

Temperature-dependence of magnetic domains in phase-separated manganites

Holly Miller

Department of Natural Sciences, Longwood University, Farmville, Virginia

Amlan Biswas

Department of Physics, University of Florida, Gainesville, Florida

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The temperature dependence of the magnetic domain structure for the hole-doped manganese oxide $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, was studied in both the single-crystal and thin film forms. After the samples were heated, the magnetic domain patterns were observed using a magnetic force microscope (MFM). The ferromagnetic domains of the crystal disappear at around 320 K, which is well below the bulk Curie temperature (T_C) of 345 K. In the case of the film, the ferromagnetic domains disappear at an even lower temperature of about 309 K. Evidence is presented which shows that the coexistence of the charge ordered insulating and ferromagnetic metallic phases in the thin films is responsible for this anomalous behavior.

Introduction

Hole-doped manganese oxides (manganites), known mostly for exhibiting the phenomenon of colossal magnetoresistance (CMR) have attracted attention over the recent years due to their other unique properties such as 100% spin polarization [2] and first-order paramagnetic to ferromagnetic transition at the Curie temperature. These unique properties point to hitherto unknown phenomena and can also lead to possible device applications. It has been shown recently that there is a coexistence of the ferromagnetic metallic (FMM) and charge-ordered insulating (COI) phases in these materials for a certain temperature range [3]. Such coexistence is due to the presence of two possible ground states (the FMM and COI phases), which are very close in energy. This coexistence leads to an extreme sensitivity to external perturbations in these materials [4]. Such sensitivity to perturbations can possibly provide us with a new material for magnetic sensors and magnetic storage media [5].

Hole-doped manganites have the general formula $RE_{1-x}AE_xMnO_3$ (RE=rare earth ion and AE=alkaline earth ion) and are formed by replacing some of the rare-earth ions (La, Pr, Nd) in the parent compound $LaMnO_3$, by an alkaline earth ion (Ca, Sr). This process of replacing ions is called doping. Doping introduces a mixed valency of Mn ion viz. Mn^{3+} and Mn^{4+} . The perovskite structure of a manganite shown in Fig. 1(a) can better demonstrate this concept [6]. The mixture of valence Mn^{3+}/Mn^{4+} leads to a rich and varied phase diagram (see Fig. 1 (b)and (c)) [7], which includes the COI and FMM phases for different values of doping. In fact, below their Curie temperature, manganites

such as $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) are fully spin-polarized ferromagnets. The objective of this project is to directly map the domain structure of LSMO and examine it as function of temperature. Given that the Curie temperature (T_C) of LSMO is about 350 K, it is expected to retain most of its spin polarization at room temperature (300 K). However, manganite-based devices have shown limited success to date, and it has been difficult to

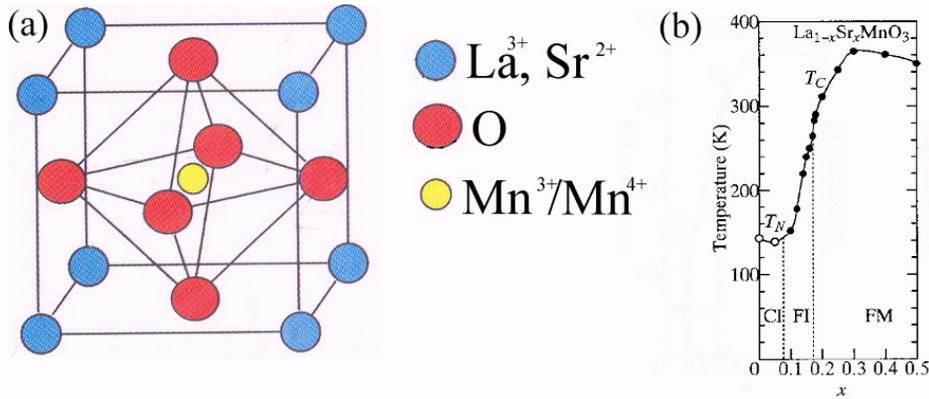
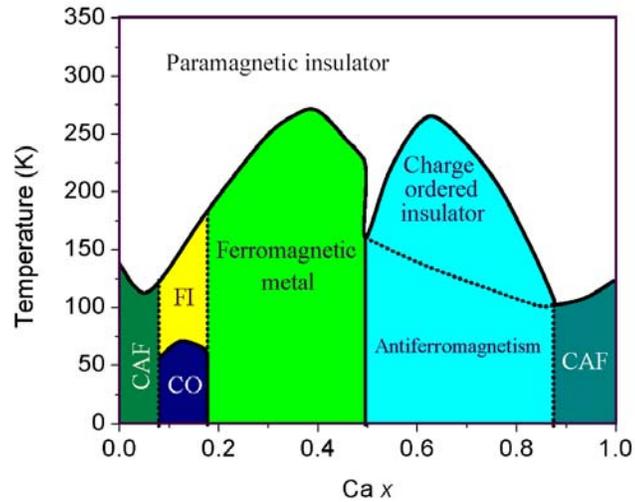


Fig. 1 (a) Hole-doped manganites with perovskite structure include a manganese atom enclosed by six oxygen atoms. Depending on the amount of doping, a mixture of rare earth and alkaline earth cations fill in the eight corners of the cube. (b) The Phase diagram of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ at different values of doping. As shown, when $x=0.3$ the magnetism is expected to remain ferromagnetic until just above 360 K. (c) Phase diagram of $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ containing a charge-ordered region.



fabricate devices which function well at room temperature [8]. One of the motivations for studying the domain dynamics is to better understand why such devices composed of this specific material do not exhibit the expected features at room temperature. Does the strain due to the substrate on thin film multilayers nucleate a competing phase, which affects

the FM domain dynamics of ferromagnetic LSMO? In the following study we present probable evidence of the coexistence of magnetic phases in LSMO when it is subjected to external strain and show that the magnetism at the surface is significantly affected.

We have examined LSMO in both single crystal and thin film forms. The single crystal represents a strain free sample, which is measured as a standard, and the thin film provides an LSMO sample subjected to external strain. Using the magnetic force imaging (MFM) mode of the atomic force microscope (AFM), we obtained images of the topography and also magnetic domain patterns [9]. Using a coiled heating device we imaged the samples at several temperatures from about 300-340K. We observed large changes in the magnetism well below the materials' Curie temperature. In the crystal form, we have observed a significant difference between the topographical and magnetic images at the lower temperatures. This shows that the topography and magnetism are not correlated. As we approach 320 K the contrast of the magnetic image disappears. However, in the thin film the topography and magnetism are more correlated and we observed a change in the magnetism at a lower temperature than the crystal (309 K).

Instruments, Materials, and Methods

We imaged LSMO in the crystal and thin film forms-both with a doping of $x \approx 0.3$. The crystal sample used in our experiment is 3.5×2.2 mm with a thickness of 0.7 mm. The films were grown on LaAlO_3 (LAO) and NdGaO_3 (NGO) substrates by pulsed laser deposition and were 500 Å in thickness. LAO has a lattice mismatch of about 2 % with LSMO, and NGO is nearly lattice matched with LSMO.

Using the atomic force microscope (AFM), we took images of both the topography (tapping mode) as well as the magnetic domain patterns (magnetic force microscopy). We used a commercial tip of etched silicon coated with cobalt chromium. The cantilever to which the tip is attached has a resonance frequency of 59.3 kHz. Another key component of the AFM is the scanner, which is an expandable/contactable cylindrical device. This scanner is made of the piezoelectric material, lead zirconate titanate (PZT).

As mentioned before, the magnetic images were obtained using a magnetic force microscope (MFM). When using the lift mode option, the cantilever ascends to a preset lift scan height and measures the magnetic force between the tip and sample at a higher distance than that of the topography. By scanning the topography first, the MFM has an advantage called the look-ahead gain. This feature enables the cantilever to better anticipate the higher and lower-lying regions. It accounts for the topography while reacting to magnetic influences on the second trace. The magnetic forces are measured using force gradient detection. In our particular experiment we used phase detection, an option that uses the cantilever's oscillations relative to the drive of the piezo to form an image.

In order to study the effects of temperature on the magnetic domains we designed and built a heating device that fit on top of our piezo yet still allowed for scanning. In order to avoid destroying its polarization, we had to thermally isolate the heated platform from the piezo tube. This was achieved by separating the heated copper platform from the magnetic sample holder using three pieces of boron nitride. We singly coiled manganin wire (which has a high resistivity and a low temperature coefficient of resistivity) around

the copper platform. After obtaining our first set of results we decided to make a double-coiled heater. To ensure that the magnetic field produced from our first (single-coiled) heater did not contribute to our results [10]. The double coil produces a magnetic field in both directions producing a net magnetic field of zero. The temperature was measured using a diode thermometer. The calibration curves (current vs. temperature) for the two heaters are shown in Appendix A.

We started by magnetizing the tip by simply placing it along with the tip holder on a permanent magnet for roughly 10 minutes. Our first scans were of a magnetic tape provided by the manufacturers of the AFM. The scans of the tape can be compared to the images given in the instruction manual. This is a way to check that the MFM tip and imaging parameters are in the proper imaging condition. Using different drive amplitudes and lift scan height distances, one can change the values to best match the illustrations in the manual. A sample image is shown in Appendix B.

After imaging the tape, we started collecting data on the LSMO samples. We first examined the crystal. By passing different currents through our heating device, we were able to heat the sample to the desired temperatures. Beginning with room temperature (about 301 K), we increased the temperature in increments of about 1-5 K. After obtaining data on the crystal, we then studied the effects of temperature on the magnetic domain patterns of the thin films.

Results

Overall, with a rise in temperature we noted a change in the magnetic domains of both the crystal and thin films. Upon measuring the crystal's magnetization using an

MPMS SQUID magnetometer [11], we found the T_C to be 345 K. In Fig. 2, the crystal's topography and magnetism are shown (a) at 301 K (room temperature) and (b) at 335 K.

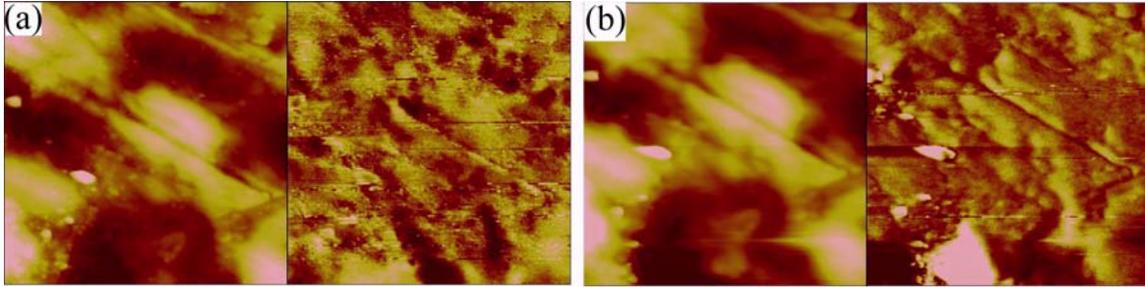


Fig. 2 The ferromagnetic domain structure at different temperatures. The topography (left images) shows little to no change when increasing the temperature from 301 K (a) to 335 K (b). However there is an obvious difference in the magnetism (right images) upon heating. The images are $5\mu\text{m} \times 5\mu\text{m}$.

Here it is quite noticeable that as the temperature is increased, the magnetic domains are not as obvious, and we are left with an image that is considerably close to that of the topography. Fig. 3 shows the changes in magnetism of the same region on the crystal with temperature. As seen, the magnetic image becomes similar to the topography

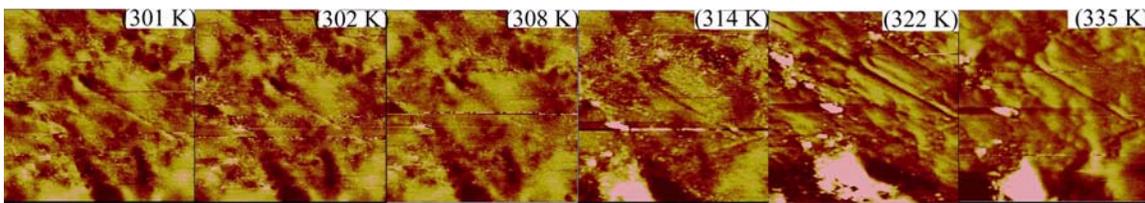


Fig. 3 Magnetic images of the crystal from 301-335 K. The domains begin to disappear at 314 K. Images are $5\mu\text{m} \times 5\mu\text{m}$.

between 314 K and 322 K. The presence of the topography suggests the disappearance of the FMM domains and a transformation into a non-ferromagnetic phase. We use the term

“non-ferromagnetic,” although it is most likely a paramagnetic phase. To check the dependence of the magnetic images on the topography, we took the cross-correlation between the magnetic and topographic images. Fig. 4 shows the cross-correlation of the topographic

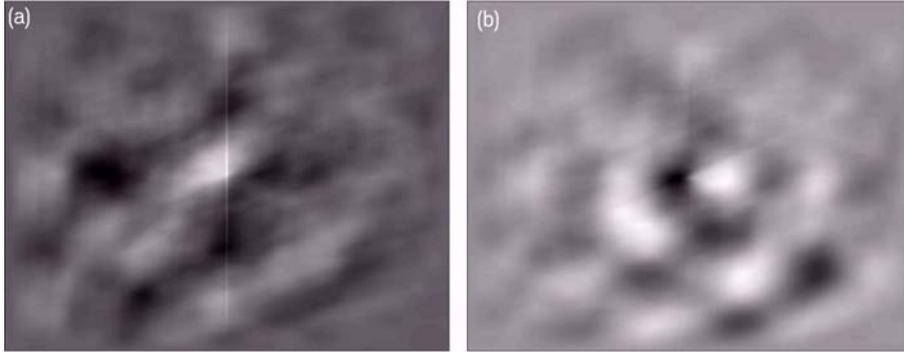


Fig. 4. Cross-correlation of the topographic and magnetic images at (a) 301K and (b) 335 K. The brighter areas indicate high correlation. The center of the image represents zero displacement between the topographic and magnetic images (see also Fig. 7).

and magnetic images at the two extreme temperatures [(a) 301 and (b) 335 K]. The brighter regions denote the positions of high correlation between the topography and magnetic image (center of the image is zero displacement between the topographic and magnetic images) [12]. An important observation is that these brighter regions tend to be scattered as opposed to being centered. The broad features in the cross-correlation images suggest low correlation between the topography and magnetism.

In the thin films, we noticed a change in the magnetic domains at a lower temperature. The insulator-metal transition of the thin is at 360 K. Fig. 5 shows the change in the magnetic domain structure of the thin film (on NGO substrate) from 301 K (a) and 315 K (b). At lower temperatures the magnetic image shows some space between

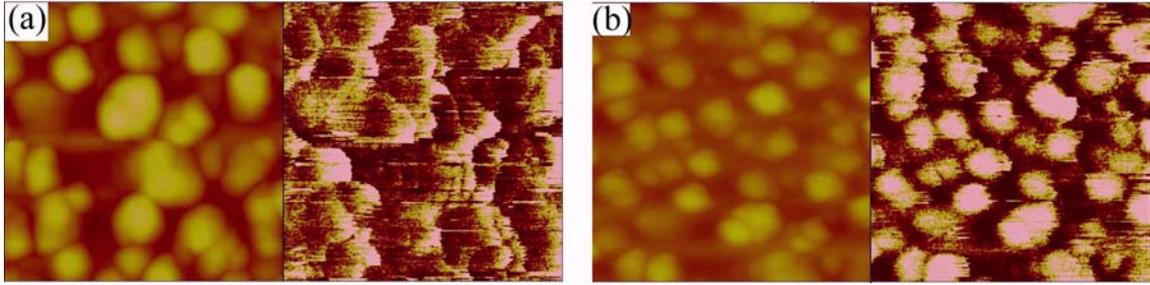


Fig. 5. Thin film images of LSMO on NGO at **(a)** 301 K and **(b)** 315 K. Both the topographic and magnetic images show a region of size 450nm x 450nm.

the domains. Whether this space is paramagnetic insulating or charge-ordered insulating cannot be confirmed from these images. In Fig. 6 changes in the magnetism of the same film on NGO become evident at 309 K, a lower temperature than the crystal. Fig. 7 provides the cross-correlations of the topography and the magnetism of this thin film at (a) 301 K and (b) 315 K. The thin film displayed a higher correlation between the

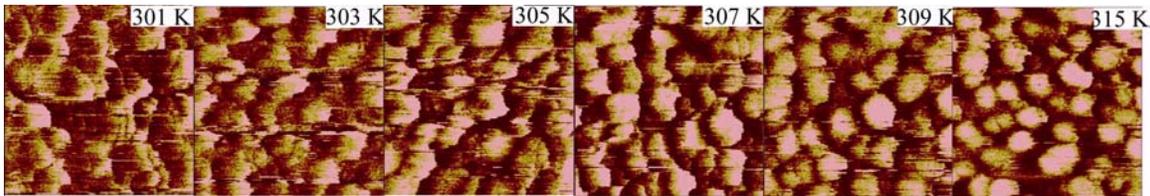


Fig. 6. Changes in magnetic domains in the thin film. The topography appears at about 307 K but dominates from 309 K. This is different from the crystal (see **Fig. 3**).

topography and magnetism at all temperatures. However, with no heat applied (301 K), the point of highest correlation is somewhat offset from the origin. For the highest temperature studied (315 K), the point that identifies the closest relationship between the two (topography and magnetism) is almost directly centered. This finding shows that at higher temperatures, the topography dominates the magnetic image and domain structure

has been wiped out. At lower temperatures (below 309 K), the topography and magnetism are still highly correlated but offset from the center by a certain amount. This suggests that the magnetism is linked to the topography, with the higher regions (top of the islands in the film) showing magnetic domains and the edges of the islands showing no magnetism [12].

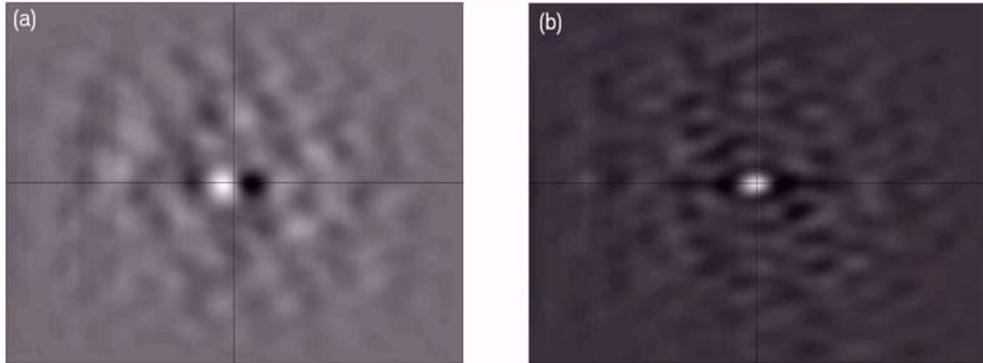


Fig. 7. Cross-correlation of topographic and magnetic images taken of LSMO film on NGO substrate. Compared to the cross-correlations of the crystal (in Fig. 4), the thin film shows the bright spot (high correlation) moving more towards the origin from (a) 301 K to (b) 315 K

Discussion and Conclusion

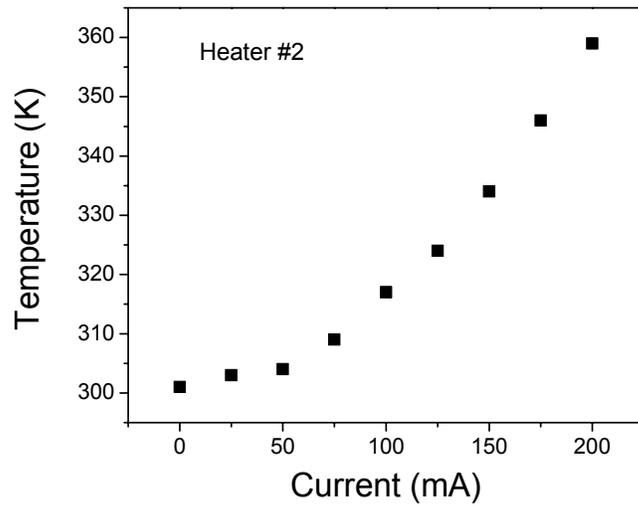
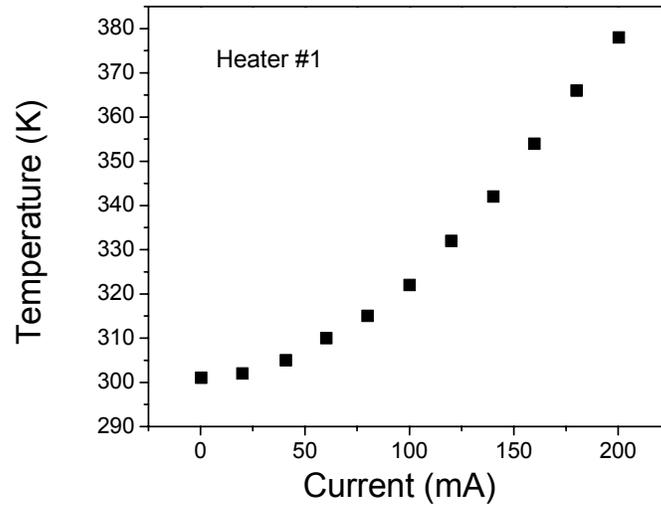
Overall, the two forms of LSMO examined in this study displayed a loss in well-defined magnetic domain patterns when subjected to heating. The interesting feature is that this loss of definition occurs well below the T_C of both the crystal and the film. Initially, with no heat applied, the crystal displays domain structures of about 500 nm in diameter. In Fig. 3, we see that while slowly approaching 315-320 K, the domains of the crystal tend to disappear. This temperature is well below its T_C of 345 K. The thin film in Fig. 6 shows smaller domains (about 100 nm in diameter) and exhibits gaps in between these regions. These gaps can be identified as possible charge-ordered regions formed

due to non-uniform strain [12]. The thin films reach a paramagnetic phase at an even lower temperature than that of the crystal, although the thin films bulk T_C is about 360 K (which is *higher* than the crystal's T_C). A possible explanation for the difference in the two is that a strain induced charge-ordered state is formed between the ferromagnetic islands in the thin film [13-15]. These charge-ordered regions result in the loss of ferromagnetism at the surface at much lower temperatures than the bulk T_C of the thin film. This observation is an important factor in determining the performance of manganite-based devices. To better understand why the domains of the crystal also disappear at a temperature slightly below T_C , we will investigate nearly strain free thin films of LSMO.

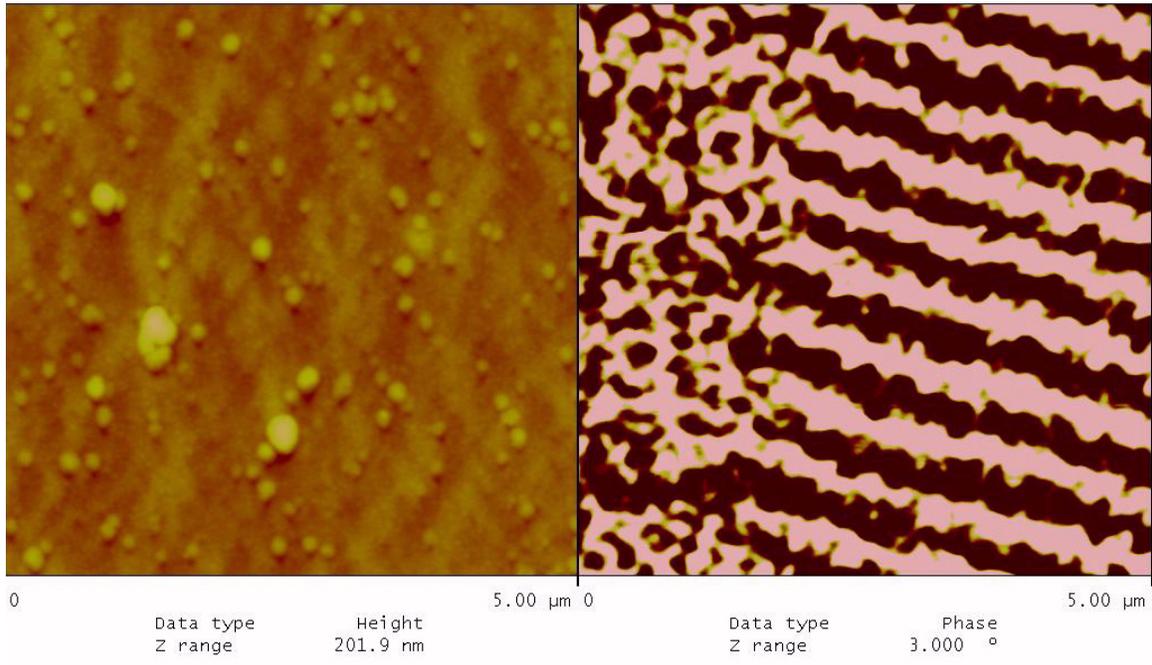
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Appendix A: Current vs. Temperature calibration curves for the two heating devices.



Appendix B: A sample image of the magnetic tape used to standardize the MFM.



References

- [1] J.M.D. Coey, M. Viret, and S. von Molnar, *Adv. in Phys.* **48**, 167 (1999).
- [2] Gary A. Prinz, *Science* **282**, 1660 (1998).
- [3] M. Uehara, S. Mori, C. H. Chen, S.-W. Cheong, *Nature* **399**, 560 (1999).
- [4] A. Asamitsu, Y. Tomioka, H. Kuwahara, Y. Tokura, *Nature* **388**, 50 (1997).
- [5] Peter Grünberg, *Physics Today*, **31**, (May 2001).
- [6] Neil Mathur and Peter Littlewood, *Physics Today* **56**, 25 (2003).
- [7] Y. Moritomo, H. Kuwahara, Y. Tomioka, and Y. Tokura, *Phys. Rev. B* **55**, 7549 (1997).
- [8] G. Hu and Y. Suzuki, *Phys. Rev. Lett.* **89**, 276601 (2002).
- [9] Yeong-Ah Soh, G. Aeppli, N. D. Mathur, and M. G. Blamire, *J. Appl. Phys.* **87**, 6743 (2000).
- [10] Qingyou Lu, Chun-Che Chen, and Alex de Lozanne, *Science* **276**, 2006 (1997).
- [11] Yeong-Ah Soh, G. Aeppli, N. D. Mathur, and M. G. Blamire, *Phys. Rev. B* **63**, 020402 (2001).
- [12] Amlan Biswas, M. Rajeswari, R. C. Srivastava, T. Venkatesan, R. L. Greene, Q. Lu, A. L. de Lozanne, and A. J. Millis, *Phys. Rev. B* **63**, 184424 (2001).
- [13] Yeong-Ah Soh, G. Aeppli, C.-Y. Kim, N. D. Mathur, and M. G. Blamire, *J. Appl. Phys.* **93**, 8322 (2003).
- [14] Liuwan Zhang, Casey Israel, Amlan Biswas, R. L. Greene, and Alex de Lozanne, *Science* **298**, 805 (2002).
- [15] Y. Murakami, J. H. Yoo, D. Shindo, T. Atou, and M. Kikuchi, *Nature* **423**, 965 (2003).