Spin-glass behaviour and dynamical process of high field magnetisation in disordered perovskite manganites

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Abstract. In single crystals of A-site disordered perovskite manganites R₁₋ₓAEₓMnO₃ (R=Sm, Gd, Eu, AE=Ba, Sr, x=0.5, 0.45), we have observed the spin-glass behaviour. Applying pulsed high magnetic fields, a metamagnetic phase transition was observed in some samples, whereas in other samples, the magnetisation was just a smoothly varying function of magnetic field up to the saturation. The difference between the two types of behaviour seems to arise from the difference in the average ionic radius of the A-site ions and the disorder in the A-site layers. In the metamagnetic transition, magnetisation hysteresis was found to be smaller as the field sweep rate is faster, which is contrary to the ordinary spin relaxation phenomena.

1. Introduction

In recent years, much interest has been aroused in the perovskite manganites represented by a chemical formula R₁₋ₓAEₓMnO₃ (R and AE denote rare earth ions and alkaline earth ions, respectively) because of the rich phenomena originating from the coupling among charge, spin, orbital and lattice systems [1]. A variety of electric and magnetic phase transitions between different phases arise depending on temperature, species of ions in the R and AE sites and their concentration x. In a magnetic field, they show colossal magnetoresistance, metal-insulator transitions and other field-induced phase transitions. The phase transitions originate from the competition between the superexchange antiferromagnetic interaction accompanied by the long range orders such as the charge order or orbital order and the double exchange type ferromagnetic interaction. Akahoshi et al. recently demonstrated that the A-site disordered single crystals of half-doped manganite R₀.₅Ba₀.₅MnO₃ where the R ions and Ba ions in the A-site are randomly distributed have remarkably different properties from the A-site ordered crystals [2]. The random potentials due to the nano-scale disorder suppress the long range orders and spin glass-like behaviour is observed at low temperatures when the ionic radius of R is relatively small (R=...
It is of interest to investigate how the spin glass state is affected by the external field. Takeshita et al. observed the pressure-induced transition from the spin glass insulator to a ferromagnetic metal in disordered Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ [3]. In this paper we study the effect of high magnetic fields in A-site disordered perovskite manganites R$_{1-x}$AE$_x$MnO$_3$ (R=Sm, Gd, Eu, AE=Ba, Sr, x=0.5, 0.45) in steady and pulsed high magnetic fields.

2. Experimental

Single crystals of the A-site disordered samples were grown by the floating zone method at CERC. Magnetisation and magnetoresistance were measured in high magnetic fields generated up to 50 T by a pulsed magnet at IFW Dresden and up to 150 T by the single-turn coil technique at Berlin. The former and the latter systems use a capacitor bank of 1 MJ (10 kV) and 200 kJ (60 kV), respectively. There is a large difference in the duration time of the magnetic field between the two systems: it is 20 ms in the former case and 6.5 $\mu$s in the latter case. Thus we can investigate the magnetisation in different field intensities and also with different sweep rates. This is very convenient for studying the relaxation phenomena. The magnetisation was measured by the standard pick-up coil technique. Measurements in a steady field up to 14 T were made using a Quantum Design PPMS.

3. Results and discussion

All studied disordered crystals showed a cusp-like peak in the low field magnetisation vs. temperature curve at some freezing temperature $T_f$ with 40 K $\leq T_f \leq$ 50 K. The resistivity steeply increases with decreasing temperature. From these results we can deduce that the samples are in the spin glass-like insulating state below the temperature $T_f$. Figure 1 shows the magnetisation curve of Eu$_{0.5}$Ba$_{0.5}$MnO$_3$. The measurements were made up to 50 T using the non-destructive pulse magnet and up to 130 T using the single turn coil system. The data obtained by the two different magnet systems show an excellent agreement indicating the accuracy of the measurements. The magnetisation increases smoothly up to the nearly saturated value without any signature of the phase transition. However, we can see an almost linearly increasing part between 13 T and 24 T. This part can be more clearly defined by taking the derivative $dM/dH$,
in which distinct changes on both sides of the field range are seen. Similar behaviour was also observed in other samples with R = Gd and Sm, and a crystal with a little different composition Sm$_{0.55}$Ba$_{0.45}$MnO$_3$. Figure 2 depicts $dM/dH$ for Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ exhibiting a linear $dM/dH$ between 10 T and 20 T. No metamagnetic phase transition is observed in these crystals, and the magnetic field-induced alignment of spins takes place in a large range of fields.

In samples with Sr partly replacing Ba, Sm$_{1-x}$(Ba$_{1-y}$Sr$_y$)$_x$MnO$_3$ ($x$=0.5 with $y$ = 0.5 and 0.25; and $x$ = 0.45 with $y$ = 0.5), a clear metamagnetic phase transition was observed. Figure 3 shows the magnetisation curve in Sm$_{0.55}$(Ba$_{0.5}$Sr$_{0.5}$)$_{0.45}$MnO$_3$ with the freezing temperature $T_f$ =46 K. With increasing magnetic field, the magnetisation $M$ tends to saturate below 8 T at an intermediate value, followed by the metamagnetic transition. It should be noted that the metamagnetic transition was still observed at 72 K $\gg T_f$. As the temperature is raised, the hysteresis is decreased and vanishes at about 75 K. The increase of the magnetisation is accompanied by a huge reduction of the resistance showing the insulator-metal transition.

We can notice that between the samples which show the metamagnetic phase transition and those which show no transition but just the change of the slope of the magnetisation curve, there are significant differences in the averaged radius of A-site cations $<r_A> = (1-x)r_{R3+} + xr_{AE2+}$ and the variance of the ionic radii $\sigma^2 = \sum_x r_i^2 - <r_A>^2$. $<r_A>$ is a parameter characterizing the Mn-O-Mn bond angle, and it is well known for perovskite manganites that as $<r_A>$ increases, the crystal favors the ferromagnetic metallic state. The magnitude of the disorder of the samples, on the other hand, can be evaluated by the variance $\sigma^2$. Following Ref. [4], we plot the samples of this experiment in Fig. 4 showing $<r_A>$ as x-axis and $\sigma^2$ as y-axis. Note that along the diagonal from the lower left to the upper right corner of the diagram an increasing suppression of the charge and orbital order is expected. In the same figure, the field of the metamagnetic phase transition ($H_t$) and the upper field where the slope shows the abrupt change ($H_{t2}$, cf. Fig.2) are plotted as a function of $<r_A>$. From such a diagram, it can be deduced that large disorder suppresses the first order phase transition, whereas it occurs for those compounds situated closer to the lower left corner of the diagram. The possible presence of a charge-ordered phase which may cause the metamagnetic transition has been considered. The absence of a charge order transition in the temperature-dependent low-field magnetisation

![Figure 3](image1.png)  
Figure 3. Magnetisation curve for Sm$_{0.55}$(Ba$_{0.5}$Sr$_{0.5}$)$_{0.45}$MnO$_3$ indicating the metamagnetic transition at different temperatures.

![Figure 4](image2.png)  
Figure 4. Plot of the investigated samples on the plane of the variance $\sigma^2$ vs. $<r_A>$ (closed symbols). Index for each data point symbol indicates R($x$, $y$), where R is the rare earth ion of the sample. The transition field $H_t$ and the field where the slope of the magnetisation changes ($H_{t2}$ in the hatched area) are also plotted as a function of $<r_A>$ (open symbols).
Figure 5. Comparison of the magnetisation curve for Sm$_{0.55}$(Ba$_{0.5}$Sr$_{0.5})_{0.45}$MnO$_3$ measured in a superconducting magnet with a sweep rate of 0.012 T/s and in a pulse field with $\sim$5000 T/s.

Figure 6. Comparison of the $M - H$ curve for Sm$_{0.55}$(Ba$_{0.5}$Sr$_{0.5})_{0.45}$MnO$_3$ measured in short pulse field with a duration time of about $\Delta t = 6.5 \mu s$ and long pulse field with $\Delta t = 20$ ms.

has been carefully proved for one of the metamagnetic crystals. The observed tendency of $H_t$ and $H_{t2}$ to increase with $< r_A >$ is unexpected. It could be dominated by an effect of the increasing disorder in the samples and is subject to further studies.

Another interesting feature is the magnitude of the hysteresis. Figure 5 shows the comparison of the magnetisation curves measured in long pulse and steady fields. In the latter case, the transition takes place at a higher field and the hysteresis is larger. This is contrary to typical spin relaxation phenomena. When the field sweep rate is even faster, the transition field and the hysteresis show no significant sweep rate-dependence any more as shown in Fig. 6. Therefore, the involved relaxation time is longer than the ms range. The phenomenon is reminiscent of the recently found sharp jump of the magnetisation [5, 6]. In fact, we have observed a sharp jump in a certain range of the field sweep rate. The underlying magnetisation process is a current matter of research. The phenomenon is related to the potential barrier height the system must overcome to transit to the field-dependent ground state during the field sweep. In the slow sweep, gradual alignment of spins to the field will reduce the magnetic energy. This leads (i) to a lower energy difference between competing magnetisation states and may further lead (ii) to a higher potential barrier. The local release of thermal energy from an aligning cluster may provide the activation necessary for the alignment of adjacent spins/clusters. This can be more efficient for the fast sweep due to (i) and may even lead to an avalanche type behaviour. More details will be reported in a future paper.

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References

[1] Tokura Y 2006 Rep. Prog. Phys. 69 797.
[2] Akahoshi D, Uchida M, Tomioka Y, Arima T, Matsui Y and Tokura Y 2003 Phys. Rev. Lett. 90 177203.
[3] Takeshita N, Terakura C, Akahoshi D, Tokura Y and Takagi H 2004 Phys. Rev. B 69 180405.
[4] Tomioka Y and Tokura Y 2004 Phys. Rev. B 70 014432.
[5] Fisher LM et al. 2004 Phys. Rev. B 70 212411.
[6] Mahendiran R et al. 2002 Phys. Rev. Lett. 89 286602.