Direct Imaging of the First Order Spin Flop Transition in the Layered Manganite
\[ \text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7 \]

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The spin-flop transition in the antiferromagnetic layered manganite \[ \text{La}_{1.4}\text{Sr}_{1.6}\text{Mn}_2\text{O}_7 \] was studied using magnetization measurements and a high-resolution magneto-optical imaging technique. We report the direct observation of the formation of ferromagnetic domains appearing at the first order spin-flop transition. The magnetization process proceeds through nucleation of polarized domains at crystal defect sites and not through the expansion of polarized domains due to domain wall motion. A small magnetic hysteresis is caused by the difference between the mechanisms of nucleation and annihilation of domains in the mixed state. These results establish a direct link between the magnetic structure on the atomic scale as seen in neutron scattering and the macroscopic properties of the sample as seen in magnetization and conductivity measurements.

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The naturally layered manganites of composition $La_{2-2x}Sr_{1+z}Mn_2O_7$ have attracted much recent interest since in addition to the phenomenon of colossal magneto-resistance (CMR) they exhibit a variety of coupled magnetic, electronic and structural groundstates. Depending on the doping level, $x$, the interplay of superexchange and double exchange interactions between the Mn-moments gives rise to competing antiferromagnetic and ferromagnetic spin arrangements. Recent magnetization and neutron scattering experiments on the $x = 0.30$ compound, $La_{1.4}Sr_{1.6}Mn_2O_7$, have shown that for this doping level the material is a highly anisotropic type A antiferromagnet in which the ferromagnetic MnO$_2$ bi-layers are stacked antiferromagnetically along the $c$-axis with the Mn-moments oriented along the $c$-axis. The application of a magnetic field along the antiferromagnetic axis induces a spin-flop transition between the antiferromagnetic and the polarized states. The unique feature of this material is that due to the strong coupling of the electronic and magnetic degrees of freedom this transition is accompanied by a very large, anisotropic magnetoresistance. In addition, the spontaneous formation of ferromagnetic bubble domains has recently been reported.

Using magnetization measurements and magneto-optical imaging we performed a detailed study of the magnetization process in single crystal $La_{1.4}Sr_{1.6}Mn_2O_7$. Magneto-optical imaging allows for the direct observation of the formation of magnetic domains and provides details of the local magnetization process which are unattainable from bulk magnetization and/or conductivity measurements. A first order spin-flop (metamagnetic) transition between the antiferromagnetic and polarized states is observed at a field of about 1.1 kOe applied along the $c$-axis. We find direct evidence for a mixed state of coexisting spin-flop and antiferromagnetic domains. The magnetization process in the mixed state proceeds through nucleation of spin-flop domains at crystal defect sites. The expansion of polarized domains through domain wall motion in the MnO$_2$-planes appears to be of minor significance, especially in the onset region of the transition. However, we observe continuous domain growth along the $c$-axis, that is, normal to the MnO$_2$-planes. A small
magnetic hysteresis, consistent with the first order nature of the transition, is caused by the difference between the domain nucleation and annihilation processes.

Single crystals of La$_{1.6}$Sr$_{1.6}$Mn$_2$O$_7$ were melt-grown in flowing 20% O$_2$ (balance Ar) in a floating-zone optical image furnace. The sample used in this study is shaped like an irregular plate (see Fig. 2) with lateral dimension of roughly 800 x 600 µm$^2$ and thickness (along c) of 100 µm. The magneto-optical images were obtained employing a high-resolution magneto-optical imaging technique.

The field dependence of the c-axis magnetization at 20 K is shown in Fig. 1 for increasing ($M_{in}$) and decreasing ($M_{dc}$) field. The magnetization stays essentially zero (region “I”) until a critical field of $H_{SF} = 1.1$ kOe is reached at which a steep, initially linear rise of the magnetization occurs. The magnetization saturates near $H_s = 4.8$ kOe at a value of 72 emu/g. This magnetization behavior is indicative of a first order spin-flop transition at $H_{SF}$. In the field region of 1.1 kOe to 4.8 kOe (region “II”) a mixed state of coexisting antiferromagnetic and polarized domains is expected due to demagnetization effects. The spin arrangements in the various phases are indicated in the figure. Also shown is the magnetization hysteresis $\Delta M = M_{dc} - M_{in}$. The magnetization process is essentially reversible, however, clearly resolved peaks of $\Delta M$ are observed near $H_{SF}$ and at $H_s$. As discussed in more detail below these are caused by the nucleation of polarized and antiferromagnetic domains, respectively.

Fig. 2 shows magneto-optical images taken at a temperature of 21 K on the c- face of the crystal with the external magnetic field applied parallel to the c-axis. Bright contrast in these images represents high local values of the normal component of the magnetic induction, $B_z$. At applied fields below $H_{SF}$ (Fig. 2a) the sample is in the antiferromagnetic state and correspondingly there is essentially no magnetic contrast in the bulk of the crystal. With increasing field, polarized domains seen as bright spots, nucleate (see Fig. 2b, c). These images show directly the coexistence of polarized and antiferromagnetic domains as
expected for the mixed state of the first order spin-flop transition. With further increasing field, domains coalesce to form extended domain structures that exhibit clear striation along two orthogonal directions that coincide with the crystallographic (100) and (010) directions (Fig. 2d). In addition, the pattern of domains is exactly the same on successive field sweeps and is independent of an applied in-plane field. These results indicate that the polarized domains nucleate at defect sites in the underlying crystal structure.

In contrast to the behavior seen for example in the uniaxial antiferromagnets MnF$_2$ and FeCl$_2$ near the spin-flop the expansion of polarized domains through continuous domain wall motion is not observed. Instead, the magnetization process proceeds through the nucleation of new domains. Using the available literature data for the in-plane exchange energy, J$_{1} = 3.6 \times 10^8$ erg/cm$^3$, and anisotropy energy, K = 2 x 10$^6$ erg/cm$^3$, we estimate a domain wall thickness $\delta \approx \pi a (J_{1}/2K)^{1/2}$ of about 30 lattice constants, a. Such a small value implies that these domain walls can be pinned very effectively by imperfections in the underlying crystal structure which prohibits the occurrence of domain wall motion.

Figs. 2c and e show the domain pattern at a field of 1165 Oe in increasing and decreasing field, respectively. A clear hysteresis in the domain pattern is visible: on the returning branch of the hysteresis loop the magnetic structure is characterized by a much finer divided domain pattern. This is shown quantitatively in Fig. 3 which displays the field dependence of $(B_z - H)$ in the position labeled “1” in Fig. 2c. At a field of 1120 Oe on increasing field a discontinuous jump in $(B_z - H)$ by about 700 G is observed consistent with the first order nature of the spin-flop transition. On decreasing applied fields an “inverted” hysteresis is observed: the values for $(B_z - H)$ are smaller than those on the increasing field. At $H_{sp}$ the two branches intersect and the decreasing branch reaches zero at 1070 G. The almost smooth disappearance of $(B_z - H)$ is also observed for a “short” hysteresis loop up to 1165 Oe. The smooth disappearance of the spin-flop domains has previously been observed at the spin-flop transition of FeCl$_2$ and has been attributed to the shrinking of the domains along the field direction as the field is decreased (see below).
Also included in Fig. 3 is the field dependence of \((B_z - H)\) when averaged over the sample surface. This quantity is directly proportional to the global magnetization of the sample (Fig. 1), and the linear rise starting near 1100 Oe is in good agreement with Fig. 1. A value of \((B_z - H)\) of about 700 G in an applied field of 2350 Oe can be converted into a magnetization value of 35 emu/g, in good agreement with the magnetometer measurement of 30 emu/g. However, the field resolution of the images is not sufficient to discern the small hysteresis \(\Delta M/M = 10^{-3}\) seen in magnetization.

Fig. 4 shows magneto-optical images taken at 21 K on an edge face containing the ac-axes. For these images the external magnetic field is applied parallel to the c-axis, that is, parallel to the imaged surface. The location of the imaged surface is indicated by the dark line in Fig. 2c. In this geometry the imaged contrast arises from the stray fields transverse to the magnetization direction. These stray fields are positive (bright contrast) near the north pole and negative (dark contrast) near the south pole of the magnetized domain resulting in the characteristic bright-dark pairing. In low fields (Fig. 4a) there are in addition to the large domain associated with the second phase inclusion (labeled “2” in Fig. 4d) several small domains distributed throughout the thickness of the sample. On increasing field new domains nucleate (Fig. 4b) and existing domains grow along the field direction (see label “4” in Figs. 4a, b and c) until in elevated fields (Fig. 4d) a dense pattern of domains stretches from the bottom to the top surface. In particular, the domain labeled “4” evolves into the striation labeled “4” in Fig. 2d. On decreasing field a hysteresis similar to the results in Fig. 2 is observed (see Fig. 4c and d) that is characterized by a denser pattern of longer domains.

In conclusion using magnetization measurements and magneto-optical imaging we performed a detailed study of spin-flop transition in single crystal La_{1.4}Sr_{1.6}Mn_2O_7. Direct evidence for a mixed state of coexisting spin-flop and antiferromagnetic domains is presented. The magnetization process in the mixed state proceeds through nucleation of spin-flop domains at crystal defect sites. The expansion of polarized domains through...
domain wall motion in the MnO$_2$-planes appears to be of minor significance. However, we observe continuous domain growth along the c-axis, that is, normal to the MnO$_2$-planes. A small magnetic hysteresis, consistent with the first order nature of the transition, is caused by the difference between the domain nucleation and annihilation processes.

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References


Figure captions

Fig. 1 Field dependence of the magnetization (left scale) and of the magnetization hysteresis (right scale) at 20 K. The spin arrangements in the different regimes are shown schematically.

Fig. 2 Magneto-optical images at 21 K in various fields applied parallel to the c-axis. Label “1” in frame c marks the location of the local magnetic induction curves (Fig. 3). The dark line indicates the polished end face imaged in Fig. 4. Labels “2”, “3” and “4” in frame d mark non-magnetic (La,Sr)$_2$MnO$_4$ inclusions, a cavity and magnetic striation referred to in Fig. 4, respectively.

Fig. 3 Field dependence at 21 K of the local value of $(B_z - H)$ evaluated in position “1” (see Fig. 2c) for increasing and decreasing applied fields. Also included is the field dependence of $(B_z - H)$ when averaged over the sample surface. The inset shows the behavior near the spin-flop transition on expanded scales.

Fig. 4 Magneto-optical images at 21 K taken on the end-face marked in Fig. 2c. Labels “2” and “4” refer to the magnetic (La,Sr)$_2$MnO$_4$ inclusion and the spin-flop domains that evolves into the striation (see Fig. 2d).
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