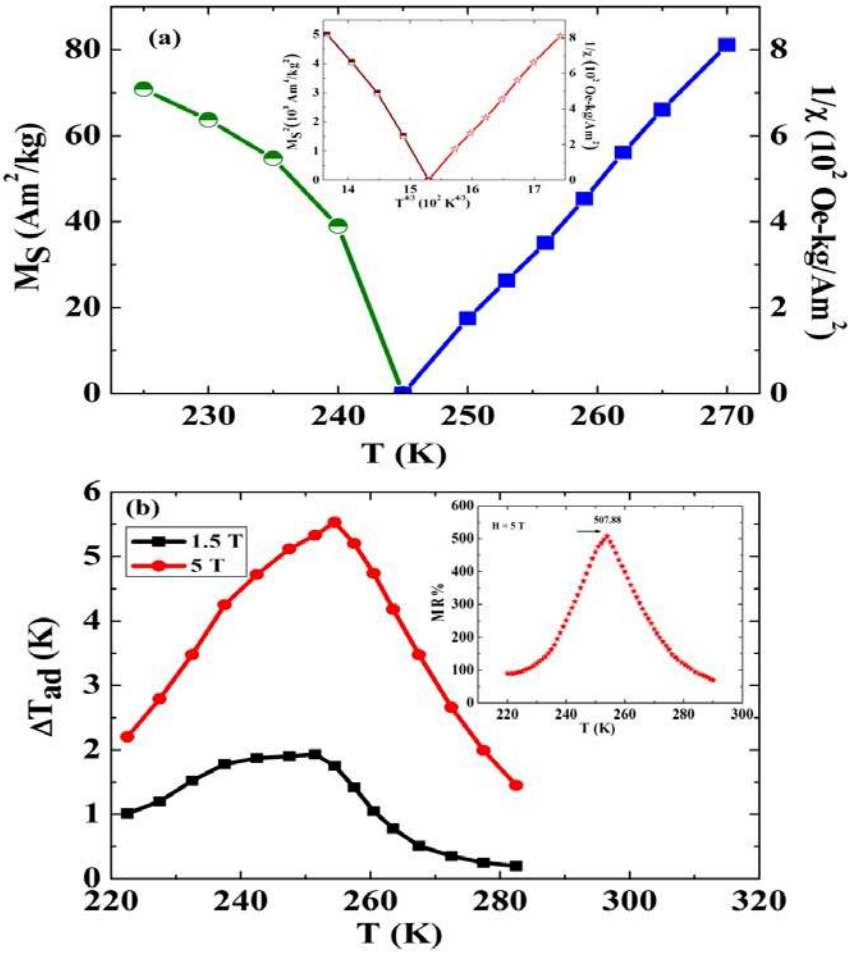

Fig. 9. $M_S(T)$ and $1/\chi(T)$ of $La_{0.7}Ca_{0.3}MnO_3$ single crystal along the *ab*-plane estimated from the Arrott plot (a). Inset: M_S^2 and $1/\chi$ plotted against $T^{4/3}$. Adiabatic temperature change ΔT_{ad}

as a function of temperature under field change of 1.5 and 5 T, respectively (b). Inset: Magnetoresistance plotted against temperature.

We estimated the saturation magnetization, $M_S(T)$, below T_C and the reciprocal magnetic susceptibility, $1/\chi$ (= $\lim_{H \to} B/M$) above T_C from Figs. 5(a) and 7, respectively. The M_S and $1/\chi$ are estimated as the values of the intersections of natural extrapolations of the M^2 (M^4) curves with the vertical and horizontal axes, respectively. The values estimated from the M^2 and the M^4 curves were almost the same, so we only show the values estimated from the M^2 curves. The obtained M_S and $1/\chi$ are shown in Fig. 9(a). We also show the M_S^2 and $1/\chi$ vs. $T^{4/3}$ plots in the inset of Fig. 9(a), since the M_S^2 and $1/\chi$ of WIFs are predicted to show non-Fermi-liquid behaviour and to obey a $T^{4/3}$ relation according to the SCR theory of spin fluctuations. M_S^2 and $1/\chi$ show linear relations against $T^{4/3}$ over a relatively wide temperature range, in good agreement with the SCR theory around T_C.

In order to further make sure of the great potential of the present $La_{0.7}Ca_{0.3}MnO_3$ single crystal along the *ab*-plane as a candidate for magnetic refrigerant, we have measured the adiabatic temperature change, ΔT_{ad}, by the equation $\Delta T_{ad}(T, \Delta B) = -\frac{T}{c_p(T,B)} \Delta S_M(T, \Delta B)$ for magnetic field change up to 5 T. Fig. 9(b) displays the temperature-dependence of ΔT_{ad}. The estimated ΔT_{ad} value is 5.53 K for a field change from 0 to 5 T. This value is about 70% of that of Gd and higher than for many manganese oxides [22]. Large magnetoresistance (MR) is the phenomenon where resistance of a material drops drastically on application of magnetic field. It has tremendous applications in recording information on hard disks, magnetic sensors, and spin-electronic devices [37, 38]. The MR ratio, defined as $MR = (\rho(0)/\rho(H) - 1)$, exhibits a maximum value of 507.88 % at T_C under a magnetic field of 5 T, as shown in the inset to Fig. 9(b).

The value of the refrigerant capacity (*RC*) of $La_{0.7}Ca_{0.3}MnO_3$ single crystal along the *ab*-plane is calculated by numerically integrating the area under the corresponding $\Delta S-T$ curve, with the temperature difference for the full width at half maximum (FWHM) of the peak, δT_{FWHM}, used for the integration limits, as shown in Fig. 10 [39]. The maximum value of *RC* is found to be 358.17 J/kg under a magnetic field change from 0 to 5 T. For the *RC* value, however, it is necessary to take into account the hysteresis loss. We have also noted a very small (almost negligible) hysteresis loop exhibited by the M (H) curve (inset to Fig. 2). Small magnetic losses are beneficial to the magnetic cooling efficiency. Large hysteretic losses reduce the magnetic refrigeration effect by about 23% in $Gd_5Ge_2Si_2$ [40, 41], in which the magnetic

loops exceed those of perovskite like magnetic refrigerants by a factor of about 40. The large magnetic loops in $Gd_5Ge_2Si_2$ are caused by a first order structural phase transition, induced by magnetic field. The structural transition shows large hysteresis with field, which, in turn, causes the high magnetic loop and large hysteretic losses. Magnetization-demagnetization processes in $La_{0.7}Ca_{0.3}MnO_3$ single crystal lanthanum manganites are not accompanied by any structural transition, and because of this, the hysteresis loops are very small.

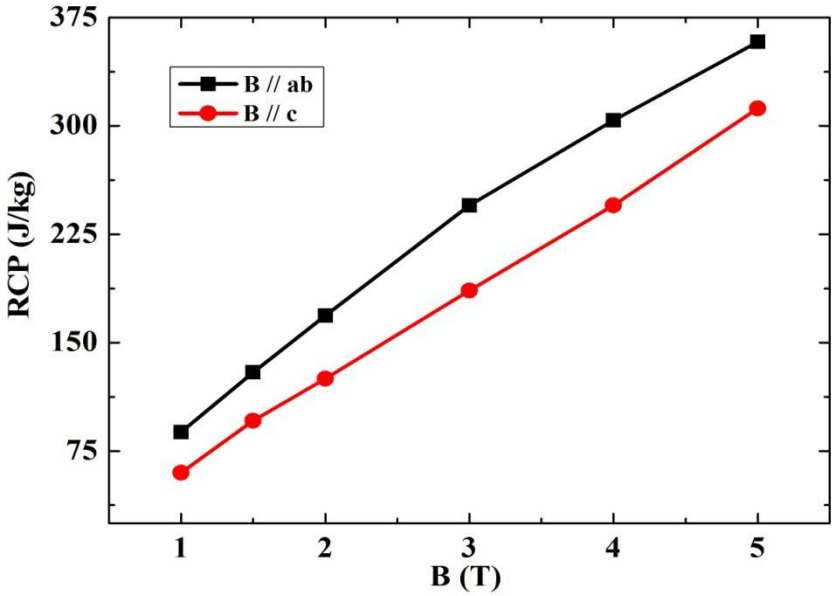

Fig. 10. Relative cooling power values (RCP) versus applied magnetic field for $La_{0.7}Ca_{0.3}MnO_3$ single crystal along the *ab*-plane and the *c*-direction.

In our case, the hysteresis loss is almost negligible, so the effective refrigerant capacity (RC_{eff}) is the same as the RC. For our sample, the value of RC_{eff} at 358.17 K is comparable to that of $Gd_5Ge_{1.9}Si_2Fe_{0.1}$ (about 355 J/kg) [41]. This value is also much larger than those of some other magnetocaloric materials for a field change of 0–5 T, such as $Gd_5Si_2Ge_2$ (about 240 J/kg) [41], $Ni_{50}Mn_{34}In_{16}$ (about 181 J/kg) [42], $LaFe_{11.0}Co_{0.9}Si_{1.1}$ (about 275 J/kg) [43], and $LaFe_{11.2}Co_{0.7}Si_{1.1}C_{0.1}$ (about 320 J/kg) [44]. Our study shows that the large values of RC_{eff} and the lack of magnetic hysteresis in $La_{0.7}Ca_{0.3}MnO_3$ single crystal favours practical application of the material near room temperature. This relatively high RC arises from the strongly asymmetric distribution of $|\Delta S_M|$ and the large $\delta T_{FWHM.}$ In addition, the composition inhomogeneity and/or non-stoichiometry of the phase may also contribute to the large $\delta T_{FWHM.}$

4. Conclusions

We have studied the MCE in $La_{0.7}Ca_{0.3}MnO_3$ single crystal. Due to the strong magnetocrystalline anisotropy, the crystal shows anisotropic magnetic entropy change. The uniformity of the ΔS_M distribution and the lack of any hysteresis loss are desirable for an Ericsson-cycle magnetic refrigerator and for high magnetic cooling efficiency. The nature of the magnetic transition is confirmed by the Landau coefficients. By adopting Takahashi's spin-fluctuation theory, we estimated the spin fluctuation parameters and calculated $1/\chi$. The convex curvature of M^2 against B/M in the form of an Arrott plot indicates that $La_{0.7}Ca_{0.3}MnO_3$ is WIF, and the characteristics of its spin fluctuations can be understood within three-dimensional SCR and Takahashi's spin-fluctuation theories, at least around T_C.

Acknowledgement

This work was supported by the Australian Research Council (DP0879070).

References

[1] S. Y. Dan'kov, A. M. Tishin, V. K. Pecharsky, K. A. Gschneidner, Jr., Phys. Rev. B 57 (1998) 3478.

[2] V. K. Pecharsky and K. A. Gschneidner, Jr., Phys. Rev. Lett. 78 (1997) 4494.

[3] V. K. Pecharsky, K. A. Gschneidner, Jr., Appl. Phys. Lett. 70 (1997) 3299.

[4] J. R. Sun, F. X. Hu, B. G. Shen, Phys. Rev. Lett. 85 (2000) 4191.

[5] H. Wada, Y. Tanabe, Appl. Phys. Lett. 79 (2001) 3302.

[6] F. X. Hu, B. G. Shen, J. R. Sun, Z. H. Cheng, G. H. Rao, X. X. Zhang, Appl. Phys. Lett. 78 (2001) 3675.

[7] D. H. Wang, S. L. Tang, H. D. Liu, W. L. Gao, Y. W. Du, Intermetallics 10 (2002) 819.

[8] S. Yu. Dan' kov, A.M. Tishin, V. K. Pecharsky, K. A. Gschneidner, Jr., Phys. Rev. B 57 (1998) 3478.

[9] Z. B. Guo, Y. W. Du, J. S. Zhu, H. Huang, W. P. Ding, D. Feng, Appl. Phys. Lett. 78 (1997) 1142.

[10] X. Bohigas, J. Tejada, E. del Barco, X. X. Zhang, M. Sales, Appl. Phys. Lett. 73 (1998) 390.

[11] A. Szewczyk, H. Szymczak, A. Wisniewski, K. Piotrowski, R. Kartaszynski, B. Dabrowski, S. Kolesnik, and Z. Bukowski, Appl. Phys. Lett. 77 (2000) 1026.

[12] M. H. Phan, S. B. Tian, S. C. Yu, and A. N. Ulyanov, J. Magn. Magn. Mater. 256 (2003) 306.

[13] Y. Sun, X. Xu, Y. Zhang, J. Magn. Magn. Mater. 219 (2000) 183.

[14] V. K. Pecharsky and K. A. Gschneidner, Jr., J. Magn. Magn. Mater. 200 (1999) 44.

[15] T. Moriya, and A. Kawabata, J. Phys. Soc. Jpn. 34 (1973) 639.

[16] Y. Takahashi and T. Moriya, J. Phys. Soc. Jpn. 54 (1985) 1592.

[17] T. Moriya, Spin Fluctuations in Itinerant Electron Magnetism (Springe-Verlag, New York, 1985).

[18] Y. Takahashi, J. Phys. Soc. Jpn. 55 (1986) 3553.

[19] M. H. Phan, S. C. Yu, N. H. Hur and Y. H. Jeong, J. Appl. Phys. 96 (2004) 1154.

[20] S. B. Tian, M. H. Phan, S. C. Yu and N. H. Hur, Physica B. 327 (2003) 221.

[21] K. A. Gschneidner, Jr., V. K. Pecharsky, A.O. Tsokol, Rep. Prog. Phys. 68 (2005) 1479.

[22] F. X. Hu, B. G. Shen, J. R. Sun, Z. H. Cheng, Phys. Rev. B 64 (2001) 012409.

[23] T. Tang, K. M. Gu, Q. Q. Cao, D. H. Wang, S. Y. Zhang, and Y. W. Du, J. Magn. Magn. Mater. 222 (2000) 110.

[24] Y. Sun, W. Tong, and Y. H. Zhang, J. Magn. Magn. Mater. 232 (2001) 205.

[25] Z. M. Wang, G. Ni, Q. Y. Xu, H. Sang, and Y. W. Du, *ibid.* 234 (2001) 371.

[26] H. X. Bohigas, J. Tejada, M. L. Marinez-Sarrion, S. Tripp, and R. Black, J. Magn. Magn. Mater. 208 (2000) 85.

[27] C. S. Hong, W. S. Kim, E. O. Chi, K. W. Lee, and N. H. Hur, Chem. Mater. 12 (2000) 3509.

[28] M. H. Phan, V. Srinivas, S. C. Yu, and N. H. Hur, J. Appl. Phys. 93 (2003) 8200.

[29] R. Gross *et al.*, J. Magn. Magn. Mater. 211 (2000) 150.

[30] S. K. Banerjee, Phys. Lett. 12 (1964) 16.

[31] A. Fujita, S. Fujieda, K. Fukamichi, J. Appl. Phys. 85 (1999) 4756.

[32] M. Yue, Z. Q. Li, X. B. Liu, H. Xu, D. M. Liu and J. X. Zhang, J. Alloys and Comp. 493 (2010) 22.

[33] X. B. Liu, D. H. Ryan, Z. Altounian, J. Magn. Magn. Mater. 270 (2004) 305.

[34] D. Bloch, J. Voiron, V. Jaccarino, and J. H. Wernick, Phys. Lett. 51A (1975) 259.

[35] H. Ohta and K. Yoshimura, Phys. Rev. B 79 (2009) 184407.

[36] B. Chen, C. Michioka, Y. Itoh, K. Yoshimura, J. Phys. Soc. Jpn. 77 (2008) 103708.

[37] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederich, J. Chazelas, Phys. Rev. Lett. 61 (1988) 2472.

[38] G. Binasch, P. Grünberg, F. Saurenbach, W. Zinn, Phys. Rev. B 39 (1989) 4828.

[39] K. A. Gschneidner, Jr., V. K. Pecharsky, A. O. Pecharsky, C. B. Zimm, Mater. Sci. Forum 315 (1999) 69.

[40] V. K. Pecharsky and K. A. Gschneidner, Jr., J. Magn. Magn. Mater. 167 (1997) L179.

[41] V. Provenzano, A. J. Shapiro, and R. D. Shull, Nature (London) 429 (2004) 853.

[42] V. K. Sharma, M. K. Chattopadhyay, S. B. Roy, J. Phys. D 40 (2007) 1869.

[43] M. Balli, D. Fruchart, D. Gignoux, J. Phys.: Condens. Matter 19 (2007) 236230.

[44] M. Balli, D. Fruchart, D. Gignoux, Appl. Phys. Lett. 92 (2008) 232505.