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Griffiths phase and temporal effects in phase separated manganites

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ABSTRACT

Phenomenological description of relaxation phenomena in magnetic and transport properties of perovskite manganites has been presented. The approach is based on generalization of some hypotheses appropriate to the Preisach picture of magnetization process for half-metallic ferromagnets and on an assumption that in doped manganites the phase separated state exists near the magnetic ordering temperature. For systems with the percolation type of a ferromagnet–paramagnet transition, distinctive features in relaxation of magnetization and resistivity have been found. The relaxation is shown to be most pronounced near the transition temperature, and to be an approximately logarithmic function of time. The theoretical results replicate a broad spectrum of behavior observed experimentally on time dependence of magnetization and resistivity of CMR systems and allow a direct comparison with available experimental data. We propose an additional experimental test to distinguish between the percolation scenario of magnetic and transport transitions in doped manganites, and the ferromagnetic polaron picture. In particular, an anomalously slow relaxation to zero of the order parameter can be considered as a key feature of the Griffiths-like phase transition in doped manganites. It is also shown that a system with the Griffiths-like state will exhibit nonequilibrium aging and rejuvenation phenomena, which in many aspects resemble that of a spin glass. We hope that experimental observation of a set of time decay properties will provide a settlement of apparently conflicting results obtained for different characteristics of phase-separated manganites.

1. Introduction

Despite intense effort towards understanding the physics of manganites, many macroscopic features of their phase separated (PS) state, including its thermodynamic properties and dynamic behavior, still remain to be studied in more detail. One of the most interesting features of the PS state is the entwining between its dynamic and static properties. Numerous experimental data directly point to the fact that magnetization $M(H,T)$ and resistivity $\rho(H,T)$ are rather sensitive to the magnetic field (H) and temperature (T) sweeping rate. The mixed-valence manganites often display slow relaxation features, such glass like dynamic effects as aging and rejuvenation that hide experimentally the real equilibrium thermodynamic state of the system [1–10].

Whereas the phase separation scenario is generally accepted (an extensive review of all aspects of phase separation in manganites can be found in [11]), the origin of colossal magnetoresistance (CMR) in manganites is still under discussion. At present there are two competing opinions. Salamon et al. were the first who argued that the Griffiths-like phase and percolation type of metal–insulator (MI) transition could be a true theoretical ground for physics of manganites [12]. According to the opposite point of view, the Griffiths phase itself is insufficient for the appearance of CMR, and the formation of ferromagnetic polarons just above T_C causes unique physical characteristics of doped manganites [13,14].

Let us recall that the Griffiths phase (GP) was first proposed by Griffiths to explain the effects of quenched randomness on the magnetization of a diluted Ising ferromagnet [15]. The GP means the existence of short-range ordering of ferromagnetic clusters in paramagnetic matrix in the temperature region $T_C(p) < T < T_G$. Here T_G stands for the ordering temperature of undiluted or homogeneous system $T_G = T_C(p=1)$ and the latter is known as the Griffiths temperature, while $T_C(p)$ is the Curie temperature of a diluted system; p is the probability of the nearest-neighbor exchange bonds to occur. For $p < p_c$ (p_c stands for percolation threshold) no long-range ferromagnetic order is established in the system. In the GP, though the system does not hold spontaneous magnetization, there exist spatially distributed regions that are devoid of disorder forming finite size clusters having ferromagnetically correlated spins. As a direct consequence of limitation of the magnetic correlation length ξ to the finite size of the clusters (ξ does not diverge at the critical point) the magnetization fails to be an analytical function at the critical point. Bray and

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Moore [16] generalized this argument for any bond distribution (instead of bonds having strength of only J and 0, as discussed by Griffiths [15]) which reduces the long-range ordering transition temperature. It is generally accepted at present that quenched disorder is a prerequisite for the formation of the GP (for more details, see review [17] and references therein).

Concerning doped manganites, in pioneering works of Salamon et al. [12], based on their results of magnetic and heat capacitance measurements, the 'Griffiths phase' paradigm in physics of these compounds was introduced. Indeed, in a manganite with a generic formula $R_{1-x}A_xMnO_3$ (where R is a trivalent rare earth and A is a divalent alkaline-earth ion) among the various forms of inhomogeneous phases, preformation of ferromagnetic (FM) clusters much above the FM long-range ordering temperature (T_C) was detected in different experiments: neutron scattering [18], ferromagnetic resonance [19], magnetic susceptibility measurements [12,20], etc. (see also recent review on this topic [21]). At present, it is widely accepted that a system which demonstrates CMR is intrinsically inhomogeneous. Even in the best crystals, a competition between different phases results in the electronic PS state [22]. Authors [12] attributed these to the existence of the Griffiths-like phase in doped manganites. Accordingly, the MI transition is achieved due to percolation, i.e., following the Griffiths-like scenario, and theoretical model of the percolated MI transition was suggested to explain the CMR effect [23]. Namely, above T_C, the FM metal clusters are isolated and a system is in insulating state. As temperature decreases, these isolated FM clusters increase in size and at T_C the percolated channel forms. After that, the relevance of the Griffiths-like phase to colossal magnetoresistance of manganites was addressed in a number of reports [24].

Yet, applicability of the percolate phase transition scenario to CMR systems was addressed in a number of reports [13,14], too. It is argued, in particular, that the unique physical characteristics of doped manganites in a temperature region above the Curie temperature T_C are, most likely, due to ferromagnetic polarons, concentration of which increases upon cooling towards the Curie temperature [25]. Indeed, a strong electron–phonon coupling due to the Jahn–Teller effect of Mn^{3+} ion was shown to play a key role in CMR behavior and, as expected, is able to facilitate the formation of polarons [26]. The existence of polarons has been extensively confirmed experimentally [27]. It was argued that correlated polarons [28] may contribute to the nanometer-scale phase separation (see also [10] and references therein). Transition from itinerant large polarons to localized small polarons was proposed to interpret the MI transition associated with the ferromagnet–paramagnet (FM–PM) transition in the CMR materials. (Models of polaron transport are discussed in detail in review [29].)

In this report, we address the question whether these two scenarios of FM–PM phase transition, the percolation type (Griffiths-like) and the second-order-like, can be distinguished by their temporal behavior. The available data point out that temporal effects in the phase-segregated state can fundamentally differ from those in the state of large (or small) localized polarons. In particular, the Griffiths-like state is characterized by anomalously slow temporal evolution (relaxation) of the order parameter [30] and, thus, appears to be particularly favorable for the existence of out-of-equilibrium features. The competition between the coexisting phases opens the possibility for the presence of locally metastable states, giving rise to such interesting time dependent effects as cooling rate dependence [9], relaxation [1–8,31] giant $1/f$ noise [32], two-level fluctuations [33], etc. [34]. The similarity between phase-segregated manganites and glassy systems was also suggested [35]. The extremely slow relaxation observed in all the above-mentioned experiments opens an interesting question about temporal phenomena in a system in the GP. As far as the authors know, a clear understanding of the system's slow dynamic behavior in the GP is still lacking, and in this report, we try to address some open questions.

Systematic studies of the resistivity magnetization relation in doped manganites below and above the Curie temperature reveal a strong interplay between transport and magnetism in this system. This experimental fact was used as a physical basis of the model [36] which considered direct relationships between magnetization $M(H,T)$, a function of magnetic field H and temperature T, and magnetoresistivity $\rho(H,T)$. Mathematically the model is based on utilization of some hypotheses appropriate to the Preisach picture of the magnetization process [37] (see also textbook [38]). One of the remarkable features of the Preisach-based approach is that it yields joint description of hysteresis and thermal relaxation based on a few simple assumptions common to both aspects of the phenomenon [39,38,40]. We embraced this opportunity to address the temporal effects for a system in the PS state.

The present paper discusses modeling of the temporal effects that leads to time dependence of magnetic and transport properties within the percolation scenario (Griffiths-like phase), and within the (conventional) FM–PM second-order-like transition. We try to replicate a broad spectrum of behavior observed experimentally on time dependence of magnetization and resistivity of CMR systems. The approach is based on generalization of some hypotheses appropriate to the Preisach picture of the magnetization process for half-metallic ferromagnets [36] and on the assumption that in doped manganites the PS state exists near the magnetic ordering temperature. For the systems under consideration, the noticeable relaxation of magnetization and resistivity has been found. The relaxation is shown to be most pronounced near the transition temperature, and to be approximately a logarithmic function of time. The theoretical results allow a direct comparison with the experimental data. We propose an additional experimental test to distinguish between the percolation scenario of magnetic and transport transition in doped manganites, and the ferromagnetic polaron picture. In particular, we show that a system with Griffiths-like state will exhibit nonequilibrium aging and rejuvenation phenomena, which in many aspects resemble those of a spin glass. We believe the experimental observation of a set of time decay properties will provide a settlement of apparently conflicting results obtained for different characteristics of PS manganites.

The main part of the paper is organized as follows. In Section 2 the phenomenological model of the MI transition and magnetoresistivity of doped manganites is briefly outlined. For the sake of simplicity, we will consider the case when the PS state is formed by two phases, the FM phase with metal type conductivity and the PM one with polaron transport. A key feature introduced here is the interplay between the external magnetic field changes and the thermal relaxation effects that determines the time evolution of the system. In Section 3 the results of numerical simulations for a system with a second-order-like FM–PM phase transition and for a system with the Griffiths-like scenario FM–PM transition are presented. Our main goal is to reveal the effects related to the field or temperature sweeping time parameter t_{exp}. In the next section, we discuss the results obtained and the available experimental data on temporal phenomena in doped manganites, and summarize our main results.

2. The model

Let us briefly outline the model used (for details, see, Refs. [36]).