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Possible spin-glass state in SmSr-manganites as the origin of the magnetization jumps

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Abstract

Magnetic field-induced step-like changes in magnetization of $\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$ manganites were studied. A strong dependence of these features on the magnetic-field sweep rate was observed. The notable overheating of the sample, starting exactly at the start of the magnetic transition, was observed upon the transition. We suggest that quenched disorder leads to the formation of an inhomogeneous (spin-glass-like) state and to subsequent magnetization jumps driven by a release of latent heat.

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1. Introduction

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].

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 $\text{Pr}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ [4], in ceramics and single crystal of $\text{Pr}_{1-x}\text{Ca}_x\text{MnO}_3$ ($x = 0.3-0.37$) [5] and in $\text{Gd}_5(\text{SiGe})_4$ alloys [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. [6] 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.

In the recent paper [7] it was shown that step-like behavior exists in the magnetization M and resistivity ρ but not in the magnetostriction of $\text{Sm}_{1-x}\text{Sr}_x\text{Mn}^{18}\text{O}_3$ ($x = 0.45; 0.5$) 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 cooled samples, but they do exist for rapidly cooled samples. In the latter case there is an additional linear term in the specific heat vs. temperature dependence. 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), suggesting this extra linear term to be of an essentially magnetic origin, possibly spin-glass-like type [8]. It was

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supposed that frozen magnetic disorder and corresponding entropy is responsible for the large overheating at the avalanche-like transition to the FM state upon increasing the magnetic field.

At this point the question arises. Is this transition really temperature-driven one or the temperature increase observed in Ref. [6], is just a consequence of the magnetic transition? In this context, the synchronous investigation of the magnetization and the temperature seems to be promising to clarify this aspect.

In this paper, we report the simultaneous measurements of the magnetization and the sample temperature in ‘real-time’ (with 10 μ s resolution). The notable sample overheating (of about 5 K) has been observed even when the sample is immersed in the liquid helium. This overheating is shown to develop simultaneously with the magnetization transition.

2. Experiment

Ceramic $\text{Sm}_{1-x}\text{Sr}_x\text{Mn}^{18}\text{O}_3$ samples with $x = 0.45, 0.5$ were prepared by a solid-state reaction technique. The enrichment of the samples by ^{18}O was performed at $T = 950^\circ\text{C}$ and at a pressure $p = 1$ bar for 200 h using the method reported in Ref. [9]. Magnetization was measured by a Quantum Design MPMS-7 SQUID magnetometer, by a vibrating sample magnetometer, and using Fitz’s technique (for high-speed measurements). High-speed (up to 100 000 samplings/s) measurements of the magnetization and the temperature were performed using a fast analog-to-digital data acquisition board (Data Translation). The temperature change was detected by the (Au + Fe)/Cu thermocouple connected to a high-speed amplifier.

$\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$ with $x \approx 0.5$ is known to be in the vicinity of the I – M and AFM–FM transition. So, for this system the electronic and magnetic state can be tuned by the application of a magnetic field or by oxygen-isotope substitution. $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{Mn}^{18}\text{O}_3$ is an insulator in the low-temperature ground state, and undergoes an insulator–metal (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 [10].

The irreversible metamagnetic AFM–FM transition was shown to be step-like after zero-field cooling. The step location was observed to depend on the sweep rate of the magnetic field: the smaller the sweep rate, the larger field value is needed to realize the transition [7]. For $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{Mn}^{18}\text{O}_3$ sample at the rate < 250 Oe/s the transition becomes smooth (Fig. 1). This suggested that the step-like character of the transition is not an intrinsic property of a compound and could be related to the heat excess, released at the transition [6].

To clarify this question we have studied the $M(H)$ steps and their relation with the temperature change at different sweep rates by our high-speed experimental setup with 10 μ s resolution. The results demonstrate the finite width of the transition of the order of 1 ms for the sample with

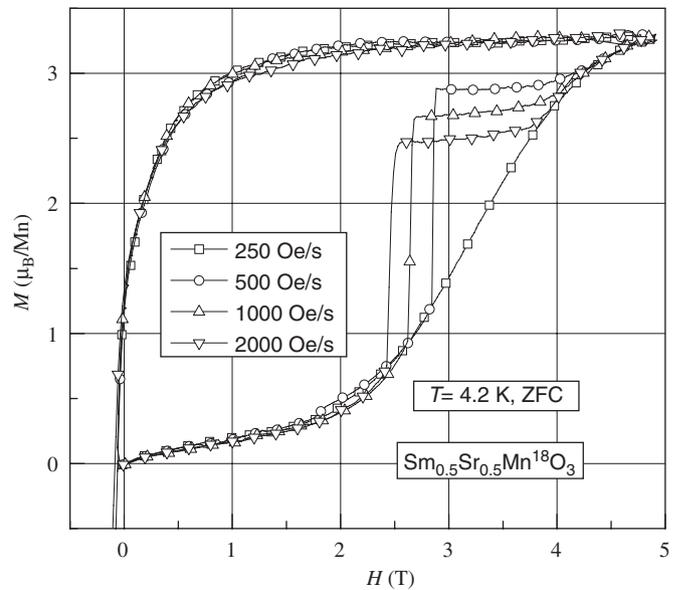


Fig. 1. The dependence of the magnetization jump on the magnetic-field sweep rate.

$x = 0.45$ (Fig. 2(a)) and approximately 10 ms for $x = 0.5$ (Fig. 2(b)). Note that these $M(t)$ curves are almost the same for different sweep rates (800–3200 Oe/s). (The curves in Fig. 2(b) were arbitrarily scaled by about $\pm 10\%$ to emphasize their coincidence). This means, that being triggered, the transition will complete in a definite time independent on the further changes of the H . Thus, our samples with a relatively small difference in Sr content exhibit a factor of ten differences 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 [11] unlike our observations.

To check the possible relation of the magnetic transition with overheating of the sample, we investigated simultaneous changes in the magnetization and the temperature. In this measurement the sample was immersed in the liquid helium bath, with the thermocouple being attached to the sample with the silver glue. The other thermocouple junction was kept in the liquid helium. The result obtained for the magnetic field sweep rate of 1600 Oe/s is shown in Fig. 3(a). Again, this temperature change is sweep-rate independent. In this figure the very short overshoot at the beginning of the transition is merely connected with the inductive signal ($\sim dM/dt$) from the sample in the thermocouple loop. (This was proven by the measurements with the opposite sign of the magnetic field where this overshoot has the opposite sign and the same amplitude). The sample temperature is clearly seen to increase for approximately 5 ms and go back in 200 ms. A notable change of exponent at 60 ms is probably connected with the change in boiling regime on sample cooling back: from film to bubble boiling. In Fig. 3(b) the overheating process is shown in more detail along with the magnetization

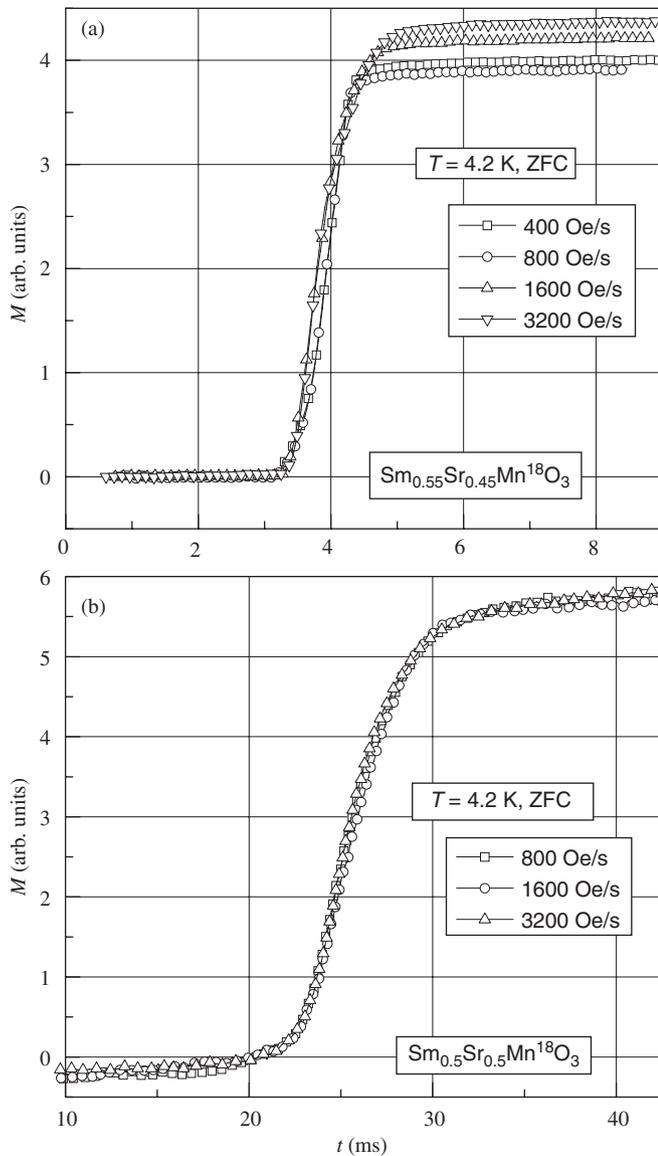


Fig. 2. Time dependence of the magnetization during the jump, obtained at different magnetic-field sweep rates.

transition (the overshoot signal is removed, the removed region marked by arrows; the magnetization is arbitrary scaled). Taking into account an imperfect thermal contact with the thermocouple, we can see very good coincidence of the magnetic transition and the temperature rise.

We suppose the following scenario for the step-like metamagnetic transition observed. Upon cooling, the AFM–FM competition results in a strongly disordered magnetic state, which has an excess specific heat [7]. FM ordering with the external magnetic field leads to the reduction of this extra entropy. In this case, the local release of the frozen entropy develops into the avalanche-like overheating of the sample because the temperature increase, in turn, tends to FM ordering [7]. So, both the magnetization and the temperature change in a jump-like way. This scenario assumes the time scale of the transition

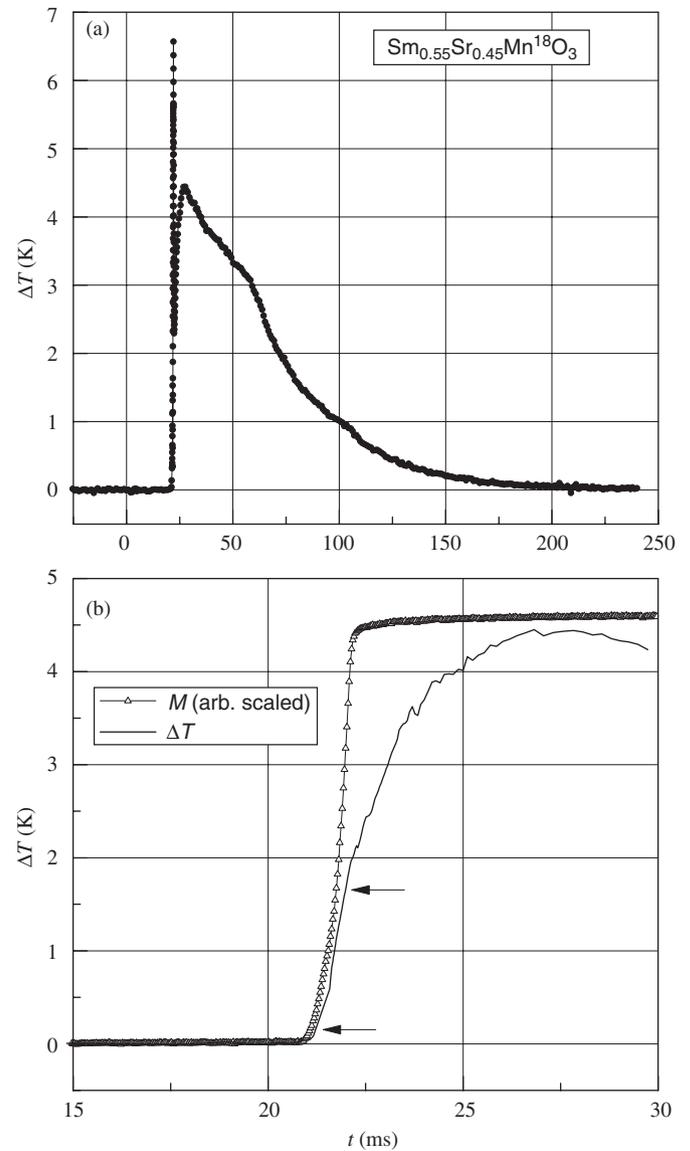


Fig. 3. Overheating of the sample during the magnetization jump. The lower panel shows simultaneous measurements of the temperature and magnetization (arbitrary scaled).

to be inversely proportional to the thermal conductivity of the sample. This is exactly the case if one compares $\text{Sm}_{1-x}\text{Sr}_x\text{Mn}^{18}\text{O}_3$ with $x = 0.45$ and 0.5 . The resistivity of $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{Mn}^{18}\text{O}_3$ is at least four orders of magnitude higher [10] so that the thermal conductivity should be lower. We recall that the observed time scale of the jump for $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{Mn}^{18}\text{O}_3$ is 10 times larger than for $\text{Sm}_{0.55}\text{Sr}_{0.45}\text{Mn}^{18}\text{O}_3$ (cf. Fig. 2(a) and (b)).

3. Conclusion

We have studied the magnetization jumps in $\text{Sm}_{1-x}\text{Sr}_x\text{Mn}^{18}\text{O}_3$ and related sample overheating upon the magnetic field induced AFM–FM transition. The sample overheating is shown to develop simultaneously

with the magnetization jump and culminates at $\Delta T = 4.5\text{K}$. The time scale of the jump (both in the magnetization and in temperature) is shown to be independent of the magnetic-field sweep rate. These means that the magnetization jump is not a magnetic field-driven process but a temperature-driven one. The overheating starts at the beginning of the magnetization change and stimulate the transition to the FM state what, in turn, results in the further release of heat. This latent heat seemingly originates from the frozen magnetic disorder (spin-glass like) and releases in the observed avalanche-like way.

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