Entropy-Driven Reentrant Behavior in CMR Manganites

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Abstract

We discuss the origin of the reentrant behaviors of insulating states above the Curie temperature of the colossal magnetoresistance manganites. We consider a system where charge ordering and ferromagnetism compete with each other. In the presence of randomness which pins charge order fluctuations, entropy-driven reentrant behaviors will appear, which explains the typical temperature dependence of the resistivity for CMR manganites.

Key words: CMR, disorder-induced insulator-to-metal transition, reentrant

1. Introduction

One of the key features of the colossal magnetoresistance (CMR) phenomena in manganites is that the resistivity shows an insulating behavior in the paramagnetic regime above the Curie temperature \(T_C\) and a metallic behavior as ferromagnetic moment appears below \(T_C\). By applying an external magnetic field, appearance of the magnetic moment shifts to higher temperatures and the insulating behavior is replaced by the metallic one, which creates CMR.

Experimentally, such behavior has been shown to be closely related to charge ordering (CO) fluctuations. Using neutron scattering measurement, Dai et al. [1] observed the existence of CO fluctuations above \(T_C\) which suddenly disappear below \(T_C\) for the CMR compounds. Note that, for the compounds with less carrier concentrations which do not show CMR but are insulating to the lowest temperatures, CO fluctuations develop to make a long-range order (LRO) as temperature decreases. It has been shown that the temperature dependence of the CO fluctuations are closely related with that of the resistivity.

One of the questions which arises from these experimental results is, provided CO fluctuations are enhanced at higher temperatures above \(T_C\), why they are disfavored to make LRO, and instead are taken over by ferromagnetic metal (FM) phase. In order to clarify the origin of CMR, it is crucial to understand this ‘reentrant’ behavior. Similar phenomena, where reentrant behavior from CO to FM are observed as decreasing temperature under a magnetic field, have also been reported in \((\text{Pr,Ca,Sn})\text{MnO}_3\) [2].

Recently, \(A\)-site ordered/disordered samples have been synthesized to make ‘disorder-control’ [3]. In \(A\)-site disordered samples which exhibit CMR, Tomioka et al. [4] observed the similar behavior of CO fluctuations above \(T_C\) using X-ray scattering and Raman measurements. Meanwhile, in \(A\)-site ordered sam-
samples which show the bicritical phase diagram without CMR, the reentrant behavior is absent. Thus we see that randomness plays an essential role to control CMR and reentrant behavior.

The authors have studied the extended double-exchange system where FM and CO compete in the presence and absence of quenched randomness, using the Monte Carlo technique [5]. Various phase diagrams as well as disorder-induced insulator-to-metal transition phenomena have been observed. In this paper, we further discuss the nature of the competition between FM and CO in various temperature regions, and investigate the origin of the reentrant behavior of CO fluctuations.

2. Entropy-Driven Reentrant Behavior

Let us consider a CO region with insulating behavior close to the phase boundary to FM region in the absence of disorder. By adding randomness, CO LRO corrupts and becomes short-range order (SRO). This is due to the anti-phase pinning of the order parameter as depicted in Fig. 1. The randomness for conduction electrons corresponds to a ‘random field’ to the commensurate CO LRO.

In other words, CO with a solid LRO is energetically unfavorable against random pinning potentials. If it is close to the phase boundary, FM becomes relatively stabilized. Due to the competition between these phases, CO disappears and turns into FM at low temperatures in the presence of randomness. We consider that this is the origin of the disorder-induced insulator-to-metal transition.

At the same time, as shown in Fig. 1, the position of domain boundaries can shift without any energy cost, which means that CO-SRO states have large entropy. Therefore, at higher temperatures, CO-SRO states will be favored by the large entropy due to CO domain configurations, which explains the enhancement of CO fluctuations observed in our previous report.

This results in an entropy-driven reentrant behavior, i.e., CO fluctuations become maximum above the FM phase. Such reentrant behavior of the CO fluctuations is accompanied by the maximum of the resistivity due to the pseudo-gap structures in the density of states as well as the optical conductivity which has been reported previously.

At $T \sim T_C$ where maximum competitions between CO and FM exist, a weak magnetic field stabilizes FM which substantially reduces the resistivity. We consider that the competition between CO and FM, as well as entropy-driven CO-SRO state above FM phase, is essential to CMR phenomena.

3. Discussion

Our proposal for the mechanism of CMR emphasizes the roles of spacial CO fluctuations with domain structures. Such physics will not be captured by local approximations and mean-field treatments. In various studies using single-site dynamical mean-fields, for instance [6,7], possibilities for LRO of CO or polaron-lattices have been neglected within theoretical frameworks. Namely, by not taking into account CO mean-fields, CO LRO is artificially suppressed within the self-consistent treatments. Although the results of these calculations in the absence of randomness resemble the experimental results of disordered samples, they cannot reproduce the systematic changes between $A$-site ordered and disordered samples.

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