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Influence of pressure on magnetization and magnetostriction jumps in the manganite $(Eu_{1-x}Gd_x)_{0.6}Sr_{0.4}MnO_3$(x=0,0.1)

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Abstract. We have demonstrated the influence of external pressure on magnetization jump in Gd substituted $Eu_{0.58}Sr_{0.42}MnO_3$. The collapse of magnetization jump of the Eu based manganite caused by the applied pressure up to 1.2 GPa is explained by a suppression of the metastable blocked state, which is inherent to the phase separated manganite system exhibiting the magnetic avalanche. On the other hand, the magnetization jump of the Gd substituted sample is enhanced under the same pressure. To further examine the nature of the magnetization jump, the influence of pressure on the isothermal magnetostriction of the parent manganite is carried out. These findings indicate the crucial role on the abrupt transition played by the frozen phase separated phase.

In a recent study[1] on the bandwidth-temperature-magnetic field phase diagram of $RE_{0.55}Sr_{0.45}MnO_3$ with nearly half doping, the bandwidth was controlled by chemical substitution and hydrostatic pressure. In particular, near a critical pressure, the character of the ferromagnetic transition varies from the first to second order. The applied pressure gives rise to a larger bandwidth accompanied by a decrease of short range CO/OO fluctuations, resulting in weakened first order transition. In this paper, we demonstrate the effect of pressure on magnetization jump and its associated magnetostriction at low temperatures observed in Gd substituted $Eu_{0.6}Sr_{0.4}MnO_3$. The $Eu_{1-x}Sr_xMnO_3$ ($x \sim 0.4$ for polycrystalline samples) system is a spin-glass like insulator at low temperature because the substitution of Eu ion with smaller ion radius for La site gives rise to both narrow band width and an increase in the quenched disorder.[2, 3] It is well known that the former and latter parameters are controlled by the average value and the variance of the RE/AE ionic radii in $RE_{1-x}AE_xMnO_3$,respectively. Several recent researches for metamagnetic transitions of doped manganites have revealed that magnetic avalanches appear at low temperatures[4]. To account for this phenomenon, a martensitic model based on lattice mismatch between competing CO and FM phases has been proposed but questions have been raised as to the validity of such a scenario. Ghivelder et al., have observed a sudden temperature rise ($\sim$ 30 K) associated with the magnetic jumps, indicating a release of

large latent heat (or entropy) from frozen magnetic to FM ordered states. According to their findings,[5] after zero-field cooling, the manganite system showing low temperature magnetic avalanche reaches a highly blocked state with a small, and almost time independent fraction of FM, which is dispersed as isolated regions within a CO matrix. This blocked state is realized at low temperatures by large energy barriers separating the AFM-CO and FM phases and strains due to the lattice mismatch between the coexisting phases. Increasing the temperature under the applied field, the system is then transfered from the blocked to unblocked states, promoting a growth of the FM phase over the majority AFM-CO matrix.

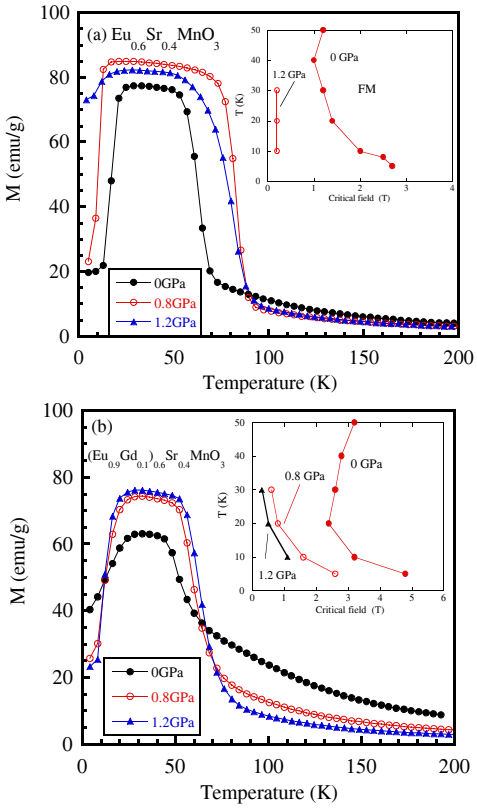

Figure 1. (a) Temperature variation of the zero field cooled (ZFC) magnetization of the $Eu_{0.6}Sr_{0.4}MnO_3$ bulk sample under 0 GPa, 0.8 GPa, and 1.2 GPa. The TH phase diagram of the parent sample is established from isothermal magnetization data after zero field cooling at selected temperatures as shown in the inset. H_a =1.5 T at 0 GPa, and 1 T both at 0.8 and 1.2 GPa. (b) ZFC magnetization curves of the $(Eu_{0.9}Gd_{0.1})_{0.6}Sr_{0.4}MnO_3$ bulk sample under 0 GPa, 0.8 GPa, and 1.2 GPa. H_a = 3T at 0 GPa, 1.5 T at 0.8 GPa, and 1 T at 1.2 GPa.

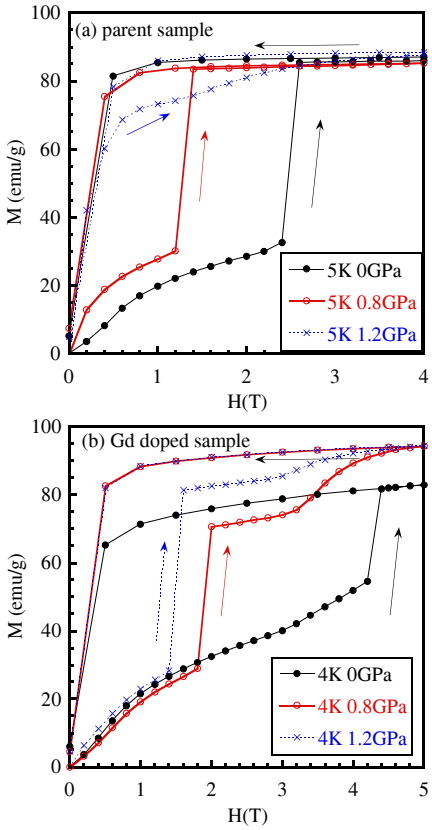

Figure 2. Isothermal magnetization curves measured under ambient and hydrostatic pressures after zero field cooling down to selected temperatures for both (a) $Eu_{0.6}Sr_{0.4}MnO_3$, and (b) $(Eu_{0.9}Gd_{0.1})_{0.6}Sr_{0.4}MnO_3$ bulk samples. For each measurement, a magnetic field is applied up to a maximum field at the sweep rate of 0.2 T/step.

Polycrystalline samples of $(Eu_{1-x}Gd_x)_{0.6}Sr_{0.4}MnO_3$ $(x = 0, 0.1)$ were prepared with a solid-state reaction method. The stoichiometric mixtures of Eu_2O_3, Gd_2O_3, $SrCO_3$, and Mn_3O_4 high

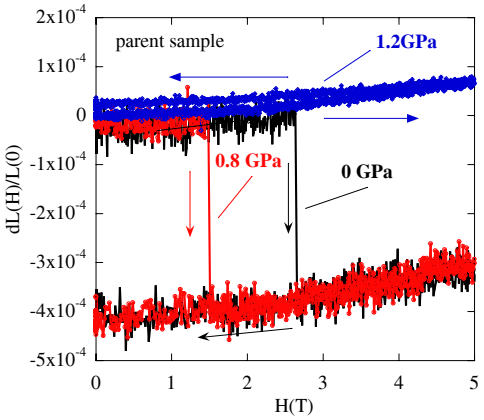

Figure 3. Isothermal magnetostriction $dL(H)/L(0)$ of the parent sample as a function of the applied pressure at 4.2 K.

purity powders were calcined in air at 1000 °C for 24 h and 1250 °C for 48 h. The products were then ground and pressed into cylindrical pellets. The pellets were finally sintered at 1350 °C for 36 h. X-ray diffraction data revealed that all samples are almost single phase with orthorhombic structures ($Pnma$). The lattice parameters of the parent sample ($x = 0$) are $a = 5.4424$ Å, $b = 7.664$ Åand $c = 5.4329$ Å, which is in fair agreement with a previous work.[2]

The magnetization measurement was carried out using commercial superconducting quantum interference device (SQUID) magnetometers both at Iwate Univ. and National Institute for Materials Science. Hydrostatic pressures in magnetization and magnetostriction measurements were applied by using a clamp-type CuBe cell up to 1 GPa. Magnetostriction was measured by using a superconducting magnet at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University.

First of all, let us show in Fig.1 the temperature variation of the zero field cooled (ZFC) magnetization of $(Eu,Gd)_{0.6}Sr_{0.4}MnO_3$ under ambient and applied pressures, to check the existence of a low temperature blocked state in the present sample. The critical field H_C at the corresponding temperatures is determined from the inflection points in isothermal magnetization curve upon increasing the applied field up to 5 T, indicating the metamagnetic transition. The negative curvature in dH_c/dT at low temperatures is an essential condition for the appearance of the magnetization avalanche as discussed in ref.[5]. As displayed in Fig.1(a), the ZFC magnetization curve shows a PM to FM transition near 60 K followed by a plateau, upon decreasing T, it then decreases rapidly until T ∼20 K, and finally remains a lower value down to

5 K. The application of hydrostatic pressure on the ZFC magnetization causes an enlargement of the FM region, resulting in a narrowed blocked region below 10 K. A volume fraction of the FM phase under pressure is estimated to be almost 97 % at the intermediate temperatures. For further applied pressure of 1.2 GPa, a low temperature blocked state is strongly suppressed and instead of it the FM phase still remains down to 4 K, indicating no steplike transition of the Eu based sample. Moreover, we attempt to measure the ZFC magnetization curve of the Gd substituted sample, to examine the influence of chemical pressure on the magnetic properties. For comparison, the critical field versus temperature boundary is plotted as a function of the applied pressure in the inset of Fig.1(b). We note that the HT phase boundary line of the Gd10% sample under 0.8 GPa is similar to that of the parent sample at ambient pressure.

Next, to examine the influence of lattice on magnetic avalanche of the bulk $Eu_{0.6}Sr_{0.4}MnO_3$, we attempt to measure the low-temperature magnetization as a function of field up to 5 T under ambient and applied pressures. At low temperatures, the magnetization jump appears just around 2.6 T upon increasing the applied field as shown in Fig.2. The application of external pressure on the magnetization depresses the critical field H_c from 2.6 T at 0 GPa down to $\sim$1.2 T at 0.8 GPa. Furthermore, the ultrasharp phase transition to the FM state disappears under an applied pressure of 1.2 GPa accompanied by a rapid suppression of magnetic hysteresis. This finding is well consistent with the low temperature MT curve showing no low temperature blocked state. To examine a close relationship between the lattice and spin coupling in the Gd substituted sample, we try to measure the effect of pressure on the magnetization curve of $(Eu_{0.9}Gd_{0.1})_{0.6}Sr_{0.4}MnO_3$. Upon lowering temperature down to 4 K, a steplike FM transition appears around 4.2 T at 0 GPa. The application of external pressure on the Gd doped sample enhances the steplike transition as depicted in Figs. 2(b). Figure 3 shows the effect of external pressure on the magnetostriction of the parent sample which is well consistent with that on the isothermal magnetization. In particular, the value of $dL(H)/L$ under an application of 1.2 GPa remains almost zero upon increasing the applied field up to 7 T and exhibits no magnetic hysteresis, indicating a spontaneous transition of the system to the ferromagnetic state before the application of a magnetic field. Our data are in fairly agreement with previous results that a negative magnetovolume effect disappears above 1.15 GPa.[6] In summary, we have shown the influence of external pressure on the magnetization jump and its concomitant magnetostriction in the polycrystalline $(Eu,Gd)_{0.6}Sr_{0.4}MnO_3$. A suppression of the metastable blocked state under pressure leads to lowering the critical field. Our data indicate the importance of the frozen phase segregated state in the doped manganite system exhibiting the magnetic avalanche.

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