Magneto-optics observation of spontaneous domain structure in ferromagnetic $La_{0.78}Ca_{0.22}MnO_3$ single crystal

2004 J. Phys.: Condens. Matter 16 5461

(http://iopscience.iop.org/0953-8984/16/30/009)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **16** (2004) 5461–5468 PII: S0953-8984(04)74714-1

Magneto-optics observation of spontaneous domain structure in ferromagnetic $La_{0.78}Ca_{0.22}MnO_3$ single crystal

G Jung[1,2], M Indenbom[1], V Markovich[2], C J van der Beek[2], D Mogilyansky[3] and Ya M Mukovskii[4]

[1] Laboratoire des Solides Irradiès, Ecole Polytechnique, 91128 Palaiseau, France
[2] Department of Physics, Ben Gurion University of the Negev, PO Box 653, 84105 Beer Sheva, Israel
[3] Institute of Applied Research, Ben Gurion University of the Negev, PO Box 653, 84105 Beer Sheva, Israel
[4] Moscow State Steel and Alloys Institute, 117936, Moscow, Russia

E-mail: jung@bgumail.bgu.ac.il

Received 11 January 2004
Published 16 July 2004
Online at stacks.iop.org/JPhysCM/16/5461
doi:10.1088/0953-8984/16/30/009

Abstract

Spontaneous ferromagnetic domains in lightly Ca-doped $La_{1-x}Ca_xMnO_3$ single crystals have been visualized and investigated by means of the magneto-optical technique. In marked difference to the magnetic contrast structures associated with magneto-crystalline anisotropy, which appear only in applied magnetic field, spontaneous ferromagnetic domains show up at low temperatures below the Curie temperature in zero applied field and are characterized by oppositely oriented spontaneous magnetization in adjacent domains. Ferromagnetic domains seen in zero field cooled samples take the form of almost periodic, corrugated stripe-like structures. Application of even a very weak magnetic field during cooling through the magnetic ordering transition changes the stripe domain structures into a bubble domain system.

1. Introduction

Rare-earth manganites with perovskite structure and composition $R_{1-x}A_xMnO_3$, where R stands for a trivalent rare-earth ion and A for a divalent alkaline ion, exhibit the colossal magnetoresistance effect (CMR) resulting from a subtle balance between spin, orbital, charge and lattice degrees of freedom. CMR manganites have a complex phase diagram in which a metal–insulator (M–I) transition occurs in close vicinity to the magnetic ordering transition [1]. The transport properties of CMR manganites at low temperatures are as yet not well understood. On the other hand, it is well-known that the electrical resistance of the magnetic domain walls

strongly influence transport properties and magnetoresistance of classical ferromagnets and CMR manganites [2, 3]. We have recently speculated that the increase of the resistivity at low temperatures in lightly Ca-doped manganite single crystals may be associated with spin-dependent transport across intrinsic twin-boundary tunnel junctions located within the domain walls separating neighbouring ferromagnetic domains with opposite orientation of the magnetic moments [4]. A serial arrangement of such junctions leads to pronounced resistance oscillations [5]. Since the domain wall resistance, independently of the mechanism of wall scattering, critically depends on the domain configuration, an effort devoted to elucidating the micromagnetic structure and topology of spontaneous ferromagnetic domains in CMR materials is welcome. Surprisingly, little is known about the magnetic domain structures in CMR perovskite single crystals. Although several papers present evidence of the appearance of magnetic contrast structures below the Curie temperature, most of them appear only in an applied magnetic field, contrary to what is expected for spontaneous ferromagnetic domains. Moreover, up until now, the majority of experiments have been performed with layered manganites and thin film samples only.

Magnetic structures in manganites have been investigated experimentally by means of magneto-optical (MO) techniques [6–13], magnetic force microscopy (MFM) [14–19], scanning SQUID (SSM) and Hall probe microscopy (SHM) [20–22], Bitter decoration [23], Lorentz electron microscopy [24–26] and neutron depolarization measurements [27]. The last two techniques are clearly most suitable for resolving fine structures with domain sizes in the range of 10 μm and smaller. Some of the methods inevitably perturb the investigated magnetic structures. For example, the field of MFM ferromagnetic cantilevers, even if very weak, may modify the domain structure in the investigated specimen. Scanning microscopy techniques have the disadvantage of a relatively small observation area. The MO technique allows visualization of large surfaces at a variable spatial resolution. Moreover, the MO technique is non-invasive since the weak anisotropy of the magneto-optical garnet indicator leaves the magnetic structure of the investigated samples nearly undisturbed. This motivated us to choose this technique in our experiments.

2. Experimental details

Investigations of the ferromagnetic domain structure in CMR manganites should be executed with appropriate care. In order to reveal spontaneous ferromagnetic domains a sample has to be cooled through the magnetic ordering phase transition under zero field-cooled conditions (ZFC). An applied magnetic field reveals, first of all, magnetic structures that are related to the material inhomogeneity and magnetic anisotropy. These may completely dominate, or even smear out, the spontaneous ferromagnetic domains. Moreover, as will be discussed below, even a very weak magnetic field, orders of magnitude weaker than the saturation field of the spontaneous domain, may significantly alter the topology of ferromagnetic domains in CMR manganites.

To the best of our knowledge the only ZFC MO study of magnetic domains in CMR manganites has been carried out using strongly anisotropic layered $La_{1.36}Ca_{1.64}Mn_2O_7$ single crystals [13]. Other known MO investigations of CMR manganites were performed either under field cooling (FC) conditions or in applied magnetic fields. Most of the reported studies concern manganite thin films, where the domain structure will be clearly dominated by the properties of the substrate. For example, FC MO imaging of $La_{2/3}Ca_{1/3}MnO_3$ thin films has revealed magnetic contrast due to the appearance of twins associated with martensitic transition in the film triggered by the cubic-to-tetragonal transition in the $SrTiO_3$ substrate [8, 9].

The image of the anisotropic field distribution on the surface of a ferromagnetic sample immersed in a uniform field can be easily confused with the expected image of

spontaneous ferromagnetic domains. For example, in the reports on magneto-optical and high resolution Bitter investigations of $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.8}Ca_{0.2}MnO_3$ single crystals, the magnetic contrast has been seen only under applied field while the zero field images were featureless [6, 23]. The correlations between magnetic and twin structures reported in [6] mean that the magnetic contrast observed in non-zero field has to be attributed to the magnetic crystalline anisotropy only, and not to the ferromagnetic domains. In this paper we report on the visualization, by means of the magneto-optical technique, of spontaneous ferromagnetic domains in a series of $La_{1-x}Ca_xMnO_3$ (LCMO) single crystals. Although we have detected the presence of spontaneous domains in crystals with various doping levels x, here we concentrate on domain structures in LCMO with a doping level close to the percolation threshold $x_C \approx 0.22$ [28]. These structures are of particular interest because of the possible correlation between the domain structure and transport properties of low-doped LCMO [29].

The LCMO crystals were grown by a floating zone method using radiative heating as reported in detail elsewhere [30]. The as-grown crystal is a cylinder of diameter 4 mm and 40 mm high, with the axis oriented close to the $\langle 110 \rangle$ crystalline direction. Magnetic and transport measurements indicated that the M–I transition occurs at a temperature very close to the Curie temperature T_C of each crystal [28, 31]. For MO imaging of the local magnetic induction distribution on the surface of the LCMO crystal, several disc-like slices, with thicknesses ranging between 0.5 and 1 mm, were cut from the crystal. The flat surfaces were polished to optical quality. The polished sample was mounted on the cold finger of the flow cryostat for magneto-optical measurements. A ferrimagnetic garnet indicator film with in-plane anisotropy was placed directly on top of the polished plate, and observed using linearly polarized light. The reflected light intensity, observed through an analyser oriented nearly perpendicular to the polarization direction of the incident light, is related to the local value of the magnetic induction component perpendicular to the crystal and to the garnet. At small magnetic fields the polarization rotation angle depends on the field linearly, leading to a quadratic dependence of the image intensity on the local magnetic field.

3. Results and discussion

Figure 1(a) shows the MO image of the polished $(\bar{1}10)$ surface of $La_{0.8}Ca_{0.2}MnO_3$ single crystal, seen after zero field cooling procedures at 31 K. The arrangement of parallel bright and dark stripe domains starts to be seen at temperatures just below the Curie temperature $T_C = 183$ K. Bright and dark stripes in this MO micrograph represent opposite orientations of the magnetic moments in adjacent domains, up and down with respect to the crystal surface. Application of an external magnetic field during cooling through the paramagnetic–ferromagnetic transition drastically changes the topology of the ferromagnetic domains. As illustrated in figure 1(b), the previously observed stripe domains are replaced by bubble domains as a result of application of a magnetic field of only 40 G during cooling through T_C. This demonstrates that even very weak magnetic fields, such as those produced by the tip of a MFM cantilever, can significantly influence the domain pattern in CMR manganites. An external magnetic field is normally needed to stabilize the bubble domains. Surprisingly, the bubble pattern in the investigated field cooled crystals remains stable even in the remanent state, after turning off the field.

Figure 2 shows MO micrographs of the [110] surface of $La_{0.78}Ca_{0.22}MnO_3$ crystal ZFC cooled to 90 K, as seen in zero and nonzero applied fields of opposite orientation. Spontaneous corrugated stripe-like domains are evident over a significant part of the surface. The restriction of the area occupied by the visible domains in the MO micrograph can be related to an apparently insufficient degree of polishing and flattening of the surface which resulted in a limited area of close adherence of the MO indicator to the crystal surface. As will be shown

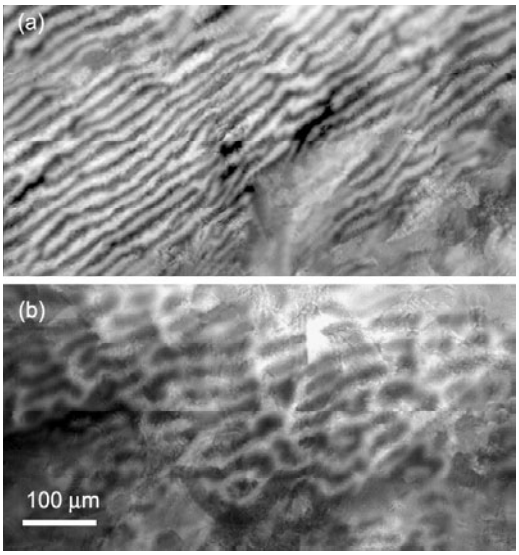

Figure 1. MO image of the ($\bar{1}10$) surface of the $La_{0.8}Ca_{0.2}MnO_3$ single crystal: (a) after zero field cooling to 31 K, (b) after FC to 30 K in 40 G.

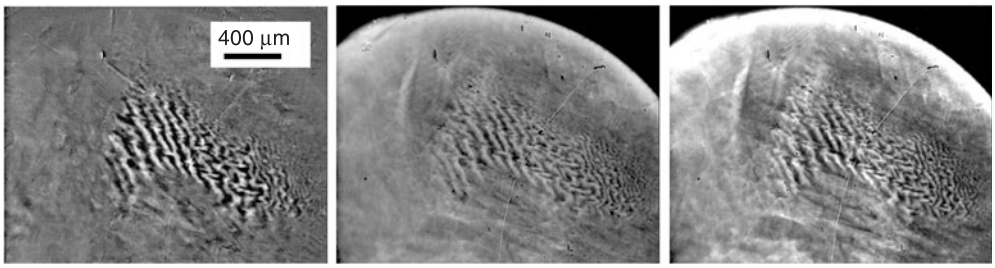

Figure 2. MO image of the (110) surface of the $La_{0.78}Ca_{0.22}MnO_3$ crystal after zero field cooling to 90 K. The external magnetic field H applied perpendicularly to the sample surface is: (a) $H = 0$, (b) $H = 460$ G, (c) $H = -460$ G.

latter, the surface field of the spontaneous ferromagnetic domain is very weak and an even small gap between the MO indicator and sample surface may result in the loss of image detail. Yet another reason why the domain structure is not seen in the entire surface may be the stress introduced by the polishing process [32].

Application of an external field, in the direction perpendicular to the sample surface, not only magnetizes the sample, the circular shape of which appears in the MO image seen in figures 2(b) and (c), but also reveals yet another kind of magnetic contrast. This contrast however, is related only to the magnetic anisotropy associated with twin domains and not to the spontaneous ferromagnetic domains. Different directions of the easy magnetization axis in neighbouring twin domains results in a difference between the applied field components normal to the surface in adjacent twin domains [6, 33]. With the reversal of the direction of the external field, twin domains lying closer to the easy magnetization axis always show higher induction, even though the orientation of the induction changes so as to be aligned with the direction of the externally applied field. By contrast, the spontaneous domains retain

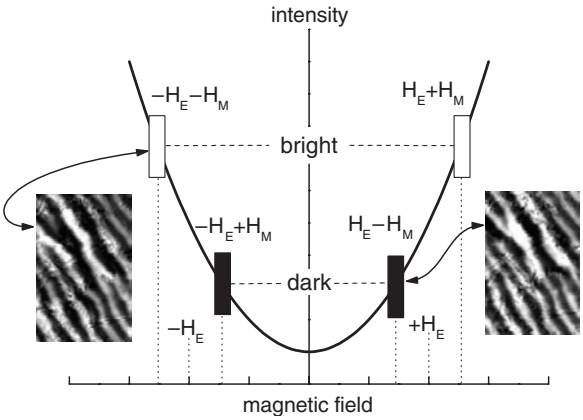

Figure 3. Mechanism leading to a change of the ferromagnetic domain intensity with changing direction of the applied magnetic field H_E. The image of the domain with a local value of the surface magnetic field $+H_M$ is bright for the field direction coinciding with the direction of the domain field (positive fields) and becomes dark for the opposite direction of the applied field.

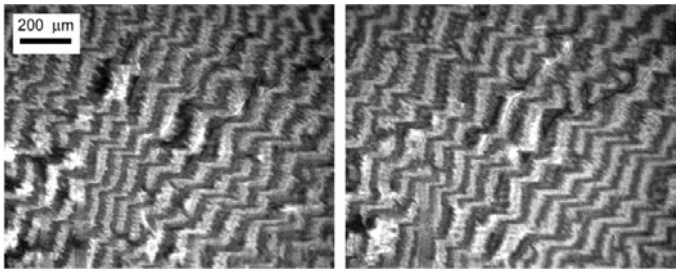

Figure 4. MO image of two different realizations of spontaneous domain patterns in an $x = 0.22$ LCMO single crystal obtained by ZFC to 150 K, returning to temperatures above T_C and subsequent ZFC cooling back to 150 K.

the direction of their spontaneous magnetization under the reversal of the field direction, as reflected in the mutual interchange of bright and dark domains in figures 2(b) and (c). The mechanism leading to the intensity interchange with changing polarity of the applied field is explained in figure 3. MO images taken with non-zero applied field also reveal that the rough corrugation pattern of the spontaneous domains is caused by their intersections with underlying crystallographic twin domains.

The character of the spontaneous domain structure remains similar when the crystal is taken through a number of thermal cycles between low temperature and some temperature well above T_C. However, the detailed geometry of the domain structure is different after each cycle, as shown in the expanded views of figure 4. Simultaneously, the geometry of the magnetic contrast due to twin domains remains rigorously identical for all realizations.

The spontaneous domain structure is relatively robust and its general shape does not change significantly upon application of the maximum magnetic field available in our MO setup, as illustrated in figures 2(b) and (c). However, the maximum available field is relatively weak, of the order of 0.5 kG, and directions of spontaneous moments are clearly not inverted by such a field, i.e. the saturation field is much larger than that which we can apply in our experiment. Detailed scrutiny of significantly magnified images revealed that the application of available

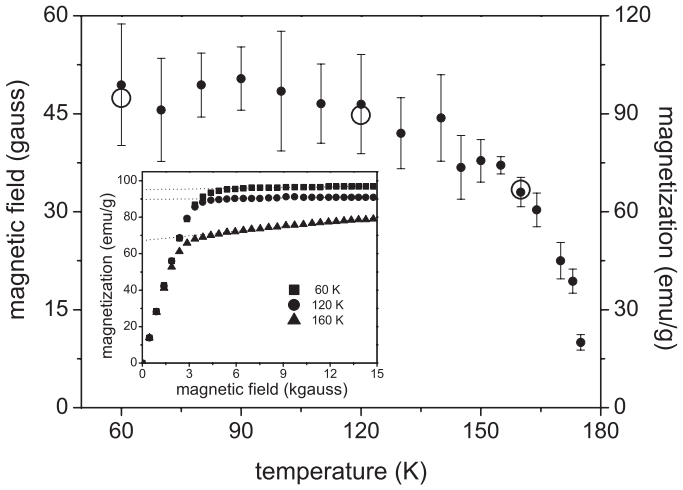

Figure 5. Temperature evolution of the normal component of the magnetic field at the domain surface in $x = 0.22$ LCMO as evaluated from the MO contrast. The open symbols represent calibrating magnetization data obtained from the independent magnetometric measurements shown in the inset.

external fields causes only slight motion of the domain walls resulting in a slight increase of the thickness of the domains of spontaneous magnetization oriented along the applied field direction.

By calibrating the intensity of the magneto-optical image contrast against the known applied magnetic induction in the areas far away from the sample we were able to measure the values of the normal component of the magnetic field at the domain surface $H_\perp$. The value of $H_\perp$ evaluated in this way (figure 5) increases with decreasing temperature and saturates at low temperatures, similar to the temperature evolution of the spontaneous magnetization in classical ferromagnets. Note however, that the absolute value of $H_\perp$ is very low, below 100 G. This can be explained by the fact that we are measuring only the normal component of the field, strongly reduced by the presence of closure structure at the sample surface [32]. The domain branching into closure domains is visible as circular spots within the domain structure in an enlarged image shown in figure 4. They became significantly more pronounced at lower temperatures.

To calibrate and verify the MO measurements we have independently evaluated the spontaneous magnetization by an extrapolation to zero field of the high field part of the volume magnetization versus applied field curve, measured with a vibrating sample magnetometer (inset to figure 5). The result, for three temperatures, is shown as the open symbols in figure 5. These well coincide with the magneto-optically determined temperature dependence of the saturation magnetization. This fact confirms that the observed MO contrast indeed corresponds to the spontaneous magnetization of the magnetic domains.

Within experimental error, the width of the domain structure remains almost temperature-independent, as seen in figure 6. Note that the spontaneous domains are relatively wide. The domain width of several dozens of microns is orders of magnitude larger than the typical nanometric size of the domains due to phase separation [34]. The size difference reflects the fact that phase separated domains differ primarily in their electronic properties, whereas the magnetic order, in particular in low-doped LCMO system, may remain the same [1, 33]. A significant increase of data scatter and value of the associated measurement

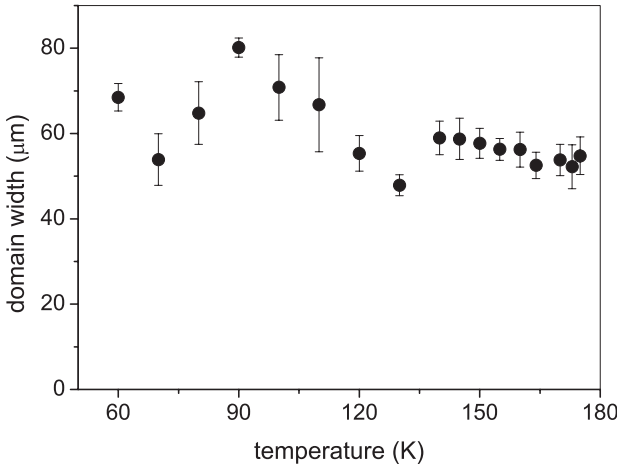

Figure 6. Temperature evolution of the spontaneous domain width in $x = 0.22$ LCMO.

error at temperatures below 140 K is due to changes in the domain structure occurring below this temperature. As will be discussed in detail elsewhere, the stripe-like domain structure undergoes a strong corrugation and buckling transition, most likely due to the temperature changes of the magnetic anisotropy.

In summary, we have demonstrated that the magneto-optical technique is suitable for the direct unperturbed investigations of spontaneous ferromagnetic domains in CMR manganites. The surface field of the spontaneous domains is very weak, much weaker than the expected saturation fields. We have shown that the domain structure is very sensitive to any externally applied magnetic field, a fact that requires a critical revision of several published experimental results. In particular, our observations put doubt on the results of several experiments in which an external field was needed to reveal the domain pattern. In particular, we disagree with the claims that external fields much weaker than the domain saturation or coercive fields cannot significantly change the domain pattern [23]. All investigated manganite samples, including those with unpolished surfaces, have shown pronounced magneto-optic contrast structures after application of the field at low temperatures. This dominating magnetic contrast is due to twin domains and results from magneto-crystalline anisotropy only. Care should be taken not to confuse these structures with spontaneous ferromagnetic domains.

Acknowledgment

This research was supported by the Israeli Science Foundation administered by the Israel Academy of Sciences and Humanities (grant 209/01).

References

[1] Dagotto E, Hotta T and Moreo A 2001 *Phys. Rep.* **344** 1
[2] Kent A D, Yu J, Rüdiger U and Parkin S S P 2001 *J. Phys.: Condens. Matter* **13** R461
[3] Ziese M 2002 *Rep. Prog. Phys.* **65** 143
[4] Yuzhelevski Y, Markovich V, Dikovsky V, Rozenberg E, Gorodetsky G, Jung G, Shulyatev D A and Mukovskii Ya M 2001 *Phys. Rev.* B **64** 224428
[5] Dikovsky V, Yuzhelevski Y, Markovich V, Rozenberg E, Gorodetsky G, Jung G, Shulyatev D A and Mukovskii Ya M 2002 *Phys. Rev.* B **65** 144439

[6] Khapikov A, Uspenskaya L, Bdikin I, Mukovskii Ya M, Karabashev S, Shulyatev D and Arsenov A 2000 *Appl. Phys. Lett.* **77** 2376
[7] Lin Z W, Cochrane J W and Russell G J 1999 *Appl. Phys. Lett.* **74** 3014
[8] Vlasko-Vlasov V K, Lin Y K, Miller D J, Welp U, Crabtree G W and Nilitenko V I 2000 *Phys. Rev. Lett.* **84** 2239
[9] Vlasko-Vlasov V K, Lin Y, Welp U, Crabtree G W, Miller D J and Nilitenko V I 2000 *J. Appl. Phys.* **87** 5828
[10] Tokunaga M, Tokunaga Y, Yasugaki M and Tamegai T 2001 *J. Magn. Magn. Mater.* **226–230** 852
[11] Welp U, Berger A, Miller D J, Vlasko-Vlasov V K, Gray K E and Mitchel J F 1999 *Phys. Rev. Lett.* **83** 4180
[12] Welp U, Berger A, Miller D J, Vlasko-Vlasov V K, Gray K E and Mitchel J F 2000 *J. Appl. Phys.* **87** 5043
[13] Welp U, Berger A, Vlasko-Vlasov V K, Li Q, Gray K E and Mitchel J F 2000 *Phys. Rev.* B **62** 8615
[14] Lu Q, Chen C-C and de Lozanne A 1997 *Science* **276** 2006
[15] Suzuki Y, Wu Y, Yu J, Ruediger U, Kent A D, Nath T K and Eom C B 2000 *J. Appl. Phys.* **87** 6746
[16] Wu Y, Matshushita Y and Suzuki Y 2001 *Phys. Rev.* B **64** 220404
[17] Soh Y-A, Aeppli G, Mathur N D and Blamire M G 2000 *J. Appl. Phys.* **87** 6743
[18] Soh Y-A, Aeppli G, Mathur N D and Blamire M G 2001 *J. Magn. Magn. Mater.* **226–230** 857
[19] Popov G, Kalinin S V, Alvarez T, Emge T J, Greenblatt M and Bonnell D A 2002 *Phys. Rev.* B **65** 064426
[20] Fan X-J, Koinuma H and Hasegawa T 2002 *Phys. Rev.* B **65** 144401
[21] Fukumura T, Sugawara H, Hasegawa T, Tanaka K, Sakaki H, Kimura T and Tokura Y 1999 *Science* **284** 1969
[22] Fukumura T, Hasegawa T, Sugawara H, Tanaka K, Sakaki H, Kimura T and Tokura Y 2001 *Appl. Phys. Lett.* **78** 2023
[23] Barkov F L, Vinnikov L Ya, Tulina N A, Zver'kov S A, Arsenov A, Mukovskii Y M and Shulyatev D A 2003 *Europhys. Lett.* **61** 242
[24] Lloyd S J, Mathur N D, Loudon J C and Midgley P A 2001 *Phys. Rev.* B **64** 172407
[25] Asaka T, Anan Y, Nagai T, Tsutsumi S, Kuwahara H, Kimoto K, Tokura Y and Matsui Y 2002 *Phys. Rev. Lett.* **89** 207203
[26] Mori S, Asaka T and Matsui Y 2002 *J. Electron. Microsc.* **51** 225
[27] Yusuf S M, Sahana M, Hegde M S, Dörr K and Müller K H 2000 *Phys. Rev.* B **62** 1118
[28] Markovich V, Yuzhelevski Y, Gorodetsky G, Jung G, van der Beek C J and Mukovskii Ya M 2003 *Eur. Phys. J.* B **35** 295
[29] Jung G, Markovich V, Yuzhelevski Y, Indenbom M, van der Beek C J, Mogilyansky D and Mukovskii Ya M 2004 *J. Magn. Magn. Mater.* **272–276** 1801
[30] Shulyatev D A, Arsenov A A, Karabashev S G and Mukovskii Ya M 1999 *J. Cryst. Growth* **198/199** 511
[31] Markovich V, Fita I, Puzniak R, Tsindlekht M I, Wisniewski A and Gorodetsky G 2002 *Phys. Rev.* B **66** 094409
[32] Hubert A and Schafer R 1998 *Magnetic Domains* (Berlin: Springer)
[33] Van Aken B B, Meetsma A, Tomioka Y, Tokura Y and Palstra T T M 2002 *Phys. Rev.* B **66** 224414
[34] Radaelli P G, Ibberson R M, Argyriou D N, Casalta H, Andersen K H, Cheong S-W and Mitchell J F 2001 *Phys. Rev.* B **63** 172419