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Quenched-disorder-induced magnetization jumps in $(Sm,Sr)MnO_3$

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Magnetic field induced steplike changes in magnetization and resistivity of $Sm_{1-x}Sr_xMnO_3$ manganites were studied. A strong dependence of these features on the cooling rate was observed. Magnetostriction, however, does not show the presence of large strain in our samples. From all these features we can rule out the conventional explanation of magnetization jumps as a consequence of martensitic transition. We propose instead that quenched by fast cooling disorder leads to the formation of an inhomogeneous metastable state and to subsequent magnetization jumps.

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Magnetic field-induced first order phase transitions attract a lot of attention both in conventional antiferromagnets (AFM)[1] and in mixed-valence manganites (see Ref. 2 and references therein) as well as in some pseudobinary systems.[3,4] In AFM these transitions are usually reversible and relatively broad.[1] In diluted metamagnets (for example, $Fe_xMg_{1-x}Cl_2$) they may be steep (avalanchelike) and hysteretic.[5] In manganites such transitions may be sharp or broad, reversible or strongly hysteretic[6] depending on chemical composition and temperature, and are often accompanied by structural and insulator-to-metal (I-M) transition.[7–9]

Recently, the field-induced phase transition to a ferromagnetic (FM) state was shown to be discontinuous at low $T < 5$ K in ceramic Mn-doped $Pr_{0.5}Ca_{0.5}MnO_3$,[10,11] in ceramics and single crystal of $Pr_{1-x}Ca_xMnO_3$ ($x=0.3-0.37$)[12,13] and in $Gd_5(SiGe)_4$ alloys.[3,4] The position and number of steps depend on the magneto-temperature history and on the magnetic field sweep rate.[3] This was interpreted as the result of some kind of martensitic transformation. However, this scenario is not clear because grain boundaries in ceramics could be intrinsic barriers for domain-wall movement. Ghivelder *et al.*[14] have observed a huge temperature increase at such abrupt field-induced transition (from 2.5 to 30 K), whereas the specific heat before and after transition differs only by 10%. This implies that a large (magnetic) entropy is frozen in the sample and abruptly released upon increase of the magnetic field. The authors of Ref. 14 have proposed a model in which the local AFM-FM transition releases the heat locally and triggers a heat avalanche leading to the observed magnetization jumps. In this case, the step should have some finite characteristic time scale of the order of a thermal relaxation time. However, only the magnetic field width of the step was discussed and was found to be less than 2 Oe[12] or even strictly zero.[14]

In this paper we show that steplike behavior exists in the magnetization M and resistivity ρ but not in the magnetostriction of $Sm_{1-x}Sr_xMn^{18}O_3$ ($x=0.45$, 0.5) ceramics. These steps have a characteristic time scale of the order of 1 ms which does not depend on the magnetic field sweep rate. Moreover, the low-field low-temperature magnetic state itself strongly depends on the zero-field cooling rate. There are no M and ρ steps for slowly (1 K/min) cooled samples, but they exist only for rapidly cooled samples. In the latter case there is an additional linear term in the specific heat vs temperature dependence. We suggest that frozen magnetic disorder and corresponding entropy is responsible for the large overheating at the avalanchelike transition to the FM state upon increasing the magnetic field.

Ceramic $Sm_{1-x}Sr_xMn^{18}O_3$ samples with $x=0.45$, 0.5 and $(NdEu)_{0.55}Sr_{0.45}Mn^{18}O_3$ were prepared by a solid-state reaction technique, described in Ref. 15. The Nd/Eu ratio was selected to fit the Sm ionic radius. The magnetic and electric behavior of SmSr and (NdEu)Sr samples is qualitatively identical [see Fig. 3(b)]. The enrichment of the samples by ^{18}O was performed at $T=950$ °C and at a pressure $p=1$ bar for 200 h using the method reported in Ref. 16. Magnetization was measured by a Quantum Design MPMS-7 SQUID magnetometer and by a vibrating sample magnetometer. High-speed (up to 100 000 samplings per second) measurements of the magnetization were performed using Fitz's technique and a fast analog-to-digital data acquisition board (Data Translation). Resistivity, specific heat, and magnetostriction were measured in PPMS-9 cryostat. To measure magnetostriction we used WK-06-062AP-350 strain gauges (Vishay Intertechnology) bonded to the sample with a proper epoxy. The striction was detected by the change in resistance of the strain gauge.

$Sm_{1-x}Sr_xMnO_3$ with $x \approx 0.5$ is known to be in the vicinity of the I-M and AFM-FM transition.[17] So, for this system the electronic and magnetic state can be tuned by the application of a magnetic field or by oxygen-isotope substitution. $Sm_{0.5}Sr_{0.5}Mn^{18}O_3$ is an insulator in the low-temperature ground state, and undergoes an I-M transition after ^{18}O-to-^{16}O substitution, after reduction of the doping level to $x=0.45$, or under application of the magnetic field of the order of 1 T.[18] Our magnetization data (Fig. 1) show that there are

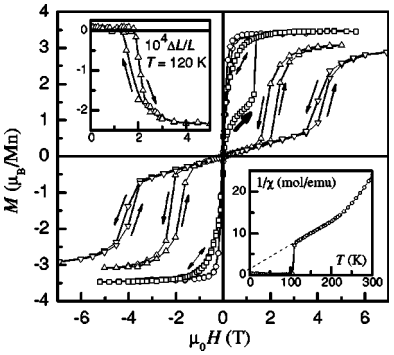

FIG. 1. Magnetization loops for $Sm_{0.55}Sr_{0.45}MnO_3$ recorded after zero-field cooling from room temperature. The data for the first increase of the magnetic field are shown in the first quadrant and marked with bold arrows if different from stable curves (III quadrant). $T=5$ K (□), 55 K (○), 120 K (△), and 140 K (▽). The upper left inset shows magnetostriction at 120 K, the lower right one presents a high-temperature tail of inverse susceptibility at $\mu_0 H = 0.1$ T.

two types of field-induced transition for ^{18}O samples: at low temperatures there appears an irreversible transition from an AFM to FM state, and at high temperatures $T > T_c$—a reversible albeit slightly hysteretic PM-FM transition. Magnetostriction of about 2×10^{-4} is clearly seen to accompany the PM-FM transition (upper inset) in accordance with the data of Ref. 19. The $1/\chi$ data just above T_c show a tendency for AFM interactions that competes with FM ordering at intermediate temperatures.

The irreversible metamagnetic AFM-FM transition is shown to be steplike (Fig. 2) after zero-field cooling. We have observed that the step location depends on the sweep rate of the following magnetic field: the smaller the sweep rate, the larger the field value needed to realize the transition. For the $Sm_{0.5}Sr_{0.5}Mn^{18}O_3$ sample at the rate $\leqslant 250$ Oe/s the transition becomes smooth. This shows that the steplike transition is not an intrinsic property of a compound. Two questions arise at this point: (i) how sharp are these steps and (ii) is M a function of the magnetic field along all the steps or is only the start of the transition triggered by the magnetic field?

To resolve these problems we have studied the $M(H)$ steps at different sweep rates with our high-speed experimental setup with 10 μs resolution. The results shown in the inset to Fig. 2 demonstrate the finite width of the transition of the order of 1 ms. Note that this curve is the same for different sweep rates (200–3000 Oe/s). Being triggered, the transition will complete in a definite time independent on the further changes of the H. In the case of $Sm_{0.5}Sr_{0.5}Mn^{18}O_3$, this time scale is approximately 10 times higher (not shown). Thus, our samples with a relatively small difference in Sr content exhibit a factor of 10 difference in the transition time. This fact cannot be easily reconciled with the scenario of a martensitic transition because the microstructure of both samples is identical. Moreover, a distribution of avalanches in martensitic transformations has usually no characteristic time scales[20] unlike our observations.

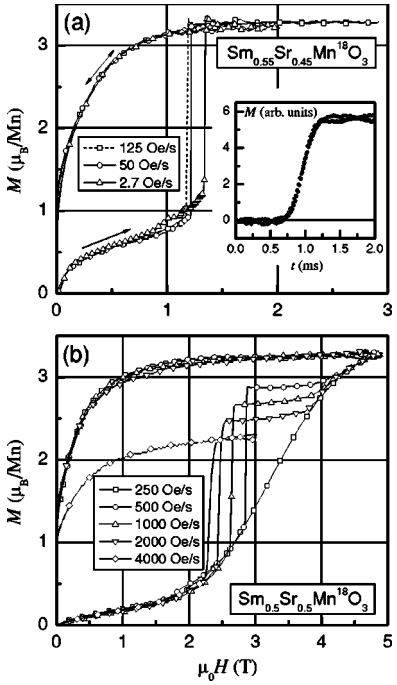

FIG. 2. The dependence of the magnetization jump on the magnetic field sweep rate for $Sm_{0.55}Sr_{0.45}Mn^{18}O_3$ (a) and $Sm_{0.5}Sr_{0.5}Mn^{18}O_3$ (b) at $T=5$ K. Inset shows the time dependence of the magnetization during the jump. This dependence is the same for the sweep rates in the range 200–3000 Oe/s.

To further elucidate the origin of such sharp transitions, we have checked the effects of the cooling rate on the low-temperature magnetic state (Fig. 3). The quenched-in disorder should be strongly affected by the cooling rate. In the case of martensitic transformation,[20] a slow cooling rate leads to a low defect concentration so that the strains are released via large avalanches. On the other hand, fast cooling results in a large number of defects and thus leads to a sequence of many small avalanches. However, as we will discuss below, the behavior observed in our experiments with different cooling rates shows the opposite tendency. This is in our opinion a strong argument against the interpretation of the jumps as a consequence of martensitic phenomena.

Two cooling rates were used in our experiments. In the first, slow cooling, the sample was cooled at the rate of 1 K/min from 300 K to 5 K in zero applied field for transport and specific heat measurements and 100 Oe for magnetization experiments. In the second, fast cooling regime, the cooling rate was 10 K/min for resistivity and specific heat measurements and approximately 20 K/min for magnetization. Usually, no specific information is provided in the literature on the employed cooling rate in manganite research, and we assume that most results are obtained using a relatively fast cooling rate.

Two key points should be noted in the data of Fig. 3. The first is a huge thermal hysteresis for the data obtained on heating and cooling, which is generally considered to be a fingerprint of a first order phase transition. The second is a striking difference in the low-temperature states of the

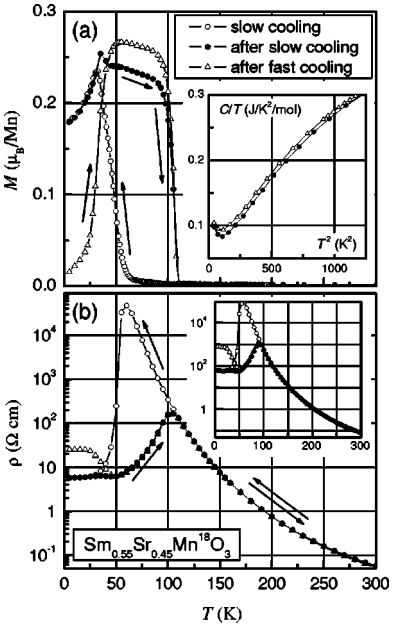

FIG. 3. The temperature dependences of the low-field (0.01 T) magnetization (a) and zero-field resistivity (b) of $Sm_{0.55}Sr_{0.45}Mn^{18}O_3$ obtained for different temperature histories: (○) slow cooling, (●) heating after slow cooling, and (△) heating after fast cooling. Insets show the temperature dependence of the specific heat (a) and resistivity (b) of $(NdEu)_{0.55}Sr_{0.45}Mn^{18}O_3$ for the same temperature histories.

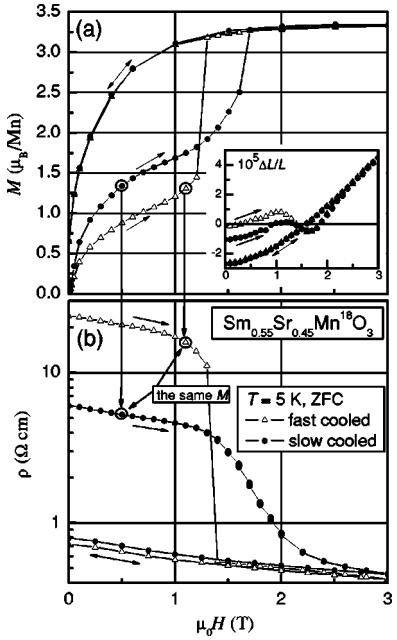

FIG. 4. Magnetization (a) and resistivity (b) loops for $Sm_{0.55}Sr_{0.45}Mn^{18}O_3$ recorded at 5 K after slow and fast cooling in zero field, which demonstrate the absence of jumps for slowly cooled samples. The inset shows a minor effect of the cooling rate on the magnetostriction. Here the parabolic increase is caused by the magnetoresistance of the gauge itself.

fast- and slow-cooled samples. For the former, we observe a low-magnetization state with a relatively high resistivity. For the latter, the magnetization is 10 times higher and the resistivity is smaller. The additional linear term in the specific heat appears for the fast cooled sample as shown in the inset to Fig. 3(a). These differences diminish upon heating and disappears at $T \approx 45$ K.

The low-temperature magnetization and resistivity loops after slow and fast cooling (Fig. 4) have qualitatively different behavior. The steplike transition in $M(H)$ and $\rho(H)$ exists only for fast cooled samples, whereas this transformation is smooth in slow cooled samples. Comparison of the data of Figs. 4(a) and 4(b) shows that the fast-cooled samples have significantly larger resistivity even at the same value of M [compare, for instance, $\rho(H=0.5, 1.1$ T)]. This is an evidence for a quenched disorder in fast-cooled samples (cf. Ref. 21). The other evidence of an additional disorder is shown in the inset to Fig. 3(a). The fitting of the temperature dependence of the specific heat is the same for both cooling rates except an additional linear term of the order of 20% in the fast-cooled sample. (Note, that conductivity in this case is even lower and the electron term could not be the origin of this change.) Corresponding extra entropy $\Delta S = 0.25$ J/K/mol is of the same order as the entropy change upon PM-FM transition (~ 0.6 J/K/mol), thus we believe this extra linear term to have essentially magnetic origin, possibly spin-glass-like type.[22] However, there is almost no additional strain (less than 10^{-5}) in the fast-cooled sample and the magnetostriction is smooth for both histories. This, together with the opposite dependence of the behavior of our samples on the cooling rate, mentioned above, in our opinion rules out the description of these phenomena as a consequence of a martensitic nature of the transition.

Upon cooling, the AFM-FM competition may result in a strongly disordered magnetic state, which has an excess specific heat and enhanced magnetocaloric effect.[23] Ferromagnetic ordering with the external magnetic field would lead to the reduction of this extra entropy. In this case, the local release of the frozen entropy may result in the avalanchelike overheating of the sample because the higher the temperature the more tendency to ferromagnetism is observed at low temperatures (cf. the data for 5 and 55 K in Fig. 1). So, both the magnetization and resistivity change in a jumplike fashion. Such effect was observed recently[14] on the steplike transition. This scenario assumes the time scale of the transition to be inversely proportional to the thermal conductivity of the sample. This is exactly the case if one compares $Sm_{1-x}Sr_xMn^{18}O_3$ with $x = 0.45$ and 0.5. The resistivity of $Sm_{0.5}Sr_{0.5}Mn^{18}O_3$ is at least four orders of magnitude higher[18] so that the thermal conductivity should be lower. We recall that the observed time scale of the jump for $Sm_{0.5}Sr_{0.5}Mn^{18}O_3$ is 10 times larger than for $Sm_{0.55}Sr_{0.45}Mn^{18}O_3$.

Summarizing, we studied the magnetization and resistivity jumps in $Sm_{1-x}Sr_xMn^{18}O_3$, especially their dependence on the cooling rate. The slow (1 K/min) cooled samples demonstrate a smooth AFM-FM transition upon increase of the magnetic field, in contrast to the fast (~ 10 K/min) cooled samples where this transition is steplike with a characteristic time scale of 1 ms. The results obtained (the

dependence of the jumps on the cooling rate; the constant time scale of the jumps, independent on the field-sweep rate; the behavior of specific heat; the absence of noticeable magnetostriction) disagree with the often used interpretation that the magnetization steps originate from the strain release at a martensitic transition. We suggest that the frozen disorder and the associated entropy is the origin of an excess specific heat which is released in an avalanchelike way upon applying a magnetic field, resulting in a heat burst in the sample and in steps in the magnetization and resistivity.

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