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Power-law distribution of avalanche sizes in the field-driven transformation of a phase-separated oxide

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At low temperature ($T \sim 1.5$ K), the isothermal magnetization curves of a phase-separated manganese oxide ($Pr_{0.6}Ca_{0.4}Mn_{0.96}Ga_{0.04}O_3$) are found to exhibit a large number of small steps. Analysis of this phenomenon shows several features which are typical of avalanchelike dynamics. These findings lend further support to the martensitic origin of the magnetization steps encountered in phase-separated manganites.

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I. INTRODUCTION

The mixed-valent manganites with general formula $R_{1-x}A_xMnO_3$ (R being a trivalent rare-earth ion, and A a divalent alkaline-earth ion), are known to be subject to phase separation phenomena, i.e., the coexistence of domains corresponding to different magnetoelectronic states, in a chemically homogeneous material.[1] The balance between such competing states is sensitive to many parameters. In particular, the application of a magnetic field often leads to the development of a ferromagnetic (FM) component, growing at the expense of the surrounding antiferromagnetic (AF) phase(s).

For Mn-site substituted manganites of formula $Pr_{1-x}Ca_xMn_{1-y}M_yO_3$ (where $x \sim 0.5, y \sim 0.05$ and M is a cation destabilizing the Mn sublattice), it was recently observed that the growth of the FM fraction as the field is increased exhibits a peculiar steplike character.[2–4] A spectacular manifestation of this effect is the staircaselike shape of the magnetization curves versus field $[M(H)]$ at low temperatures. A recent neutron diffraction experiment, carried out on $Pr_{0.5}Ca_{0.5}Mn_{0.97}Ga_{0.03}O_3$, demonstrated directly the coexistence of FM and AF phases, as well as the burstlike growth of the former at the expense of the latter when the field is increased.[5] In this class of compounds, the CE and/or pseudo-CE type AF ordering is accompanied by a collective orbital ordering (OO) of the $Mn^{3+}d_{z^2}$ orbitals, which distorts the basic unit cell.[6] Therefore, a local transformation from this AFOO state to the FM state generates long-range anisotropic stresses inside the material. We have proposed that the occurrence of steps in the growth of the FM fraction results from this martensitic character of the field-induced AFOO/FM transformation.[7]

The present paper reports on a new phenomenon that has been observed by recording the field dependence of the magnetization at temperatures ($T < 2.5$ K) lower than those investigated so far. For Mn-site substituted compounds showing only a few steps at $T \geqslant 2.5$ K, the $M(H)$ curve at $T = 1.5$ K exhibits numerous small magnetization steps with a range of amplitudes. In this paper, we present an experimental study of this phenomenon by focussing on one of these compounds. We report on investigations of the reproducibility, self-similarity, and size distribution of the steps. These results are found to be consistent with general expectations for systems governed by avalanchelike dynamics.

II. EXPERIMENTAL DETAILS

This study has been carried out on the compound $Pr_{0.6}Ca_{0.4}Mn_{0.96}Ga_{0.04}O_3$, hereafter denoted as [PrCa40]Ga4%. Ceramic samples were prepared following a standard solid state reaction.[2] The steplike magnetization curves shown by this material for $T \geqslant 2.5$ K have been reported previously.[3,8] This compound has a phase-separated ground state, made up of a sizeable FM component ($\sim 20\%$ in volume) embedded within a short-range AFOO matrix.

The $M(H)$ curves were recorded by means of a vibrating sample magnetometer (VSM Oxford Instruments) down to a base temperature of 1.5 K, collecting data at a constant sampling rate of 1 Hz whilst continuously sweeping the magnetic field. The paper focuses on the data recorded with a sweep rate of 0.1 T/min, leading to a spacing between data points equal to ~ 0.0016 T. To ensure a good sample thermalization, we worked with small, thin samples (mass ~ 10 mg) which were glued with General Electric varnish on the sample holder. The temperature was monitored and remained within the range 1.500 ± 0.003 K over the time required to record a $M(H)$ loop up to 8 T at 0.1 T/min. The influence of the sweep rate value could only be investigated within a limited range (i.e. 0.1–1 T/min), in order to avoid problems with the temperature stability (at lower sweep rates) and because of experimental limitations (the maximum sweep rate is 1 T/min). All the hysteresis loops were recorded after a zero-field-cooling (ZFC) from 300 K, i.e., starting from a state with no spin, charge or orbital ordering.

III. RESULTS AND DISCUSSION

The main panel of Fig. 1 shows a $M(H)$ curve recorded at $T = 1.5$ K for [PrCa40]Ga4%. The rounding of the curve in the low-field range is a manifestation of the significant FM component that exists in the ground state of this compound. Above ~ 2.5 T, M starts increasing via a succession of numerous small steps. For $H > 4.5$ T, this jerky shape is re-

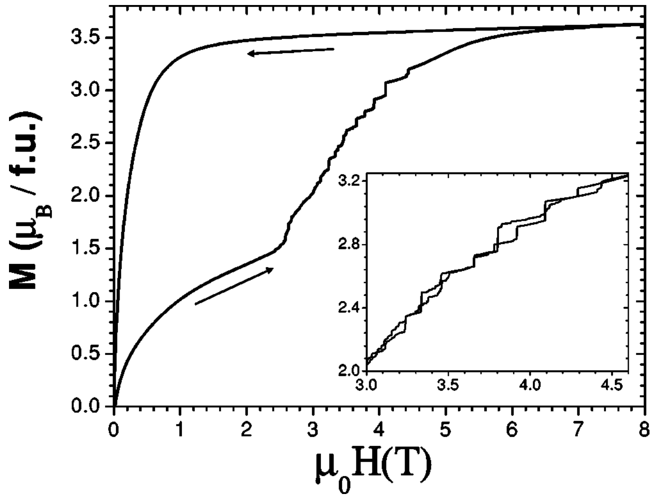

FIG. 1. $M(H)$ loop recorded at $T=1.5$ K, after a zero-field cooling, on $Pr_{0.6}Ca_{0.4}Mn_{0.96}Ga_{0.04}O_3$. The arrows indicate the directions of the field variation. The inset is an enlargement of the field-increasing branches of two independent runs recorded on the same sample.

placed by a smooth tail reaching the saturation magnetization at $\sim$8 T. Note that the value $M_{\mathrm{Sat}} \sim 3.62\ \mu_B/\mathrm{f.u.}$ contains a contribution from the Pr^{3+} ions, in addition to the full polarization of the Mn spins (3.44 $\mu_B/\mathrm{f.u.}$ for the complete FM state). The reverse leg of the loop is almost flat down to $\sim$1 T, before decreasing rapidly towards zero owing to the standard disorientation among the FM domains. Increasing the field after such a half-loop leads to a curve (not shown) that is superimposed onto the field-decreasing branch, which demonstrates that the field-induced transformation to the FM state is totally irreversible at this temperature. All these features of the $M(H)$ at 1.5 K were observed in the three samples of [PrCa40]Ga4% which were investigated. It should also be emphasized that similar curves showing a multitude of small jumps in $M(H)$ at 1.5 K have been observed in many other $Pr_{1-x}Ca_xMn_{1-y}M_yO_3$ compounds, for different values of x (e.g., 0.4 and 0.5), different values of y (e.g., 0.03, 0.04, and 0.05), and for various substituting cations M (e.g., Ga, Al, and Sn).

The behavior presently found at $T=1.5$ K is qualitatively different from the magnetization jumps reported previously for this class of materials. At higher temperatures, the transformation to the FM state was always found to take place via a few, large jumps, separated by wide plateaux.[2–4] For instance, for [PrCa40]Ga4% at $T=2.5$ K, the $M(H)$ curve contains only two large jumps of amplitude equal to $\sim$1.5 and $\sim$0.8 $\mu_B/\mathrm{f.u.}$, taking place at $\sim$1.5 and $\sim$3.5 T, respectively.[3] Note that such abrupt jumps were also found in the present VSM measurements. It must be emphasized that while the patterns of large steps are reproducible for [PrCa40]Ga4% at $T \geqslant 2.5$ K,[3,8] the exact locations of the small steps at $T=1.5$ K vary significantly from one run to the other. The stochastic character of the jerky transformation is illustrated in the inset of Fig. 1. A similar behavior was recently observed during magnetization reversal of Fe films.[9] Regarding the location of the jumps in phase separated manganites, it should be noted that the microstructure (i.e., the details of the grain structure in our ceramic samples) can be expected to play a role, as previously demonstrated in the higher-T regime ($T \geqslant 2.5$ K).[10] Note that such an influence of microstructural features is consistent with a martensiticlike behavior.

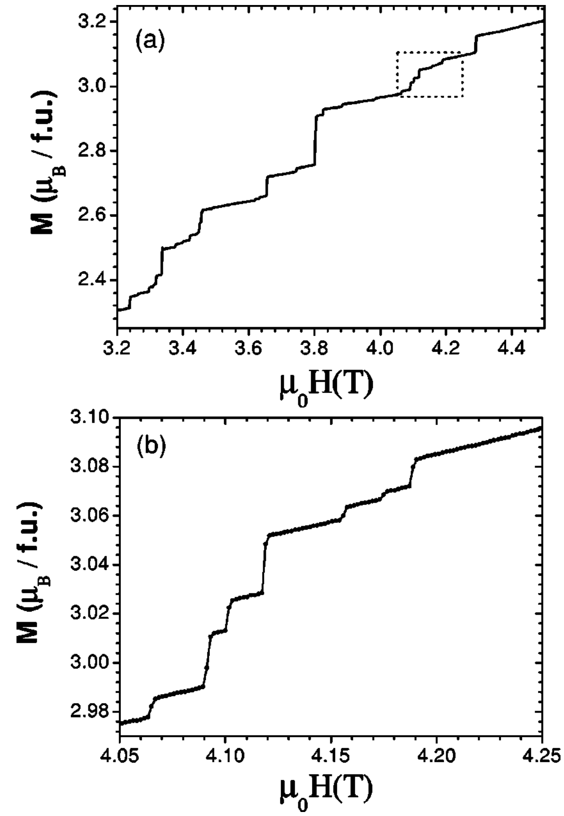

FIG. 2. Field-increasing branch of a $M(H)$ loop at $T=1.5$ K, shown at two scales: (b) is an enlargement of the dotted window drawn in (a).

Figure 2(a) is an enlargement of the central part of the field-increasing branch of a $M(H)$ curve at $T=1.5$ K. One can clearly see that M increases through a succession of jumps and plateaux of different amplitudes and widths, respectively. Figure 2(b) displays an enlargement of the portion of the curve contained in the dotted window of Fig. 2(a). It is found that the global aspect of the data at this smaller scale remains the same as that of the whole curve. Such a phenomenon of self-similarity is an expected feature of avalanchelike behavior. Related observations were recently discussed by Carillo *et al.* in work on curves of *released energy versus load* along a martensitic transformation.[11]

The avalanchelike behavior only appears at very low temperatures, i.e., below 2 K for the three samples investigated. A detailed study carried out on one of these samples has shown that the crossover in temperature is quite sharp: At a fixed sweep rate of 0.1 T/min, the $M(H)$ curves systematically show many small steps at 1.5 K, whereas there are only a few large steps at 1.75 K. The investigation of the influence of the sweep rate, dH/dt, was made difficult by the stochastic character of the avalanchelike regime (variations from run to run for a fixed dH/dt). However, by recording several series of $M(H)$ curves with different dH/dt, it has been

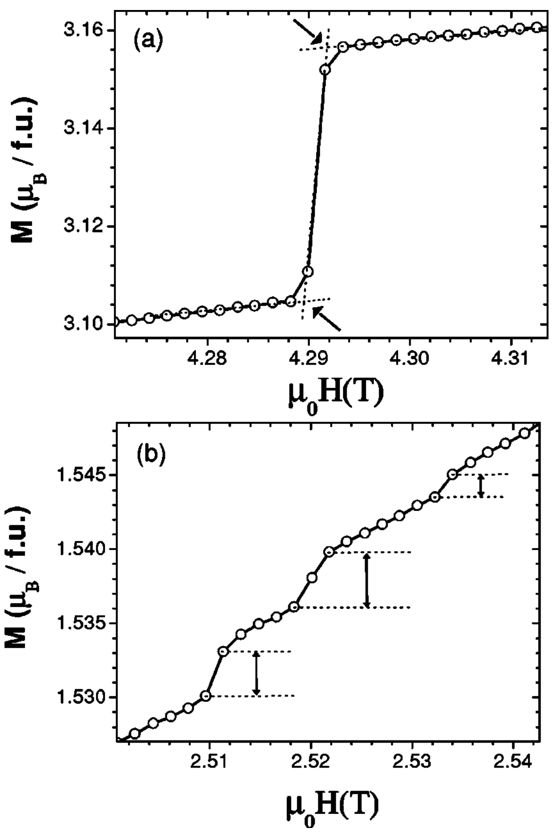

FIG. 3. Illustration of the constructions used to evaluate the height of the magnetization jumps. For large steps (a), ΔM is derived from the intersection points denoted by the arrows (see text). For small steps (b), ΔM is directly estimated from the data points flanking the jumps, as denoted by the arrows.

found that the number of steps along the magnetization curves tends to decrease as dH/dt is increased (by approximatively a factor of 2 between 0.1 and 1 T/min). The influence of this parameter deserves further investigations based on a broader range of dH/dt values (presently limited by the practical constraints discussed in Sec. II).

The distribution of the amplitudes of the jumps has been analysed for the data of Fig. 1, which corresponds to the situation (1.5 K and 0.1 T/min) exhiting the largest number of steps. The way these amplitudes have been estimated is displayed in Fig. 3. For big steps [see Fig. 3(a)], there are often one or two intermediate points along the jump. The height of the magnetization step (ΔM) was derived from constructions similar to those shown on Fig. 3(a), using the points of intersection with linear extrapolations of the curve on both sides of the jump. For the smallest jumps, the estimate of ΔM is more problematic, as illustrated in Fig. 3(b). Such small steps were often found to be separated by small field intervals, which prevented us from using the above procedure. Thus, for the smallest steps (i.e. below $\sim 0.01\ \mu_B/\mathrm{f.u.}$), ΔM was evaluated by directly considering the magnetization values of the points flanking the jump [see the dotted lines on Fig. 3(b)]. It must be recognized that there is a sizeable relative uncertainty for the smallest ΔM values. Figure 3(b) also shows that it is not possible to detect steps with amplitudes lower than $\sim 0.002\ \mu_B/\mathrm{f.u.}$

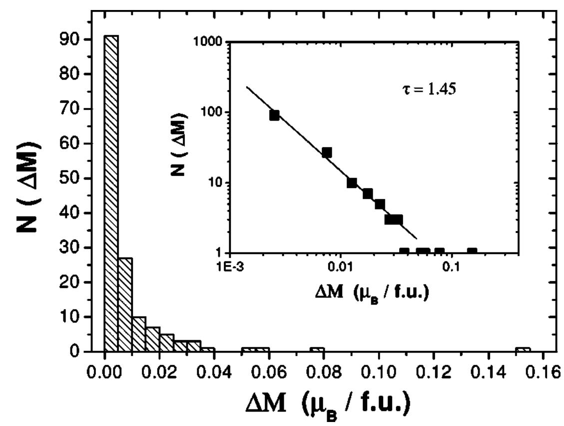

FIG. 4. Histogram of the distribution of magnetization jumps along the field-increasing branch of an $M(H)$ loop at 1.5 K (main panel). The inset shows the same data in a log-log plot. The solid line is a fit corresponding to a power law $N(\Delta M) \propto \Delta M^{-\tau}$, with $\tau = 1.45$.

Figure 4 displays a histogram of the size distribution $[N(\Delta M)]$ of the 151 steps identified along the $M(H)$ curve at $T=1.5$ K (Fig. 1), between 3 and 4.5 T.[12] The inset of Fig. 4 shows a log-log plot of the distribution corresponding to the main panel. Neglecting the binning classes with $N=1$, one observes a relationship of the type $N(\Delta M) \propto \Delta M^{-\tau}$, with $\tau \sim 1.45 \pm 0.15$. It must be emphasized that the value of this exponent may be dependent on the sample, as well as on other parameters such as the temperature and the field sweep rate. Unfortunately, the number of jumps contained in the $M(H)$ curves collected at different temperatures and sweep rates was too small (less than 30) to obtain a reliable estimate of τ.

Avalanchelike systems are known to be characterized by a power law distribution of the avalanche sizes. Experimental observations of this kind have been reported for a large variety of phenomena: For example, earthquakes,[13] martensitic transformations,[14,15] Barkhausen noise,[16,17] etc. From a theoretical point of view, the value of the exponent τ is not supposed to depend on the details of the system, but essentially on its dimensionality. For three-dimensional systems (3D), the expected mean-field value is $\tau = 3/2$.[18,19] Most of the analysis of avalanchelike systems are based on either random field Ising models (RFIM) (Refs. 20–22) or self-organized criticality (SOC).[18,23,24] Numerical simulations within the framework of 3D RFIM give values of τ ranging in between 1.6 and 1.8,[21,25] while an approach in terms of SOC leads to 1.35.[23] Our experimental result ($\tau \sim 1.45$) is found to be reasonably consistent with these general predictions. For avalanchelike dynamics, it should be noted that the exponent τ is expected to be sensitive to the variation rate of the driving parameter,[17,26,27] i.e., the magnetic field sweep rate in our case (as is mentioned above, this prediction cannot be checked with the present data).

We have previously discussed the possibility that the large steps observed in the $M(H)$ curves of Mn-site substituted manganites are related to the martensitic nature of the field-induced transformation from an AFOO phase to the FM phase.[7] We propose that the series of smaller steps observed at lower temperatures also result from the same phenomenon. Using lower temperatures makes the system sensitive to smaller energy barriers (related to the elastic energy associated with the internal strain field developing from the AFOO/FM interfaces), which leads to the occurrence of a larger number of smaller steps.

In standard martensitic systems, the avalanche size distributions have been mostly investigated through the amplitudes of the peaks in acoustic emission (AE) spectra.[11,14,28] These studies led to values of τ in the range 2.3–3.1, i.e., quite different from ours. It must be emphasized, however, that the experimental τ value can be strongly affected by the nature of the physical quantity used to characterize the "avalanche size."[28] While our measured ΔM values are expected to be proportional to the volume of material involved in each avalanche, such a relationship is not so straightforward when considering the peak amplitudes of AE spectra. Therefore, rather than comparing our data with such experimental results, let us consider studies using the size distribution of the ΔM values. According to the expected universality of the avalanche dynamics, the τ values derived from such distributions should not depend on the exact nature of the domain walls involved in the transformation. For instance, a value of $\tau \sim 1.7$ was derived from the distribution $N(\Delta M)$ for the field-induced transformation of a Cu-Al-Mn intermetallic alloy (spin-glass phase to ferromagnetic spin-glass phase transition).[29] Actually, most of the studies based on $N(\Delta M)$ distributions have dealt with the analysis of the Barkhausen jumps in ferromagnets. For 3D systems, the experimental values of τ extracted from such an analysis[21,30,31] were found to lie in the range 1.3–1.8. Our τ value is thus consistent with such studies of Barkhausen jumps, i.e., stochastic movements of Bloch walls. In our case, the domain walls in play are the AFOO/FM interfaces, which separate domains that are not only magnetically but also structurally different.[5,7] Note that the latter feature is at the origin of the martensitic character of the transformation.

Even though our data is consistent with both theoretical expectations and experimental results related to avalanche processes, it must be recognized that the statistics of the ΔM distribution will have to be improved in order to get a more precise determination of τ. Better statistics are also required to properly address the influence of some parameters such as the magnetic sweep rate. We suggest that investigations at still lower temperatures ($T < 1.5$ K), with a pickup coil technique to directly detect the magnetic flux variations, should extend the range of accessible avalanche sizes that can be measured.

IV. CONCLUSION

Magnetization-vs-field curves of a phase-separated manganite are found to exhibit a multitude of small jumps at low temperature ($T \sim 1.5$ K). We propose that this feature reflects the stepwise growth of the FM component in the phase-separated state (coexistence of FM and AFOO domains). The burstlike nature of this growth can be ascribed to the structural distortions that exist between the AFOO and FM phases, which give a pronounced martensitic character to the field-driven transformation of these manganites.

The magnetization jumps were found to have a stochastic nature and a self-similar character. Moreover, the quantitative analysis of a $M(H)$ curve showing a large enough number of jumps has led to a size distribution following a power law with an exponent $\tau \sim 1.45$. All these features are typical of processes taking place through avalanches, as is the case for martensitic transformations. The present results give strong support to the martensitic interpretation of the stepwise field-induced transformations in this class of manganites.

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