Staircase effect in metamagnetic transitions of charge and
orbitally ordered manganites.

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Abstract

This paper reports on peculiar metamagnetic transitions which take place
in antiferromagnetic, charge and orbitally ordered (AFCOO) manganites. At
very low temperatures, the virgin magnetization curves of some of these com-
pounds exhibit several, sharp steps giving rise to a staircase-like shape. This
staircase effect is shown to be sensitive to various experimental parameters.
Several potential interpretations of the staircase effect are discussed in rela-
tion to this set of results. A martensitic-like scenario, involving a leading role
of the structural distortions associated to the collapse of the orbital ordering,
is found to be the most plausible interpretation.
I. INTRODUCTION

Mixed-valent manganites of formulation $\text{RE}_{1-x}\text{AE}_x\text{MnO}_3$ (RE being a trivalent rare-earth ion, and AE a divalent alkaline-earth ion) exhibit a great variety of fascinating properties, such as charge ordering (real-space ordering of the $\text{Mn}^{3+}$ and $\text{Mn}^{4+}$ species) and colossal magnetoresistance (collapse of the resistivity by several orders of magnitude under application of an external magnetic field).\(^1\) Charge ordering is often accompanied by a long-range ordering of the $\text{Mn}^{3+} e_g$ orbitals, leading to prominent structural distortions. In $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ compounds with $x \approx 0.5$, the small A-site cationic radius and the $\text{Mn}^{4+}/\text{Mn}^{3+}$ ratio close to 1 contribute to the setting up of a particular ground state, which is both charge and orbitally ordered, insulating and antiferromagnetic of CE type (hereafter referred to as AFCOO). Application of magnetic fields can affect the subtle energy balance in these systems, and trigger the development of a ferromagnetic, conducting state (hereafter referred to as F). This process is at the origin of the most spectacular effects of colossal magnetoresistance in manganites. However, very high magnetic fields are required to melt the robust AFCOO state present for $x = 0.5$ (about 25 T at 4 K).\(^2\) For compositions shifted towards $x = 0.3$, lower fields are needed (e.g., about 5 T for $x \approx 0.35$).\(^3\) Mn-site substitutions can also weaken the AFCOO state,\(^4\) making it susceptible to undergo an AFCOO-F transition at low-T under moderate fields.

Recently, we observed very intriguing metamagnetic transitions in such $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{1-y}\text{M}_y\text{O}_3$ compounds, $\text{M}$ being a magnetic or nonmagnetic cation.\(^5,6\) In some cases, the virgin magnetization curve at low-T indeed displays successive, abrupt steps as the field is increased. This staircase-like shape of the $M(H)$ curves disappears as $T$ is increased above about 10 K. Similar features were observed in resistivity and specific heat measurements.\(^5\)

In this paper, we investigated by isothermal magnetization loops several aspects of this staircase effect: sample dependence, role of temperature, cycling effects, mode of field variation, role of microstructure, type of disorder affecting the AFCOO stabil-
ity. Most of the experiments were carried out on a set of samples of the composition \( \text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3 \). Complementary measurements were also performed at very low temperatures in \( \text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3 \) compounds with \( 0.35 \leq x \leq 0.40 \).

II. EXPERIMENTAL DETAILS

All the samples used in this study are ceramics, except one crystal which was grown in a mirror furnace. The \( \text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3 \) and \( \text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3 \) ceramic samples were prepared by conventional solid-state reaction. Stoichiometric mixtures of the oxides \( \text{Pr}_6\text{O}_{11} \), \( \text{CaO} \), \( \text{MnO}_2 \) and \( \text{Ga}_2\text{O}_3 \) were intimately ground and the powders, first heated at \( 1000 \, ^\circ\text{C} \), were pressed in the form of bars. The sintering was made at \( 1200\, ^\circ\text{C} \) and at \( 1500\, ^\circ\text{C} \) for 12 h, then the bars were slowly cooled to \( 800\, ^\circ\text{C} \) at \( 5\, ^\circ\text{C} /\text{h} \), before being quenched to room temperature. A \( \text{Pr}_{0.63}\text{Ca}_{0.37}\text{MnO}_3 \) crystal was cut out of the central part of a several-cm-long specimen grown in a mirror furnace using a floating-zone method. Physical characterizations of this crystal were previously reported. The purity of all samples was attested to by X-ray powder diffraction and electron diffraction studies. Energy-dispersive X-ray microanalyses yielded cationic compositions in agreement with the nominal ones, within the accuracy of this technique. Electron diffraction investigations also revealed the existence of twinning domains in all samples, including the single crystal.

The magnetic measurements were carried out by means of a Quantum Design extraction magnetometer, with fields up to 9 T and temperatures down to 2.5 K. All hysteresis loops were recorded after a zero-field cooling from room temperature. It must be noted that room temperature (RT) is larger than all transition temperatures -corresponding to spin, charge or orbital orderings- that can be encountered in these materials. Even for the unsubstituted, half-doped \( \text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3 \), the charge and orbital ordering takes place at \( T_{CO} \approx 250 \, \text{K} \), well below RT. The hysteresis loops were registered according to the following procedure: The magnetic field was increased from 0 to 9 T, and then decreased down to 0 T, with equally spaced values (0.25 T in most cases); a waiting time is imposed between the end
of the field installation and the beginning of the measurements (1 min in most cases); then, three measurements are successively recorded (all shown in the figures). Complementary experiments were carried out to investigate the influence of field spacing and waiting time values.

For $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$, that is the compound used for most of the experiments, we studied four ceramic samples coming from the same batch, and having nearly the same mass and shape. The first measurement recorded on each of these four samples -hereafter labelled as A, B, C and D- was an hysteresis loop at 5 K. This allowed us to properly address the question of reproducibility, by considering only samples being in a pure virgin state, i.e., without any previous application of high fields at low temperature.

III. RESULTS

Figure 1 shows hysteresis loops, recorded at 5 K, in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$ and $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ [hereafter denoted as (PrCa50)Ga5% and PrCa50, respectively]. In PrCa50, one observes a linear, reversible curve up to 9 T, consistent with the fact that this compound stays in a pure AFCOO state within this field range. In (PrCa50)Ga5%, the field-increasing branch of the loop also exhibits a linear behavior for magnetic fields lower than about 4 T. However, the slope is significantly larger than in PrCa50. This was confirmed by low-field ac-susceptibility curves, which also revealed the existence of a cusp around 50 K in the Mn-site substituted compounds. Such a cusp suggests that the introduced disorder on the Mn-sublattice allows the development of competing ferromagnetic interactions within the antiferromagnetic matrix. These results point to the setting of a very weakened AFCOO groundstate in (PrCa50)Ga5% as compared to PrCa50. For fields larger than 4 T, the curve of (PrCa50)Ga5% (sample A) exhibits three large jumps, separated by almost flat plateaus. Approaching 9 T, the magnetization undergoes a small increase from the last plateau, leading to a value of $3.25 \mu_B / \text{f.u.}$. Since the expected magnetization for full polarization of the Mn spins in (PrCa50)Ga5% is $3.3 \mu_B / \text{f.u.}$, the sample under 9 T is virtually in a saturated
magnetic state. As the field is decreased, one observes a rather flat curve down to about 2 T, followed by a rapid drop of magnetization yielding a remanent value of about 0.15 $\mu_B$/f.u. The inset of Fig. 1 displays following field-scans recorded on this sample, first by extending the field decrease down to -9 T, and then by re-increasing it up to 9 T. The curve between 0 and -9 T is symmetrical to the reverse leg of the first quadrant. Furthermore, the curve recorded when increasing the field from -9 T to 9 T is almost superimposed on the one obtained when decreasing from 9 T to -9 T. After the first application of 9 T, the sample thus seems to behave as a bulk, reversible ferromagnet.

Before going further, it is essential to check the reproducibility of the staircase-like behavior. Hysteresis loops at 5 K were recorded on four virgin samples belonging to the same batch. For the sake of clarity, figure 2 just shows the field-increasing parts of the loops. First, one can point out the remarkable superimposition of the linear regimes found between 0 and 4 T, attesting to the good homogeneity of the cationic composition within this set of samples. For each of them, three large steps occur within the field range 4-9 T, and the magnetization reaches about 3.25 $\mu_B$/f.u. under 9 T in all cases. However, it is clear that the characteristic fields associated with the steps slightly differ from sample to sample. A significant scatter exists also about the magnetization values corresponding to each plateau, especially for the first one. These results demonstrate that the staircase effect only exhibits a semi-quantitative reproducibility.

Figure 3 displays hysteresis loops recorded on sample B at 5, 7.5 and 10 K. All curves have nearly the same initial slope, saturation value and remanent magnetization, but the steps have completely disappeared for $T = 10$ K. At this temperature, the metamagnetic transition rather has a smooth, S-shape centered around 5 T. For $T \leq 5$ K, the transition exhibits several steps (observed down to our lowest temperature $T = 2.5$ K). The curve at 7.5 K well illustrates the crossover in temperature. As compared to lower temperatures, the curve is smoothed out from both sides of the field range, leaving just one step in the middle of the transition. This behavior is basically different from a simple broadening of the steps seen at 5 K. One can also notice that the curves with steps seem to “zigzag” around the one
at 10 K, a behavior suggesting that this last curve might be representative of an underlying equilibrium $M(H)$ curve at low-$T$.

In our first report of the staircase effect,$^5$ we already mentioned the existence of some variations in the pattern of steps as the sample is cycled between low-$T$ and RT. To get a direct insight into this point, we have successively recorded on sample C five hysteresis loops at 5 K, each of them starting from a zero-field cooled state. The results are shown in Fig. 4. For the first three loops, the step structure is well reproducible. One just observes small variations in the height of the second plateau and in the third step field ($H_{S3}$). In contrast, a marked evolution takes place in between the third and fourth runs: One observes clear upward shifts in the step fields $H_{S1}$ and $H_{S2}$, while the third step is replaced by a smooth tail. Then, the fifth loop remains well superimposed on the preceding one. Along these five runs, the initial slope and remanent magnetization remain constant, whereas there is a systematic, small decrease of the maximum magnetization. All these features have been observed in other samples, except that “switches”, such as that seen in Fig. 4 between runs 3 and 4, can occur sooner or later.

All previously shown data were recorded by using a field spacing ($f_s$) equal to 2500 Oe and a waiting time ($t_w$) equal to 1 min. Figure 5 shows three loops successively recorded on sample C at $T = 5$ K, following different procedures: (1) $f_s = 2500$ Oe with $t_w = 1$ min (run 5 of Fig. 4); (2) $f_s = 1000$ Oe with $t_w = 1$ min; (3) $f_s = 2500$ Oe with $t_w = 5$ min. The last two runs lead to the same average sweep rate (35 mT / min), including field installation, waiting time and data acquisition. For a constant waiting time (1 min), one observes that using smaller field spacings pushes the steps to higher fields. These shifts in the $H_S$ values are accompanied by upward shifts of the corresponding plateaus. Run 3 registered with $f_s = 2500$ Oe and $t_w = 5$ min yields a curve nearly superimposed on that of run 1 obtained for $f_s = 2500$ Oe and $t_w = 1$ min. Thus one can conclude that: (i) the shifts in $H_S$ observed between runs 1 and 2 can actually be ascribed to the variation in $f_s$, and not just to a cycling effect (as in Fig. 4); (ii) the impacts of increasing the average sweep rate via $f_s$ or $t_w$ are completely different. It turns out that the field spacing is actually the
relevant parameter at the origin of the difference between run 1 and 2.

Up to now, the staircase effect has only been observed in Mn-site substituted PrCa50 samples. One can wonder whether this property is related to the weakening of the AFCOO state, or more specifically to a disordering of the Mn sublattice. Thus, we re-investigated $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ compounds with $x$ values yielding AFCOO states able to be destabilized under moderate fields. We studied two ceramic samples with $x = 0.40$ and $x = 0.35$, as well as a crystal with $x = 0.37$. For all of them, smooth metamagnetic transitions were observed for temperatures down to 5 K. At 2.5 K, in contrast, the hysteresis loops display very steep transitions, as shown in Fig. 6. The ceramics with $x = 0.35$ and $x = 0.40$ present a unique, large step at the transition, while a multisteps structure takes place for the crystal with $x = 0.37$. This staircase effect is exactly the same as that encountered in Mn-site substituted compounds with $x = 0.50$. This is an important piece of information which demonstrates that the multisteps behavior is not basically related to the disordering of the Mn-sublattice induced by magnetic or nonmagnetic foreign cations. In other respects, one can wonder why the PrCa37 crystal behaves differently than the ceramics having bordering compositions. These issues will be addressed below.

IV. DISCUSSION

Let us try to consider the various, possible interpretations of the staircase-like metamagnetic transitions presently observed in antiferromagnetic, charge and orbitally ordered (AFCOO) manganites.

First, one must notice that multisteps metamagnetic transitions are not rare in the literature on antiferromagnets. This is connected to the existence of a “ferrimagnetic” domain at low-$T$ in the phase diagram, containing one or several intermediate magnetic arrangements in between the antiferromagnetic and paramagnetic phases. In such cases, it must be emphasized that the step fields are true critical fields given by combinations of different exchange fields present in the system. Therefore, they must be well reproducible
and independent of the measuring procedure, in contrast to the behaviors found in AFCOO manganites. In addition, the magnetization of each plateau should also have well defined values, a feature which again strongly departs from our observations.

The literature provides us with another series of results that could be relevant to our situation. In site-diluted antiferromagnets, it was indeed shown that metamagnetic transitions can display several abrupt steps. This was well illustrated in the case of Fe$_{1-x}$Mg$_x$Cl$_2$ for which experimental and simulated $M(H)$ curves bear close resemblance to ours. This behavior was interpreted in terms of avalanches of spin reversals in the AF structure. Random distribution of magnetic vacancies (Mg$^{2+}$ ions) generates a great variety of local situations in the spin lattice. Different values of external field are required to flip a spin in these various neighborhoods. As it is increased, the field first reaches a value inducing spin reversals within the most disturbed regions, probably close to clusters of magnetic vacancies. Once a first spin has been flipped, it modifies the environment of its neighbors which can themselves flip, and so on. This flipping process can propagate as a domino-like wave within the spin structure, resulting in sizeable avalanches. Simulations showed that these avalanches can be blocked by regions locally free of defects or by clusters of previously flipped spins. Then, the field can be further increased over a wide range without triggering new spin reversal, leading to almost constant magnetization. This process yields $M(H)$ curves with large, sharp steps connected by plateaus, just like in our case. Nevertheless, some experimental results cast doubts on the relevancy of this mechanism to our situation. Firstly, we have found that the staircase effect also exists in Pr$_{0.63}$Ca$_{0.37}$MnO$_3$, i.e., without substitution on the spin lattice itself. Secondly, the mechanism described above involves true critical fields, each of them being associated to a particular local arrangement (number and spatial distribution of adjacent vacancies). These critical fields are expected to be reproducible for a given sample. Once again, the modifications in the pattern of steps resulting from cycling or varying procedure cannot be accounted for in the frame of such a scenario.

Actually, the whole set of data indicates that the staircase effect is rather dealing with a succession of metastable states (corresponding to the plateaus), separated by boundaries
the step fields) which values cannot be perfectly reproducible. Keeping this in mind, let us turn back to the specific case of our AFCOO manganites. In such compounds, the metamagnetic transition is widely recognized to proceed through nucleation and growth of F domains within the AF matrix. The most natural idea about the occurrence of multisteps structures consists in invoking additional mechanisms which could impede the development of these nucleation/growth processes. When such a competition involves opposing trends which are very different in nature, it can yield a discontinuous response in various physical systems. For instance, it is the case in ferromagnets where Barkhausen noise results from jerky motions of domain walls, a behavior often attributed to pinning by defects. Related phenomena are also encountered in martensitic transformations (the so-called Acoustic Emission effect). In manganites, it turns out that transitions involving the COO state could have a martensitic-like character. This analogy was previously pointed out in recent publications.\textsuperscript{12–14} A martensitic transformation (MT) is basically a first-order transition (FOT) involving a shear-induced lattice distortion between a high-$T$ phase (austenite) and a low-$T$ phase (martensite). Because of a significant change in shape of the unit cell, the nucleation of first martensitic domains upon cooling induces long-range, anisotropic strains developing from the martensite/austenite interfaces.\textsuperscript{15} The resulting increase of elastic energy can transitorily block further growth of martensitic phase under continuous lowering of temperature (the external parameter driving the FOT in this case). Significant undercooling is actually required to make the transition starting again. This sequential process repeats itself along the course of the transformation. Finally, despite its intrinsic first-order character, the MT is spread over a large range of temperature between $M_S$ and $M_F$ (start and finish temperatures of the austenite to martensite transformation, respectively).

The settling of COO in manganites is well known to result in significant distortions of the unit cell. This originates from cooperative Jahn-Teller effects, giving rise to long-range ordering of the Mn\textsuperscript{3+} $e_g$ orbitals. It must be noted that such phenomena related to internal strains can be in play during either the setting or the collapse of COO domains. Furthermore, similar effects can be expected when COO domains coexist with either paramagnetic or
ferromagnetic (F) phases. The previous studies invoking the martensitic analogy referred to temperature-driven transitions between the paramagnetic and COO phases at $T_{COO}$.\textsuperscript{12,14} In the present paper, we rather consider the AFCOO-F transition, driven by application of magnetic field at low-$T$. The two phases involved in this transition have very different cell parameters, which can justify a martensitic-like character of the transformation. In order to quantify this point, let us compare the results of neutron diffraction data recorded at 10 K under zero field (3T2 at LLB, Saclay, France) in PrCa50 (a good reference for the AFCOO phase) and (PrCa50)Cr5\% (which is ferromagnetic at low-$T$ even under zero field).\textsuperscript{16,17} When going from the AFCOO (PrCa50) to the F phase (PrCa50Cr5\%), $a$ and $c$ parameters ($Pnma$ space group) are decreased by 0.6\% and 0.8\%, respectively, while $b$ is increased by about 1.7\%. Since the cell parameters of the two components are not strictly equal at 300 K, it is still more reliable to compare a distortion parameter, such as $D = (100 / 3) \sum_{i=1,2,3} (|a_i - \langle a \rangle| / \langle a \rangle)$, where $a_i$ refers to the three cell parameters, and $\langle a \rangle$ is the average value. While the $D$ values of both compounds are similar in the paramagnetic phase at 300 K [0.11 and 0.17 for (PrCa50)Cr5\% and PrCa50, respectively], these values are totally different in the ordered phases at 10 K: $D = 0.15$ in (PrCa50)Cr5\%, and $D = 1.19$ in PrCa50. These structural evolutions clearly indicate that large, anisotropic strains must develop at the F/COO interfaces during the metamagnetic transformation. In a canonical MT, the austenite phase transforms into the martensite upon lowering temperature or applying external stress. In our case, the roles of austenite and martensite are played by the COO and F phases, respectively, while increasing magnetic field just acts as lowering temperature or increasing stress. Contrary to the case of standard MT, note that the most distorted phase is the "austenite" in this analogy.

Let us now try to describe the occurrence of multisteps on $M(H)$ curves in the frame of such a martensitic scenario. At low-$T$ and under zero-field, the COO (austenite) phase is extremely predominant, as attested to by the linearity of the $M(H)$ curves. For sufficiently high magnetic fields, F domains (martensite) start growing. A slight deviation from linearity can be detected on the curves above about 3.5 T. This upward curvature, however, is very
small, which can be ascribed to the inhibiting effect of the strains at the F/COO interfaces. As the field is continuously increased, the driving force acting on the spins increases. There is a field value for which this force becomes high enough to locally overcome the elastic constraints, triggering a sudden motion of F/COO interfaces in a part of the sample. This, in turn, destabilizes the local stress field and, in an avalanche-like way, it can result in spreading the F state over large regions of the sample. Such an abrupt increase of the martensite volume fraction (F phase in our case) yields a sizeable increase of magnetization. Along this avalanche process, the magnetic energy decreases while the elastic one increases, a balance which can lead the system to be frozen in another metastable state. The overall transition may thus proceed by successive jumps between metastable states, which directly yield a staircase-like shape on $M(H)$ curves. One could argue that the discontinuous character of the transition in most MT takes place at small scales, making it hardly visible on the evolution of bulk quantities. However, there are some cases for which MT develops by successive “bursts”. For instance, the volume fraction of martensite versus temperature in Ni-Ti alloys can increase by macroscopic steps, a behavior consistent with the existence of the staircase effect seen in $M(H)$ curves of our manganites.

Let us now test more precisely the consistency of this martensitic-like scenario with the whole set of experimental results. Martensitic transformations are known to be basically sensitive to defects, which can influence the nucleation and growth of the product phase within the parent one (pre-existing defects or defects induced by previous transformations). Such a role of defects can explain some slight variations from sample to sample as shown in Fig. 2. The evolution observed between successive $M(H)$ curves can also be ascribed to the influence of defects created by the lattice distortions along the COO-F transformations. In MT, prominent cycling effects are observed when scanning the transition either by temperature or stress variations. In most cases, it was shown that defects produced by thermal cycling act as obstacles making the martensitic transformation more and more difficult. This leads to a decrease of $M_S$, which is qualitatively consistent with the increase of the step fields shown in Fig. 4. Figure 5 shows that the field spacing can affect the location of
the steps. As a general rule, the collapse of any brittle metastable state can be expected to be slightly delayed by smoothly varying the driving (destabilizing) parameter (i.e., the field in our case). The results of Fig. 5 are qualitatively consistent with such a behavior since one observes that smaller field increments tend to push up the breakdown fields (just as in sandpile experiments, when adding grains of sand one by one instead of by packs).

Another important result one has to face is displayed in Fig. 6. Why does one observe a staircase effect in the PrCa37 crystal and not in the PrCa35 and PrCa40 ceramics? Is it related to the $x$ value or to the sample nature? As soon as $x$ departs from 0.5, there must be some disorder in the charge and orbital ordering. In disordered systems for which an underlying first-order transition proceeds through avalanches, the degree of disorder was found to critically influence the development of the transition.\textsuperscript{20} For low levels of disorder, the transition takes place as a single infinite avalanche, while for high levels of disorder, it has a smooth shape made of tiny avalanches (undetectable on the hysteresis loop of a magnetic system, for instance). In between these situations, there is a critical value of disorder for which the transition proceeds through superimposition of avalanches of all sizes. One might thus speculate that some levels of disorder just below the critical value could lead to the appearance of several macroscopic steps on $M(H)$ curves. Nevertheless, this interpretation is made unlikely by the fact that both PrCa35 and PrCa40 exhibit similar shapes of $M(H)$, that are consistent with low disorders. Actually, it seems more plausible to rather invoke the difference of microstructure between crystals and ceramics. Our ceramics are made of grains of typical size close to 100 $\mu$m. A role of the grain size was previously reported in the development of martensitic-like transitions in manganites. Upon cooling across $T_{COO}$, it was found that large grains are more favorable to the establishment of the COO phase.\textsuperscript{21,14} This was ascribed to a better accommodation ability of the martensitic strains as the grain size increases. In our case, the results of Fig. 5 rather suggest that accommodation is more difficult in the crystal than in the ceramics. The literature of MT in metallic alloys shows that there are antagonist effects associated to grain size, leading to various global behaviors. There are cases where the width of the martensitic transition ($M_S - M_F$) was found to
increase with the grain size. This suggests that the hindering effects associated to internal stresses can increase with size, a feature which might be consistent with our results. The influence of microstructure on the staircase effect in manganites obviously deserves further investigations.

V. CONCLUSION

The staircase effect recently discovered in some antiferromagnetic, charge and orbitally ordered (AFCOO) manganites, has been extensively studied by magnetic measurements (isothermal $M$ versus $H$ loops). Various parameters were investigated: Reproducibility from sample to sample; influence of the temperature; cycling effects; measuring procedure. Experimental results allowed us to discard some of the interpretations which could be a priori considered. An important point is that the step fields cannot be regarded as “pure critical fields”. These characteristic fields rather separate metastable states, and their precise values can change with many parameters. It is also determinant to point out that the AFCOO to F transformation involves large structural distortions. Actually, there are striking analogies with the martensitic transformations encountered in metallic alloys. We found that all our experimental features can be reasonably well accounted for within such a scenario.

Although a staircase effect was first evidenced in Mn-site substituted compounds, the exact role of defects in this phenomenon appears to be quite complex. Firstly, we found that this effect can also take place in “pure” Mn compounds. Secondly, our martensitic interpretation is based on structural distortions which also take place in the absence of any disorder. The facts remain that the presence of disorder can be determinant since it favors the creation of deep metastable states separated by large barriers in the energy landscape, a situation well suited to the development of large avalanches.

A lot of aspects of the staircase effect will have to be further investigated. For instance, one will have to check whether it could also be observed in the metamagnetic transitions of other kinds of AF groundstates encountered in manganites, such as the A- or C-types.
Relaxation experiments (under fixed $T$ and $H$) should also be carried out to investigate further the role of temperature, addressing in particular the question of the athermal\textsuperscript{23} (or not) character of these multisteps transitions.

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VII. FIGURE CAPTIONS

FIG. 1. Magnetization loops at 5 K in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$ (circles) and $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ (squares). Inset displays complete scans between 9 and -9 T in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$.

FIG. 2. First magnetization curves at 5 K in four virgin $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$ samples coming from the same batch. The arrows just mark the average values of the three step fields.

FIG. 3. Magnetization loops in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$ at various temperatures.

FIG. 4. Five successive magnetization loops at 5 K in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$. The labels refer to the run numbering. Each loop was recorded after a zero-field cooling from room temperature.

FIG. 5. High-field parts of hysteresis loops at 5 K in $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{Mn}_{0.95}\text{Ga}_{0.05}\text{O}_3$. They were successively recorded using different values of the field spacing ($f_s$) and of the waiting time ($t_w$) after each field installation (first loop: circles; second loop: squares; third loop: diamonds).

FIG. 6. Magnetization loops at 2.5 K in three “pure-Mn” manganites: a $\text{Pr}_{0.63}\text{Ca}_{0.37}\text{MnO}_3$ crystal, as well as $\text{Pr}_{0.65}\text{Ca}_{0.35}\text{MnO}_3$ and $\text{Pr}_{0.6}\text{Ca}_{0.4}\text{MnO}_3$ ceramics.
Figure 1
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Figure 2
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Figure 3
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Figure 4
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Figure 5
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Figure 6
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