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Domain structure of the antiferromagnetic insulating state in $Nd_{0.5}Sr_{0.5}MnO_3$

I.O. Shklyarevskiy[a,*], A.B. Chizhik[a], S.L. Gnatchenko[a], P.J.M. van Bentum[b], P.C.M. Christianen[b], J.C. Maan[b], K.V. Kamenev[c], G. Balakrishnan[d], D. McK Paul[d]

[a] *B. Verkin Institute for Low Temperature Physics & Engineering, National Academy of Science of Ukraine, 47 Lenin Ave., 61164 Kharkov, Ukraine*

[b] *Research Institute for Materials, High Field Magnet Laboratory, University of Nijmegen, Toernooiveld 1, NL-6525 ED Nijmegen, Netherlands*

[c] *Department of Physics & Astronomy, The University of Edinburgh, Mayfield Road, Edinburg EH9 3JZ, UK*

[d] *Department of Physics, University of Warwick, Coventry CV4 7AL, England, UK*

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Abstract

Optical reflectivity studies of the ferromagnetic metal (FMM) to antiferromagnetic insulator (AFI) phase transition were performed on $Nd_{0.5}Sr_{0.5}MnO_3$ single crystals in a wide temperature and magnetic field range. The formation of a domain structure in the AFI state was observed. On the basis of the experimental results and symmetry analysis we conclude that these domains are crystal twins, confirming the symmetry lowering of the crystal at this phase transition. The twin domain structure of the AFI state in the $Nd_{0.5}Sr_{0.5}MnO_3$ is visible in reflected unpolarized light due to a different tilting of the surface of the domains. We find no evidence for intermediate FMM + AFI stripe phase.

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Perovskite manganese oxides with the basic formula $R_{1-x}A_xMnO_3$ (where R is a trivalent rare-earth ion, and A is a divalent alkaline ion) are of great interest because of their unusual magnetic and transport properties [1]. The most spectacular effect in the manganites is the colossal magnetoresistance observed in the vicinity of the Curie temperature. Moreover, these compounds exhibit interesting phenomena such as charge and orbital ordering, Jahn–Teller effect and magnetic or electric field induced phase transitions. In some of them, for example $Nd_{0.5}Sr_{0.5}MnO_3$, a temperature or magnetic field induced phase transition from antiferromagnetic insulating (AFI) to a ferromagnetic metallic (FMM) state takes place [2–4]. The nature of this phase transition is rather

*Corresponding author. Research Institute for Materials, High Field Magnet Laboratory, University of Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, Netherlands. Tel.: +31-024-36-53052; fax: +31-024-36-52440.

E-mail address: igors@sci.kun.nl (I.O. Shklyarevskiy).

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complicated and its understanding is yet to emerge.

The ferromagnetic ordering in $Nd_{0.5}Sr_{0.5}MnO_3$ occurs at $T_c \approx 255$ K. At T_{M-I} near 160 K a spontaneous first-order FMM–AFI phase transition takes place. This transition is accompanied by charge ordering, i.e. spatial ordering of the holes and corresponding orbital ordering of the Mn^{3+} and Mn^{4+} ions. In the temperature range $T < T_{M-I}$ the application of a magnetic field leads to melting of the charge ordering in $Nd_{0.5}Sr_{0.5}MnO_3$ and restores the FMM state [2]. The electroconductivity during this transition varies over two orders of magnitude and the magnetization changes by $2.5\,\mu_B$ per Mn atom [2]. In addition, the FMM–AFI phase transition in $Nd_{0.5}Sr_{0.5}MnO_3$ is accompanied by considerable changes in lattice parameters ($\sim 10^{-3}$–10^{-2}) and unit cell volume ($\sim 10^{-3}$) [2,5]. It was suggested in early studies [2] that the crystal has orthorhombic symmetry both in the FMM and AFI states. However, recent synchrotron and neutron diffraction investigations have shown that the FMM–AFI transition is accompanied by a lowering of the crystal symmetry from orthorhombic (space group Imma) to monoclinic (space group $P2_1/m$) [5,6].

In this paper we report on a detailed magneto-optical investigation of the recently discovered domain structure of $Nd_{0.5}Sr_{0.5}MnO_3$ in the AFI state, which can be simply imaged using unpolarized white light [7]. The explanation for the unusual optical contrast in the images is based on the exceptionally strong tilting of the crystal surface in the various twin domains.

The $Nd_{0.5}Sr_{0.5}MnO_3$ crystal was melt-grown by a standard floating zone method and had a cylindrical form of 3 mm in diameter. After X-ray analysis, which showed it to be a single crystal, the samples were cut out of the crystal pendicular to its growth direction with thicknesses varying from 0.5 to 3 mm. For our experiments the samples were polished with diamond paste. To remove the surface strain that occurs as a result of polishing they were annealed at 950°C during 20 h in air.

The samples were mounted in a continuous flow He-cryostat placed in a Bitter magnet providing magnetic fields up to 150 kOe. The image of the sample surface produced by a reflectance optical microscope using nonpolarized white light from a filament lamp was recorded with a CCD-camera connected to a video-recorder. The video images were subsequently analysed to determine the reflected light intensity in the various domains.

The magnetization measurements were carried out with a moving sample magnetometer in a variable temperature cryostat in fields up to 200 kOe.

It was found previously that there are distinct changes in the unpolarized reflected light intensity at the FMM–AFI phase transition in $Nd_{0.5}Sr_{0.5}MnO_3$ [7]. This effect provides the possibility of optical observation of the domain state that is formed at this first-order phase transition. With temperature decrease a spontaneous FMM–AFI phase transition takes place at $T \approx 154$ K, while temperature increase results in a reverse transition at $T \approx 162$ K. Thus, the first-order FMM–AFI phase transition is strongly hysteretic. Coexistence of FMM and AFI phases was observed only in a temperature region of about 2 K. The AFI phase has clearly visible domain structures that remain unchanged in the whole temperature range (20–154 K) at zero magnetic field. There are also no variations of the domain structure if a weak magnetic field is applied. Above a critical value of the field B_c, the AFI ordered state melts and the FMM state is restored. In Fig. 1, representative sequence of images illustrates the domain formation. In the image (a) the magnetic field is above B_c and a homogeneous FMM phase is observed. After reducing the field below the hysteretic B_c, domain growth is starting from various nucleation points. Image (b) gives a snapshot of this process. Finally, at still lower fields a uniform AFI phase is found (c) but with distinct optical contrast for the adjacent domains.

After each temperature or field-induced phase transition to the FMM state and subsequent reverse transition to the AFI state the newly formed domain structure can be completely different from the previous one. From this fact one can conclude that the domain structure in the AFI state emerges at the FMM–AFI phase transition and is not the visualization of a domain

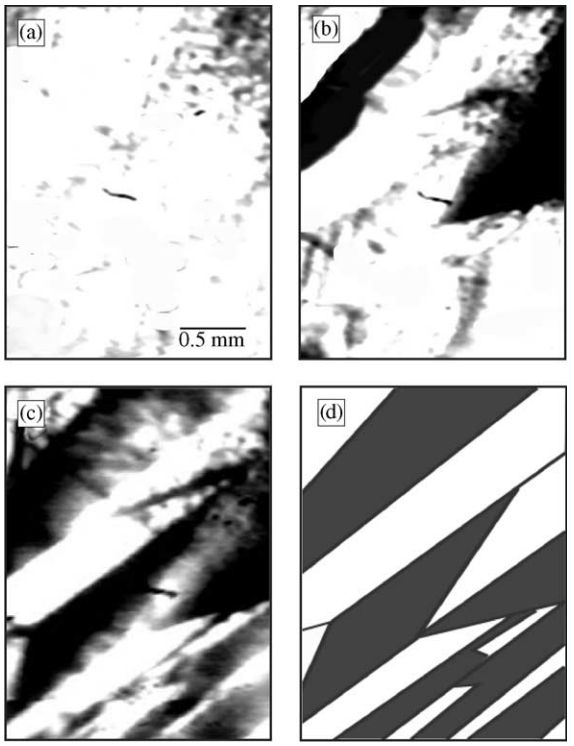

Fig. 1. Images of the field-induced FMM–AFI phase transtion in $Nd_{0.5}Sr_{0.5}MnO_3$. The temperature of the sample $T = 150$ K. External field: (a) 18 kOe, (b) 12 kOe, (c) 0 kOe, (d) Schematic representation of the AFI domain structure shown in (c).

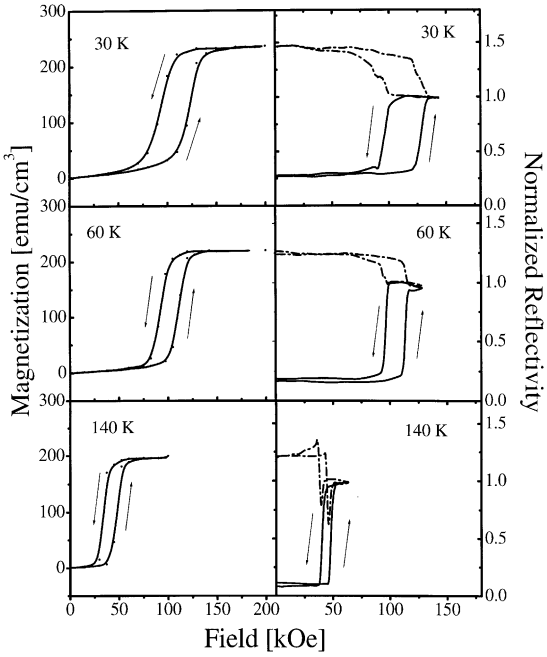

Fig. 2. Magnetization of the crystal versus magnetic field for resp. 30, 60 and 140 K (left panels). The right-hand panels indicate the field normalized optical reflectivity for bright (dash-dotted lines) and dark domains (solid curves) for the same temperatures.

structure that already exists in the FMM state. We did not find evidence for an intermediate stripe state with a periodical FMM–AFI domain structure in the present $Nd_{0.5}Sr_{0.5}MnO_3$ crystals.

In Fig. 2, we compare the magnetization data for the same crystal with the optical reflectivity as determined from the video images. The left-hand panels represent the magnetization $M(B)$ for three representative temperatures. The right-hand panels show the intensity of the reflected light from different domains, normalized to the intensity in the FMM state. The solid curves represent the intensity on the dark domains, where the reflectivity goes down in the AFI state. The dash-dotted lines give the reflectivity in the bright domains. As was mentioned above the domain structure in successive cycles is not always the same and the reflectivity on a specific part of the sample surface can first follow the solid line (dark) but then switch to the dotted line (bright). We want to emphasize that the absolute scale of the reflectivity depends strongly on the optical alignment.

From the experimental data we conclude that the change in the reflectivity for both types of domains coincides with the (hysteretic) AFI–FMM phase transition as seen in the magnetization. More specifically, the dark and bright domains are always formed at the same fields and temperatures; in other words they correspond to thermodynamically equivalent phases. It is reasonable to suggest that these domains are crystal twins emerging at the FMM–AFI phase transition accompanied by lowering of the crystal symmetry from orthorhombic to monoclinic.

The $B-T$ phase diagram of the $Nd_{0.5}Sr_{0.5}MnO_3$ based on the results of the optical studies is shown in Fig. 3. Transition field values in the phase diagram were determined from the $I(B)$ dependencies and correspond to the centre of the field interval of the mixed AFI + FMM phase. Open

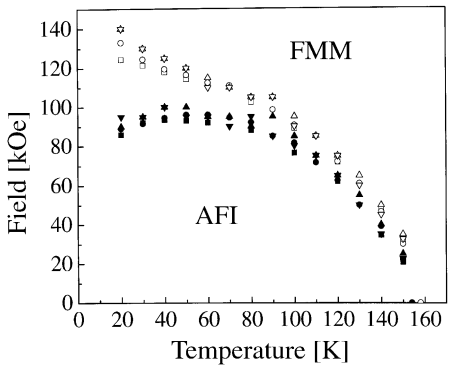

Fig. 3. $B-T$ phase diagram of $Nd_{0.5}Sr_{0.5}MnO_3$ based on both optical reflectivity and magnetization. Open symbols correspond to the phase transition from AFI to FMM state and closed symbols corresponds to the inverse transition from FMM to AFI state. Transition fields were determined from the spatially integrated $I(B)$ (circles), bright (up triangles) and dark (down triangles) domains as well as from the $M(B)$ dependencies (squares).

symbols correspond to the AFI–FMM phase transition observed with increasing magnetic field and closed symbols correspond to the reverse transition in decreasing field. The difference between the transition fields obtained for these two cases corresponds to the hysteresis in the $I(B)$ curves. It is clearly seen from Fig. 3 that the hysteresis decreases at higher temperatures. Transition field values obtained from the $M(B)$ dependencies are also indicated in the phase diagram and are in satisfactory agreement with the results of the optical experiment.

However, the origin of the optical contrast in the reflected unpolarized light at the crystal twins still remains to be explained. During visual observation of the domain structure in the AFI state we found that variation of the angle of incidence of the light on the sample resulted in changes of the optical contrast between domains. Dark domains transformed to bright or vice versa, and at a certain angle of incidence the optical contrast between domains disappeared. Moreover, the contrast between domains also vanished when using a microscope objective with sufficiently large aperture. This suggests that during crystal twin formation the surface of the sample becomes corrugated and the surfaces of the twins have a different tilt angle.

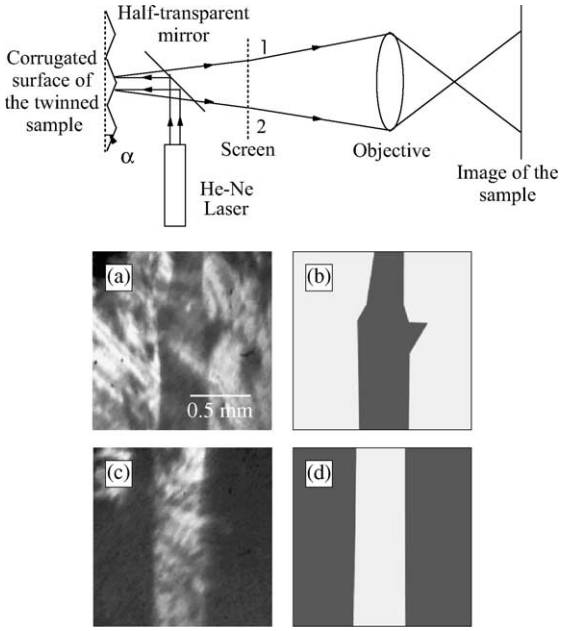

Fig. 4. Top: Optical setup to demonstrate the origin of the optical contrast between crystal twins. Bottom: (a) domain contrast with half of the reflected beam blocked. (c) Same as in (a) but with the other half of the reflected beam blocked. (b) and (d) Schematic representation of the domain structure shown in (a), respectively (c).

To check this suggestion the following experiment was performed. The basic scheme is presented on the upper panel of Fig. 4. A beam from a He–Ne laser illuminates an area of the sample. The reflected light is used to image the surface using a microscope objective. On an intermediate screen we determined the intensity distribution in the reflected beam. In the FMM phase we observed a single bright spot on the screen corresponding to the specular reflection of the optically polished surface. However, in the AFI domain phase we found two spots. If the optical alignment was such that the light of both spots was used in the image only weak optical contrast was seen between the various domains. However if we blocked half of the beam, using only one of the two bright spots to produce the image, we found a strong optical contrast. Blocking the other half of the beam led to an inversion of the contrast (Fig. 4(a) resp. (c)). Figs. 4(b) and (d) are the corresponding schematic representations of the domain structure. This

strongly suggests that the optical contrast between the domains is caused by a different angle of specular reflection for the two types of domains. The tilt angle of the domain surface was estimated to be $\alpha \approx 1^\circ$.

According to Refs. [5,6], the symmetry of $Nd_{0.5}Sr_{0.5}MnO_3$ changes from orthorhombic (space group Imma) in the FMM phase to monoclinic (space group $P2_1/m$) in the AFI state. The formation of a twin domain structure in the AFI state was previously observed in $Pr_{0.5}Sr_{0.5}MnO_3$ by electron diffraction techniques [8]. At the structural transition the crystal loses the following symmetry operations: two reflection planes $\bar{2}_x$, $\bar{2}_y$ and the rotation axes 2_x and 2_y (monoclinic axis is z-axis). The lost symmetry operations are indeed twinning operations. Two types of crystal twins can be formed at this phase transition. These twins differ by the sign of the shift deformation in the xy-plane and transform one into another by the twinning operations. It is easy to check, that in the present case two orthogonal solutions are possible with coherent domain walls arranged in the $x=0$ and $y=0$ crystal planes. This symmetry analysis is consistent with the optical observations showing two types of thermodynamically equivalent domains and preferential domain walls along the perpendicular a- and b-axis. The angle between two prevalent orientations of the domain walls is slightly less than 90° (70–80°) due to a small misalignment of the crystal cut with respect to the monoclinic c-axis as confirmed by X-ray analysis.

Finally, let us consider some properties of the intermediate state observed near the FMM–AFI phase transition. As it was mentioned above no magnetic intermediate state, i.e. thermodynamically stable stripe domain structure, forms at this transition. Usually, the absence of the intermediate state can be related to widening of the phase transition due to sample inhomogeneities. However, the present sample exhibits a sharp phase transition, and therefore the absence of the intermediate state cannot be attributed to sample inhomogeneities.

Apparently, a large energy of the interphase wall caused by the strong elastic strains is the main reason why the intermediate state does not form. These elastic strains have to arise in the vicinity of the interphase wall as a result of the dramatic changes ($\varepsilon \approx 1 \times 10^{-2}$) [4] in the crystal lattice parameters at the FMM–AFI phase transition. For the present crystals we estimate the domain wall energy $\sigma \approx 6 \times 10^5\,\mathrm{erg/cm^2}$, leading to a typical period D of the stripe domain structure of the magnetic intermediate state of $D \approx 2\,\mathrm{cm}$, which is much larger than the size of the sample. This explains the absence of the intermediate state at the FMM–AFI phase transition in our experiments.

In conclusion, we have shown that a twin domain structure arises in the AFI state of $Nd_{0.5}Sr_{0.5}MnO_3$, related to the lowering of the crystal symmetry at the FMM–AFI phase transition. The crystal twins have been imaged by unpolarized light. It was shown that the reason of the optical contrast between two types of twins is based on the local tilting of the surface of the twin domains. As far as we know, this is the first observation of a twin domain structure by this method. It was also shown that no periodic stripe phase forms at the FMM–AFI phase transition, which is related to the large energy of the interphase wall.

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