MAGNETISM

Fluctuations of the Order Parameter in $R_{0.55}Sr_{0.45}MnO_3$ Manganites near the Metal–Insulator Phase Transition

F. N. Bukhanko* and A. F. Bukhanko

Donetsk Institute for Physics and Engineering named after O. O. Galkin, National Academy of Sciences of Ukraine, ul. Rozy Lyuksemburg 72, Donetsk, 83114 Ukraine

*e-mail: buhanko@mail.fiz.ac.donetsk.ua

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Abstract—The magnetic phase transformations induced by changes of the composition, external magnetic field strength, and temperature in manganites with a nearly half-filled conduction band in the vicinity of the metal–insulator phase transition have been investigated experimentally. It has been found that the substitution of rare-earth ions (Sm) for Nd ions with a larger ionic radius in $R_{0.55}Sr_{0.45}MnO_3$ manganites leads to a linear decrease in the Curie temperature $T_C$ from 270 to 130 K and a transformation of the second-order ferromagnetic (FM) phase transition into a first-order phase transition. The results of measurements of the alternating-current (ac) magnetic susceptibility in the $(Nd_{1–y}Sm_y)_{0.55}Sr_{0.45}MnO_3$ system indicate the existence of a Griffiths-like phase in samples with a samarium concentration $y > 0.5$ in the temperature range $T_C < T < T^*$ (where $T^* ≈ 220$ K). For samples with $y > 0.5$, the magnetization isotherms at temperatures above $T_C$ exhibit specific features in the form of reversible metamagnetic phase transitions associated with strong fluctuations of the short-range ferromagnetic order in the system of Mn spins in the high-temperature Griffiths phase consisting of ferromagnetic clusters. According to the results of measurements of the ac magnetic susceptibility in the $(Sm_{1–y}Gd_y)_{0.55}Sr_{0.45}MnO_3$ system for a gadolinium concentration $y = 0.5$, there is an antiferromagnetic (AFM) phase with an unusually low critical temperature of the spin ordering $T_N ≈ 48.5$ K. An increase in the external static magnetic field at 4.2 K leads to an irreversible induction of the ferromagnetic phase, which is stable in the temperature range 4.2–60 K. In the temperature range 60 K $< T < 150$ K, there exists a high-temperature Griffiths-like phase consisting of clusters (correlations) with a local charge/orbital ordering. The metastable antiferromagnetic structure is retained in samples with gadolinium concentrations $y = 0.6$ and 0.7, but it is destroyed with a further increase in the gadolinium concentration upon the transition to the spin-glass state. The magnetization isotherm obtained with variations in the external static magnetic field in the field range ±70 kOe at 4.2 K and the temperature dependence of the ac-magnetic susceptibility $\chi$ suggest that, in the $Gd_{0.55}Sr_{0.45}MnO_3$ ceramics, there is a mixed two-phase low-temperature state consisting of the quantum Griffiths phase with a characteristic divergence of $\chi(T)$ near $T = 0$, which was embedded in the spin-glass matrix with the spin “freezing” temperature $T_G ≈ 42$ K. The low-temperature state with quantum fluctuations exists in the $(Sm_{1–y}Gd_y)_{0.55}Sr_{0.45}MnO_3$ system for $y ≥ 0.5$.

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1. INTRODUCTION

Phase transformations and critical phenomena in manganites $A_{1–x}A’_xMnO_3$ ($A = $ La, Pr, Nd, Sm; $A’ = $ Ca, Sr, Ba) with a nearly half-filled conduction band ($x ≈ 0.5$) were considered earlier in the review by Tokura [1]. It was shown that a wide variety of electronic and magnetic properties of these compounds is determined by the competition between the metallic ferromagnetic (FM) phase and the insulating antiferromagnetic (AFM) phase (usually, the CE type) with the charge/orbital (CO/OO) ordering. The main parameter that controls the competition of these two phases is the energy of charge transfer $e_r$ by electrons or, in other words, the width of the conduction band $W$, which is determined by the average radius $\langle r_A \rangle$ of the rare-earth ion substituting for lanthanum in the $A$-positions of the perovskite matrix. A change in the ionic radius $r_A$ leads to a bicritical phase diagram [2], in which the ferromagnetic phase (large ionic radii $r_A$) is separated from the antiferromagnetic phase (small ionic radii $r_A$) by a first-order phase transition. A disorder arising from local distortions of $A$-positions upon a random substitution of cations with a small ionic radius for cations with a large ionic radius (quenched disorder) can significantly change the phase diagram and magnetoelectronic properties in the vicinity of the bicritical point at the boundary between two competing phases. The behavior of manganites with a weak disorder near the bicritical point was theoretically considered by Murakami and Nagaosa [3]. Characteristic features that should be observed in the experiment near the bicritical point were predicted within the RG-model including ther-
mal and quantum fluctuations near the boundary between two phases, which corresponds to the critical width of the conduction band $W_c$. These effects occur because of a giant fluctuation of several (ferromagnetic, antiferromagnetic, orbital, and charge) order parameters due to the bicriticality which drastically increases near the boundary between two phases. As was shown in a number of theoretical studies [4–7], the phase diagram becomes asymmetric in manganites even with a moderate disorder. In this case, the ferromagnetism is suppressed but continues to exist at finite temperatures, whereas the CO state disappears and, instead, at low temperatures, there arises a glass state with the spin “freezing” temperature $T_G$, which is independent of the further decrease in the average ionic radius $\langle r_A \rangle$.

Based on numerous experimental results, Tomioka and Tokura [8] constructed an extended phase diagram of $R_{0.55}A_{0.45}MnO_3$ manganites with a weak structural disorder, in which the critical temperatures of the electronic and magnetic phase transitions are given as functions of the average cation radius $\langle r_A \rangle$ in $A$-positions in the radius range 1.26–1.40 Å. The phase diagram is separated into two equal parts with respect to the bicritical point corresponding to the average radius $\langle r_A \rangle_{cr} \approx 1.33$ Å: at $\langle r_A \rangle \leq \langle r_A \rangle_{cr}$, the phases with charge and antiferromagnetic spin ordering are stabilized in the low-temperature range, whereas at $\langle r_A \rangle \geq \langle r_A \rangle_{cr}$, the metallic ferromagnetic phase dominates.

The manganites with a strong structural disorder are characterized by an asymmetric phase diagram: the ferromagnetism is suppressed but continues to exist at finite temperatures, whereas the CO state disappears and, instead, at low temperatures, there arises a glass state. According to the electronic phase diagram constructed in [9] for manganites $R_{0.55}Sr_{0.45}MnO_3$ ($R = Pr$, Nd, Sm, Eu, Gd) with a strong structural disorder, the critical value of the average cation radius $\langle r_A \rangle_{cr}$ $\approx 1.33$ Å corresponds to the $Sm_{0.55}Sr_{0.45}MnO_3$ compound. The compounds $Sm_{1-x}Sr_xMnO_3$ exhibit some unusual properties, especially in the range of Sr critical concentrations $0.4 \leq x \leq 0.5$. They have a relatively low Curie temperature of the transition to the ferromagnetic state $T_C \sim 130$ K and possess a colossal magneto- resistance near $T_C$.

The phase transition between the metallic ferromagnetic state and the insulating state with strong short-range charge/orbital correlations was investigated in $(Sm_{1-x}Gd_x)_{0.55}Sr_{0.45}MnO_3$ ($0 \leq x \leq 1$) single crystals [9]. It was found that, owing to local lattice distortions, the structural disorder increases the electronic and magnetic fluctuations and prevents the formation of a long-range order. In the $(Sm_{1-x}Gd_x)_{0.55}Sr_{0.45}MnO_3$ system, the low-temperature state with gadolinium concentrations in the range $0 \leq x \leq 0.5$ in zero magnetic field exhibits metallic properties, whereas at $x = 0.7$ the metallic state induced by an external magnetic field arises only in the field $H \sim 3$ T at temperatures below 50 K. An increase in the magnetic field leads to the stabilization of the metallic ferromagnetic state. Near the metal–insulator phase transition, there are discontinuities in the temperature dependences of the resistance and magnetization of the samples, which are accompanied by a hysteresis that is characteristic of the first-order phase transition. A systematic investigation of the electronic and magnetic properties of $(Sm_{1-x}Gd_x)_{0.55}Sr_{0.45}MnO_3$ single crystals has demonstrated that the metallic ferromagnetic state persists with increasing gadolinium concentration $x$ up to the value $x \sim 0.5$. In this case, the temperature $T_C$ of the paramagnetic–ferromagnetic phase transition decreases linearly from $\sim 130$ to $\sim 50$ K. For gadolinium concentrations $x \geq 0.6$, the spin-glass phase is stabilized with a spin “freezing” temperature $T_G$ in the vicinity of 50 K.

Earlier, it was found that the $(Sm_{0.5}Gd_{0.5})_{0.55}Sr_{0.45}MnO_3$ ceramic samples in zero external magnetic field are characterized by a sharp peak of the magnetic susceptibility near the temperature of 48.5 K with a small temperature hysteresis, which does not depend on the measuring frequency and is accompanied by an increase in the susceptibility with decreasing temperature. A similar peak of the magnetic susceptibility is characteristic of the phase transition to the antiferromagnetic state with long-range charge/orbital ordering [10]. The magnetization isotherms in static and pulsed magnetic fields at temperatures below 60 K demonstrate an irreversible metamagnetic transition to a homogeneous ferromagnetic state with a critical field of the transition, which does not depend on the temperature of measurements. The irreversible metamagnetic transition, apparently, is associated with the destruction of the insulating state with long-range charge ordering. The magnetic-field-induced reversible phase transition to the ferromagnetic state (similar to the metamagnetic transition in the low-temperature phase) caused by the destruction of local charge/orbital correlations is observed at temperatures in the range $60$ K $\leq T \leq 150$ K. With an increase in the temperature, the critical transition fields increase almost linearly and the field hysteresis disappears.