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High magnetic-field study of the magnetization of layered manganite $Nd_{2-2x}Sr_{1+2x}Mn_2O_7$ single crystals

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Abstract

The magnetization of $Nd_{2-2x}Sr_{1+2x}Mn_2O_7$ layered manganite single crystals (with $x = 0.5, 0.4, 0.25$) has been studied using high magnetic fields up to 30 T at 4.2 K. In these compounds, which are antiferromagnetic at low temperatures, induced metamagnetic transitions have been observed. These transitions become very sharp for doping level values in the range $x = 0.25$–0.4, reaching saturation-like behavior and displaying a large hysteresis. Maximum field magnetization is much lower than expected for the full ferromagnetic configuration of the Nd and Mn moments, and a large high field susceptibility is observed beyond the transition. The observed magnetization at maximum field within the *ab*-plane is lower (up to a 17%) than along the *c* direction. However, the value of the critical magnetic field of the transition is in all the cases larger for the magnetization within *ab* than along *c*. These transitions have been ascribed to spin-flipping within the Mn moment sublattice.

Keywords: Layered manganites; Metamagnetic transitions; Colossal magnetoresistance

1. Introduction

The interest for understanding the physics underlying the colossal magnetoresistance (MR) phenomena in perovskite manganites has given a strong impetus to the study of these compounds and their layered variants. The study of layered manganites $(R,M)_{n+1}Mn_nO_{3n+1}$ (R = rare-earth ion and M = divalent cation) offers the opportunity of observing the interplay between spin, charge and lattice in low dimensions, versus the three-dimensional behavior of perovskite manganites. For $n = 2$ the crystal structure is built up of perovskite bilayers of corner-linked MnO_6 octahedra forming infinite sheets, and separated by a (R,Sr)O layer along the *c*-axis. The resultant tetragonal $Sr_3Ti_2O_7$ type-structure is very anisotropic and the study of single-crystalline samples is essential for a deep understanding of the physical properties.

Magnetoresistive effects are, in many manganite systems, associated with the existence of a ferromagnetic ground state induced by the application of magnetic field. The competition between ferromagnetic (FM) and antiferromagnetic (AF) interactions is very strong and in layered manganites

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a FM ground state is observed only in the La–Sr compounds ($La_{2-2x}Sr_{1+2x}Mn_2O_7$) with a doping level in the range $0.3 < x < 0.4$ [1]. For this reason this series has been the most studied layered bulk system. Colossal MR in layered manganites was for the first time observed in La–Sr with $x = 0.4$ [2], and intrinsic low-field MR in La–Sr with $x = 0.3$ single-crystals, which was attributed to an interlayer tunneling mechanism [3].

Nevertheless, the possibility of stabilizing FM does not seem to be a prerequisite for the observation of enhanced MR effects, and low field MR has also been reported in AF Nd–Sr with $x = 0.25$ single-crystals [4] and Sm–Sr with $x = 0.2$ polycrystals [5]. The charge ordered state tends to be more stable close to the doping level $x = 0.5$. In fact, a charge ordered state has been reported in La–Sr with $x = 0.5$ [6]. Recent high field measurements on this compound show the existence of magnetic hysteretic transitions associated with irreversible changes in the MR, that have been attributed to field induced changes in the spin and orbital structure [7]. In this paper, we present a high-field magnetization study performed on single crystals of the layered Nd–Sr compounds, which are AF at low temperatures.

2. Experimental

The crystals were grown using the floating-zone technique with an IR image furnace (NEC, SC-N35HD). The growth rate used was 7 to 8 mm h^{-1} with feed and seed rods rotating in opposite directions at 30 r.p.m. The crystals were oriented and cut into a cube (of approximate dimensions $1 \times 1 \times 1\,mm^3$) such that one of the '$z$' axes of the cube was along the crystallographic c-axis and the 'x' and 'y'-axis were contained within the ab plane.

The magnetization and magnetotransport measurements were performed using the long-pulse magnetic field facility at the University of Zaragoza-CSIC (see Ref. [8] for a detailed description of this installation). The pulse, 2.5 s long, can reach a maximum field of 31 T having a raising time of 0.15 s. The signal due to the induced variation in the magnetization of the sample is detected using compensated pick-up coils. The sample is extracted at the tail of the decaying field, in order to determine the existing remanence. The measurements have been performed at 4.2 K, with the magnetic field applied parallel to the c direction (M_c) and perpendicular to it (M_{ab}). The reproducibility of the observed magnetic behavior has been checked in different samples for a given composition.

3. Results and discussion

X-ray powder diffraction analysis of the compounds showed that the crystallographic structure was consistent with a body-centered tetragonal unit cell (I4/mmm; $Z = 2$). Impurity lines which has been ascribed to the presence of intergrowths were detected for one of the samples, $Nd_{1.5}Sr_{1.5}Mn_2O_7$, [4].

The magnetization isotherms M_{ab} and M_c obtained at 4.2 K for the $Nd_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.5$, 0.4 and 0.25) single crystals are shown in Figs. 1(a), (b) and (c), respectively. These compounds present AF order with T_N in the range 100–200 K, so that the applied field tends to induce the ferromagnetic state. A similar qualitative behavior is observed in compounds with $x < 0.5$ (Figs. 1(b) and (c)), where both the magnetization curves along c and within the ab-plane display a sharp metamagnetic transition at field values higher than 10 T. Saturation-like behavior is reached, but the maximum field magnetization is much lower than the value expected for a perfect ferromagnetic configuration of the manganese and rare-earth moments. As an example, in Nd–Sr with $x = 0.4$, M_c at 30 T reaches only 93% of the full FM state magnetization. This fact is connected with the very large susceptibility value observed in the isotherms beyond the metamagnetic transition (around 0.03 μ_B/T for M_c in Nd–Sr with $x = 0.4$). A large hysteresis is observed between the field-increasing and field-decreasing processes. In the field-decreasing process the system can follow different metastable configurations, as has been measured for M_c, with either one or two transitions detected in different measurements. As is well known, this metamagnetic hysteretical behavior is characteristic of a first-order phase transition. A small spontaneous FM contribution is observed

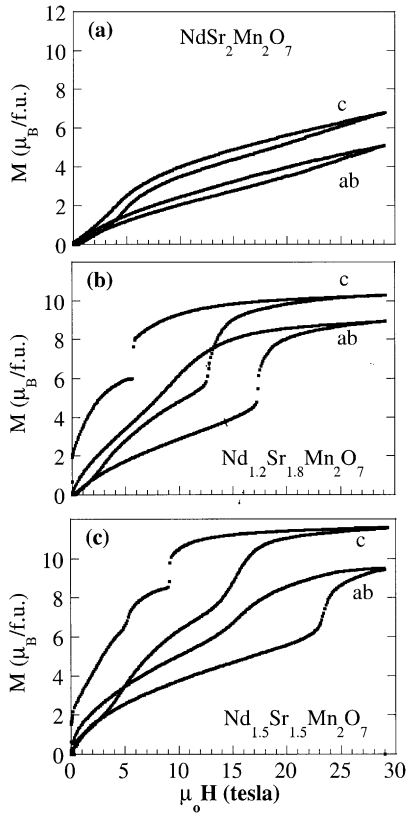

Fig. 1. Magnetization isotherms measured on single crystals of the layered Nd–Sr series along the *c*-direction and within the *ab*-plane at 4.2 K.

at low fields in the Nd–Sr with $x = 0.25$ sample (Fig. 1(c)). This contribution has been ascribed to the presence of FM intergrowths ordering at 270 K [4], not present in the remaining samples.

Note that in the $x < 0.5$ compounds the critical field value, at which the sharp magnetization jump is observed, is several teslas larger in the *ab*-plane isotherms than in the isotherms along *c*. However, the magnetization reached after the transition is lower within the plane than along the *c*-axis. This fact is probably related to the existence of a smooth transition that is induced when the field (below 5 T) is applied along the *c*-axis, which is not observed in the basal plane magnetization. These features make evident the complex anisotropic magnetic behavior of these layered manganite systems.

Decreasing the doping level from $x = 0.4$ to 0.25 leads to an increase of the critical field of the metamagnetic transitions (see Figs. 1(b) and (c)). This variation can be due to the modification of the exchange interactions of the Mn sublattice as a consequence of the variation in the lattice parameters of the different compounds. Nevertheless, the influence of a change in the effective rare-earth anisotropy can also play a role: in this structure two sites with different symmetry are available for the Sr^{2+} and Nd^{3+} cations, and depending on the composition, a different relative partial ordering is obtained [9].

In order to get a deeper insight on the nature of the observed metamagnetic transitions, preliminary measurements of the magnetotransport behavior of $Nd_{1.5}Sr_{1.5}Mn_2O_7$ have been performed [10]. These have revealed the existence of a large decrease in the MR, correlated with the low and high field metamagnetic transitions. Taking into account that in manganites the charge carriers mobility depends on the relative orientation of the Mn moments, we conclude that these moments are involved in the observed spin-flip/spin-flop metamagnetic transitions.

A qualitative change is observed in the low-temperature magnetic behavior between $x = 0.4$ and 0.5 doping levels (Figs. 1(b) and (a)). The sharp metamagnetic transitions occurring at high fields in the $x = 0.4$ compounds are very smooth than in the $x = 0.5$ compounds. Hysteretical behavior is induced but the saturated-like state is not reached even at the highest field value. This different behavior suggests a reinforcement of the AF interactions at this doping level, which could be due to the above-mentioned tendency to charge-order.

Finally, we would like to note that we have observed large jumps in the magnetorestriction correlated to the sharp high magnetic field-induced metamagnetic transitions. One can think that this effect, similar to the one observed in related manganite compounds, could be due to the existence of a field-induced crystallographic transition (with symmetry lowering) [11]. Nevertheless, in compounds where such a transition can be induced, the crystallographic transition is observed to occur spontaneously with temperature lowering, which is not the case in the present systems [12].

4. Conclusions

The low temperature, high field magnetization of single crystals of the layered manganites $Nd_{2-2x}Sr_{1+2x}Mn_2O_7$ is presented. The magnetization displays very anisotropic behavior between in-plane and out-of-plane measurements. The possibility of using high magnetic fields has allowed observing spin-flip/spin-flop metamagnetic transitions on the AF Nd–Sr compounds, which are induced at fields higher than 10 T. They have been attributed to transitions of the manganese moment configuration. The observed large high field magnetic susceptibility beyond the transitions in the $x = 0.4$–0.25 compounds is probably associated with the forced rotation of the moments of the rare-earth sublattice. The AF interactions are sharply reinforced at the doping level $x = 0.5$, which could be due to a tendency to stabilize a charge ordered state at this composition. Both the facts confirm the importance of using high magnetic fields in the understanding of the exchange interactions and magnetotransport in these layered systems. To understand the influence of the effective rare earth anisotropy, an investigation of the magnetic behavior of compounds with different rare earth substitution would be necessary.

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