Pressure induced transition from a spin glass to an itinerant ferromagnet in half doped manganite $Ln_{0.5}Ba_{0.5}MnO_3$ ($Ln=$Sm and Nd) with quenched disorder

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The effect of quenched disorder on the multiphase competition has been investigated by examining the pressure phase diagram of half doped manganite $Ln_{0.5}Ba_{0.5}MnO_3$ ($Ln=$ Sm and Nd) with A-site disorders. Sm$_{0.5}$Ba$_{0.5}$MnO$_3$, a spin glass insulator at ambient pressure, switches to a ferromagnetic metal with increasing pressure, followed by a rapid increase of the ferromagnetic transition temperature $T_C$. The rapid increase of $T_C$ was confirmed also for Nd$_{0.5}$Ba$_{0.5}$MnO$_3$. These observations indicate that the unusual suppression of the multicritical phase boundary in the A-site disordered system, previously observed as a function of the averaged A-site ionic radius, is essentially controlled by the pressure and hence the band width. The effect of quenched disorder is therefore much enhanced with approaching the multicritical region.

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One of the hallmarks of strongly correlated electron physics is the multiphase competition produced by a delicate interplay among charge, spin and orbital degrees of freedom. An appealing example of such multicriticality can be seen in perovskite manganites. Competing interactions/orders inherent in the manganites, such as double-exchange ferromagnetism vs. super-exchange antiferromagnetism and charge-orbital order vs. metallic state, tend to produce the multicritical state where external stimuli occasionally cause the dramatic phase conversion, e.g., between a metal and an insulator, or between a ferromagnet and an antiferromagnet. The colossal magnetoresistance (CMR) phenomena are believed to be a consequence of such phase competitions.

Recently, the vital role of quenched disorder in the critical region of manganites has been attracting considerable interest. The quenched disorder may result in a phase separation into the competing two ordered phases on various time-scales and length-scales, which substantially modifies the criticality and hence the response to the external field. The first suggestion came from the observation of relaxor-like behavior induced by Cr-doping on the Mn sites in the charge/orbital ordered (CO/OO) state. This is highly likely associated with the ferromagnetic (FM) metallic clusters coexisting with the background CO/OO state. The effect of quenched disorder on the A-site was subsequently examined, which shows a drastic modification of the phase diagram as a function of the average A-site ionic radius.

The half-doped manganites $Ln_{0.5}Ba_{0.5}MnO_3$ ($Ln=$ Lanthanides) are one of the ideal systems to explore the physics of quenched disorder. The perovskite manganites in this chemical composition have two possible forms of the crystal structure depending on synthetic condition. One is with the A-sites randomly occupied by $Ln^{3+}$ and $Ba^{2+}$ ions. The melt-quenched sample shows the complete solid solution of Ln and Ba ions on the A-sites with a simple cubic structure as the average structure. The other is the A-site ordered perovskite with the alternate stack of $LnO$ and $BaO$ sheets along the c-axis with intervening MnO$_2$ sheets. This is due to the large difference in the ionic radii of $Ln^{3+}$ and $Ba^{2+}$. The respective MnO$_2$ sheets in this tetragonal form are free from random potential which would otherwise arise from the Coulomb potential or local strain from the random occupation of $Ln/Ba$ on the A-sites.

The phase diagram of ordered $Ln_{1/2}Ba_{1/2}MnO_3$ as a function of ionic radius is schematically shown by the dotted line in Fig. 1. The increase of ionic radius results in the reduction of Mn-O bond bending and hence the increases of the band width. With relatively small Ln ions from Dy to Sm, the CE-type charge ordered state with diagonal orbital stripes develops at low temperatures. With larger Ln ions from Nd to La, on the other hand, the ferromagnetic metal phase and, eventually, the A-type antiferromagnetic phase with ordering of $d_{x^2-y^2}$ orbitals are stabilized upon cooling the sample. These phases meet with each other at around $Ln=Nd$, forming a well defined multicritical point. When the solid-solution of $Ln^{3+}$ and $Ba^{2+}$ is formed, however, this multicritical region is drastically altered. The solid line in Fig. 1 indicates that the phase diagram of A-site disordered $Ln_{0.5}Ba_{0.5}MnO_3$. It is clear that the phase transitions near the multicritical point are suppressed significantly. In contrast to the ordered $Ln_{0.5}Ba_{0.5}MnO_3$, the charge
and orbital ordered phase goes away and alternatively a spin glass phase without long-range order in any charge and orbital sectors emerges. With increasing the ionic radius, the ferromagnetic metal phase takes over the spin glass phase around Nd. The A-type antiferromagnetic phase, however, does not show up at low temperatures. Besides, as compared with the A-site ordered systems, the ferromagnetic phase extends to the smaller ionic radius and the ferromagnetic transition temperature, $T_C$, is appreciably suppressed near the critical region.

The contrast between the A-site ordered and the disordered systems demonstrates a substantial influence of quenched disorder on the multiphase competition in the manganites. However, it should be emphasized here that the control parameter of the disordered system is not only the bandwidth but also the degree of disorder. By changing the ionic radius and the ferromagnetic transition temperature, $T_C$, is appreciably suppressed near the critical region. Alternately, it may be understood in terms of the increased disorder with decreasing the ionic radius of $Ln$ ions from those comparable with large Ba ions.

To separate the two contributions arising from the disorder and the bandwidth, we have employed hydrostatic pressure to modify the bandwidth alone, while keeping the degree of disorder constant. We have selected two A-site disordered compounds, Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ and Nd$_{0.5}$Ba$_{0.5}$MnO$_3$, and applied hydrostatic pressures up to 10 GPa. We found a pressure induced spin glass insulator to ferromagnetic metal transition, which can be mapped onto the phase diagram obtained by examining the solid-solution effect. This clearly demonstrates that the unusual phase diagram of disordered Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ arises from the bandwidth.

The A-site solid-solutions of $Ln_{0.5}$Ba$_{0.5}$MnO$_3$ ($Ln =$ Sm and Nd) were grown by the floating-zone method in a single crystalline form. The starting polycrystalline rods were prepared by a standard solid-state reaction. The x-ray powder diffraction patterns of the resultant crystalline rods indicate the formation of single phase $Ln_{0.5}$Ba$_{0.5}$MnO$_3$ without any evidence for the A-site ordering. The resistivity and the magnetization data of disordered Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ and Nd$_{0.5}$Ba$_{0.5}$MnO$_3$, at ambient pressure, are shown in the right panel of Fig. 1. The resistivity of disordered Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ shows an insulating behavior down to the lowest temperature measured and no evidence for the long-range charge order-
is a characteristic behavior of the transition to a ferromagnetic metal. This clearly indicates that the system has experienced a pressure induced spin glass insulator to a ferromagnetic metal transition. Above the critical pressure, the transition temperature, $T_c$, goes up very rapidly with increasing pressure. These behaviors can be visually summarized as a phase diagram in Fig. 3. We immediately notice that the phase changes as a function of pressure are essentially the reproduction of those with the average ionic radius on the A-site.

It is now clear that the dominant controlling factor of the unusual phase diagram in disordered $Ln_{0.5}Ba_{0.5}MnO_3$ is the bandwidth. Then, the pressure dependence of transition temperatures in disordered $Nd_{0.5}Ba_{0.5}MnO_3$ should be connected to that of $Sm_{0.5}Ba_{0.5}MnO_3$ by shifting the pressure axis. The bottom panel of Fig. 2 indicates the evolution of the temperature dependent resistivity and the AC susceptibility with increasing pressure for the A-site disordered $Nd_{0.5}Ba_{0.5}MnO_3$. $T_C$ defined as a temperature of resistivity anomaly shows a rapid increase with increasing pressure. At high pressures, the resistivity anomaly tends to become weaker, which make the determination of $T_C$ ambiguous. In the AC susceptibility, however, we are able to see the well defined anomalies associated with the ferromagnetic transition (indicated by arrows), which provides us with a solid estimate of $T_C$. Thus obtained pressure dependence of $T_C$ is plotted in Fig. 3. By shifting by 10 GPa, the $T_C$-P curve of disordered $Nd_{0.5}Ba_{0.5}MnO_3$ seems to be smoothly connected to that of $Sm_{0.5}Ba_{0.5}MnO_3$. This provides a further evidence for

![FIG. 2: Temperature dependent resistivities under various pressures for A-site disordered $Sm_{0.5}Ba_{0.5}MnO_3$ (top) and $Nd_{0.5}Ba_{0.5}MnO_3$ (bottom). The inset for $Nd_{0.5}Ba_{0.5}MnO_3$ indicates the AC susceptibility data for the same sample.](image)

![FIG. 3: Pressure phase diagram of A-site disordered $Sm_{0.5}Ba_{0.5}MnO_3$ and $Nd_{0.5}Ba_{0.5}MnO_3$, determined from the resistivity and the magnetic susceptibility data shown in Fig. 2. The pressure axis for $Nd_{0.5}Ba_{0.5}MnO_3$ is shifted by 10 GPa.](image)
the dominance of pressure as the controlling parameter.

Another important point seen in Fig. 2 is that $T_C$ of Nd$_{0.5}$Ba$_{0.5}$MnO$_3$ tends to saturate with increasing pressure. In the saturation region, $T_C$ recovers to almost 300 K, which is close to those observed for ordered Ln$_{1/2}$Ba$_{1/2}$MnO$_3$ with Ln = Nd and Pr. These observations strongly suggest that the suppression of $T_C$ is much reduced once away from the critical region. Note again that the degree of disorder is kept constant in this experiment. We therefore conclude that the effect of quenched disorder is substantially enhanced when coupled with the (multi)criticality.

Recently, it was pointed out that the quenched disorder triggers a phase separation into ferromagnetic and charge ordered domains [13]. In their scenario, the formation of a clustered state with randomly oriented ferromagnetic clusters results in the decreases of the clean limit $T_C$ to a much lower $T_C$ in the ferromagnetic region. This accounts for the anomalous suppression of $T_C$ with approaching the critical region [14]. More recent works claim that the state below the clean limit $T_C$ is not statically clustered but homogeneous state with substantial critical charge/lattice fluctuations induced by quenched disorder [15]. Present pressure measurement alone cannot specify whether the phase separation is static or dynamic.

In summary, we have observed a pressure-induced switching of a spin glass into a ferromagnet in a half-doped manganite Sm$_{0.5}$Ba$_{0.5}$MnO$_3$ with quenched disorder, which is followed by a rapid increase of Curie temperature upon pressure. The remarkably rapid increase of Curie temperature was confirmed also in inhomogeneous multi-phase competing systems.

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