

Research Article

Metamagnetic Phase Transitions in $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ Ceramics

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The temperature dependences of *ac* magnetic susceptibility of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics provide evidence of the formation of a mixed insulating state with a specific quantum-disordered phase, in which the domains with a long-range antiferromagnetic order dominate at temperatures below $T_N \cong 48,5$ K. Irreversible metamagnetic phase transition in the ferromagnetically ordered state is carried out at the critical field $H_{c1} \sim 25$ kOe which remains constant in the temperature range from 4.2 K to 60 K. From the analysis of the magnetization isotherms, the conclusion can be made about a spontaneous phase transition into the state with charge and orbital ordering at temperatures below the critical value $T_{CO} \sim 60$ K. It is supposed that short-range charge correlations exist in unusually large interval of temperatures ~ 90 K at temperatures above T_{CO} that considerably exceeds the area of existence of local orbital correlations ~ 50 K.

1. Introduction

Change of the average radius $\langle r_A \rangle$ of a rare-earth ion replacing lanthanum in A-positions of perovskite-like matrixes in $R_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ($R = \text{Sm}, \text{Eu}, \text{Gd}$) manganites leads to the bicritical phase diagram where the metallic ferromagnetic phase is separated from the dielectric antiferromagnetic phase with charge/orbital (CO/OO) ordering by the first order phase transition [1, 2]. Prominent features which should be observed in experiment close to bicritical point have been predicted: (1) phase transition from a paramagnetic state in the ferromagnetic should be the of first order; (2) the critical temperatures of phase transitions T_{CO} and T_c close to bicritical point should decrease sharply; (3) high sensibility of the sample to external magnetic field near to interphase boundary. These effects arise because of huge fluctuation of several order parameters related to bicritical behavior, which sharply amplifies near to boundary of two phases, where more than two order parameters meet. As a result of the local lattice distortion in $R_{0.55}\text{Sr}_{0.45}\text{MnO}_3$, large structural disorder enhances the phase fluctuation between ferromagnetic metal and charge/orbital-ordered insulator and suppresses their long-range orders [3–5].

The transition between a ferromagnetic (FM) metal and an insulator with short-range charge/orbital correlation induced by the change of average radius $\langle r_A \rangle$ has been investigated for the single crystals $(\text{Sm}_{1-y}\text{Gd}_y)_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ($0 \leq y \leq 1$) [6]. A systematic study indicates that the long-range ferromagnetic order is kept up to $y \sim 0.5$ (corresponding to $R = \text{Eu}$) with T_c reduced from ~ 130 K to ~ 50 K, while it changes to a spin-glass-like insulator for $y \geq 0.6$. Strong competition was found between long-range ferromagnetic order and paramagnetic disorder or a spin-glass-like insulator with short-range charge/orbital correlation, being controlled by changes of the average radius $\langle r_A \rangle$ or the external magnetic field. No macroscopic phase separation is discerned when high-quality single-crystal specimens are used.

The results of measurements of magnetic susceptibility $\chi(T)$ and magnetization isotherms $M(H)$ in the samples of ceramics of $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ in strong static and pulse magnetic fields essentially differ from the same measurements made earlier on the single crystals. It is supposed that the reason of a significant disparity of magnetic properties of these systems is the various nature of the ground state in single crystals and the ceramics, related to different values of the structural disorder.

2. Experimental Procedures

In this work, the ceramic samples of $(\text{Sm}_{1-y}\text{Gd}_y)_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ($0 \leq y \leq 1$) were prepared using the standard solid-state reaction technique. In brief description, homogeneous powders were heat treated during three stages at temperatures of 1000, 1100°C for 10 h, and 1150°C for 24 h with intermediate grinding of as-obtained products. Compacted under pressure of 10 kbar, pellets were sintered in air at 1150°C for 10 h with succeeding temperature drop to the room temperature at the rate of 70°C/hour. Temperature dependences of magnetic susceptibility $\chi(T)$ were measured by induction method at frequencies of 1, 5, 7, and 10 kHz in the temperature range of 4,2–100 K with the help of PPMS-10. Field dependences of magnetization $M(H)$ have been measured in an interval of temperatures of 4,2–200 K in the strong pulse and static magnetic fields. Measurements in pulse fields to 300 kOe at temperatures varied from 20 to 200 K were made by nonindustrial pulse magnetometer, and measurements in static fields to 80 kOe at 4,2 K have been carried out by application of vibrating magnetometer VSM EGG, Princeton Applied Reserch.

3. Experimental Results and Discussion

According to [6], in the $(\text{Sm}_{1-y}\text{Gd}_y)_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ single crystals, the metal ferromagnetic ground state is saved with growth of Gd content only in the range of concentration of $0 \leq y \leq 0.5$. The further increase of y results in the first-order phase transition in dielectric low-temperature state with characteristic signs of spin-glass phase. Unlike the results of article [2], in the given work, the metal ferromagnetic phase was destroyed completely already for $y = 0.5$, that is, associated apparently with larger structural disorder of ceramic samples. Results of measurement of the temperature dependence of *ac* magnetic susceptibility of $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ gave evidences of formation of long-range antiferromagnetic structure in the samples with the critical temperature $T_N \cong 48,5$ K. The sharp peak of *ac* magnetic susceptibility close to 48,5 K is observed independently of the frequency of measurement (Figure 1) and with a small temperature hysteresis ~ 2 K, which strongly differs from a wide peak $\chi(T)$ in the vicinity of $T_G \approx 45$ K for single crystals [6]. The same sharp peak of magnetic susceptibility had been observed earlier in various antiferromagnets with Heisenberg, XY and Ising type of interaction, on magnetic lattices of dimensionality 1, 2, and 3 [7]. At decrease of the temperature, there is fast growth of $\chi(T)$ for $T \geq T_N$ and then a sharp drop of the susceptibility was found for $T \leq T_N$. As shown in Figure 1, sharp peak of magnetic susceptibility is imposed on continuous paramagnetic increase of a susceptibility with the temperature drop in the wide temperature interval. The magnetic susceptibility intensively rises with fall of temperature of the sample and diverges near to absolute zero of temperatures, that is, characteristic for quantum phase transitions in low-dimensional quantum Heisenberg antiferromagnets with the structural disorder [8–10]. In this phase, low-temperature

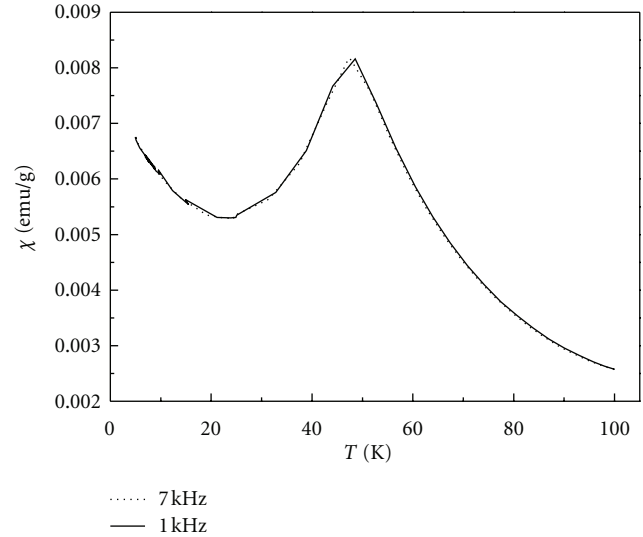


FIGURE 1: Initial *ac* magnetic susceptibility curves $\chi(T)$ in the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics in zero external magnetic field.

uniform susceptibility diverges algebraically with nonuniversal exponents. This is a signature that the quantum-disordered phase is a *quantum Griffiths phase*. Superposition of two various contributions to a low-temperature magnetic susceptibility testifies to the inhomogeneity nature of the ground state of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics in a zero external magnetic field. Apparently, in these ceramic samples, long-range antiferromagnetism coexists with short-range magnetic correlations in the large temperature interval near absolute zero. It is supposed that the mixed insulating state with a special feature of a quantum-disordered phase with dominating domains of long-range antiferromagnetism is found in the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics at the temperatures below $T_N \cong 48,5$ K. Similar mixed state close to quantum critical point had been discovered recently in the $\text{La}_{2/3}\text{Ca}_{1/3}\text{Mn}_{1-x}\text{Ga}_x\text{O}_3$ manganites in the vicinity of a metal-insulator transition [11, 12].

The variation of an external static magnetic field in an interval ± 6 T at 4,2 K had led to irreversible destruction of mixed phase accompanied by formation of the steady ferromagnetic phase (Figure 2). Field-induced stable FM phase was formed as a result of continuous metamagnetic phase transition induced by growth of an external magnetic field to extremely small critical value $H_{c1} = 25$ kOe with a sharp increase in magnetization $M(H)$ in the form of a step in height ΔM . The further increase of the field to 60 kOe resulted only in insignificant growth of magnetization. At the magnetic field reduction, the induced ferromagnetic phase is saved up to zero field. The subsequent variation field in an interval ± 15 kOe has allowed receiving of a wide hysteresis loop corresponding apparently to an anisotropic ferromagnetic state of the sample at 4,2 K. A distinction of the received magnetization curve $M(H)$ at 4,2 K is large width of the hysteresis loop ($\sim 4,4$ kOe), which corresponds to coercive field $H_c \cong 2,2$ kOe, and rather small size of magnetization of saturation in the field of 60 kOe ~ 60 emu/g, related evidently with the canted state of the manganese spins.

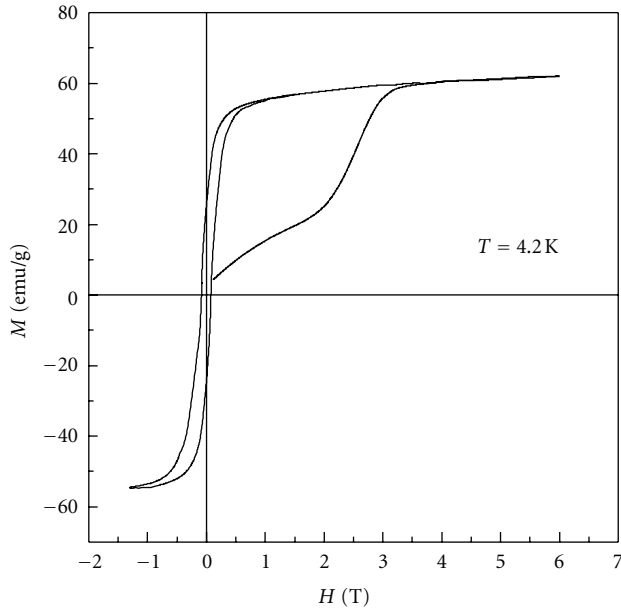


FIGURE 2: Evolution of a magnetization isotherm $M(H)$ of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ sample at 4.2 K induced by metamagnetic phase transition.

Similar behavior of the field dependence of the magnetization was observed in $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ at 5.0 K [13]. According to neutron-diffraction experiments, without a magnetic field, $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ demonstrates three phase transitions: change of the lattice symmetry at $T_B = 200$ K, antiferromagnetic ordering at $T_N = 140$ K, and canted antiferromagnetism at $T_{CA} = 110$ K. It shows a canted ferromagnetic structure accompanied with antiferromagnetic (AF) components of so-called pseudo-CE-type antiferromagnetic structure at the temperatures below T_{CA} . Furthermore, the charge ordering of Mn^{3+} and Mn^{4+} ions takes place simultaneously with a lattice distortion described as buckling, where MnO_6 octahedra show alternating tilting and cause a doubling of the periodicity of the the lattice along the [010] direction. Although the resistivity of $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ shows insulating behavior at zero field, it exhibits an insulator-metal transition at around 4.0 T. An applied field enforces an alignment of the AF components towards the field direction and drives $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ into the metallic state by actuating the double-exchange mechanism and destroying the charge ordering. This an insulator-metal transition induced by external magnetic field is accompanied also by a large magnetic hysteresis, that indicates a strong coupling between spins and charges. It has been suggested that irreversible metamagnetic phase transition in $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ can be viewed as a melting of the charge ordering.

It is well known that the field-induced insulator-metal phase transition in half-doped manganites occurs at relatively small magnetic fields $\sim 10\text{--}40$ T. Rather little value of the critical magnetic field necessary for generation of a ferromagnetic metal state is explained within the framework of double-fluid model [14, 15] by the competition between canting induced metallicity and inhomogeneity arising from the trapping of carriers by the Jahn-Teller defects. It has been

found that with growth of intensity of an external magnetic field basic state sharply leaves from the JT-distorted canted insulator state and is seized in a metal phase with the optimal (high) canting angle. In the given work, metamagnetic phase transitions in the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics located in immediate vicinity of the insulator-metal transition take place at extremely small value of the critical external magnetic field. This suggests that the phase transitions in a ferromagnetic state in $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ are induced by a delocalization of self-trapped e_g -electrons of Mn accompanied by disappearance of the local JT distortions and the locking in a metal ferromagnetic phase.

As shown in Figure 3, at temperatures smaller 60 K, isotherms of $M(H)$ in pulse fields practically coincide in form with the curves of the magnetization obtained in static fields at 4,2 K. Size of magnetization step ΔM produced by transition in the metal ferromagnetic state practically did not change with temperature growth. Metamagnetic phase transitions occur at the critical field $H_{c1} \cong 25$ kOe which remains constant up to the temperature of 60 K. Stability of the induced ferromagnetic phase in the range of temperatures of 4,2–60 K is confirmed also by the temperature dependence of ac magnetic susceptibility $\chi(T)$ (Figure 4) received at heating of the sample in zero dc field where the ferromagnetic phase has been induced by strong magnetic field at 18 K. The temperature dependence of susceptibility corresponds to phase transition from the induced ordered ferromagnetic state to the disordered paramagnetic state with critical temperature of $T_c \approx 48$ K. The temperature of 60 K in this figure corresponds to full destruction of long-range ferromagnetic ordering of Mn spins and can be considered as a boundary of stability of the induced ferromagnetic phase, which considerably exceeds the area of existence of the mixed state in zero magnetic field. Unusual feature of metamagnetic phase transitions in the range of temperatures of 4.2–60 K is the constancy of critical field H_{c1} , that is, an evidence of absence of effect of temperature changes on generation of ferromagnetic phase in the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics. It is necessary also to notice that irreversible metamagnetic phase transitions investigated in the given paper do not contain strong discontinuities of isotherms of $M(H)$ that are characteristic of earlier investigated irreversible metamagnetic phase transitions in the manganites [16, 17] both in static, and in pulse magnetic fields, and exist at sufficiently elevated temperatures. Therefore, the mechanisms of the irreversible transitions offered in these papers cannot fully explain the given experimental results.

At temperatures above 60 K, behaviour of magnetization in a strong pulse magnetic field essentially varies. As shown in Figure 3, the isotherms of magnetization in high-temperature paramagnetic phase agrees with well-known behaviour of $M(T)$ curves in a Griffiths-like phase and strongly differs from the isotherms received in low-temperature phase with the mixed insulating ground state with dominating domains of long-range antiferromagnetism. At growth of intensity of external magnetic field to the critical value H_{c1} , there is a reversible metamagnetic-like transition from paramagnetic to a ferromagnetic state in the form of a narrow step on

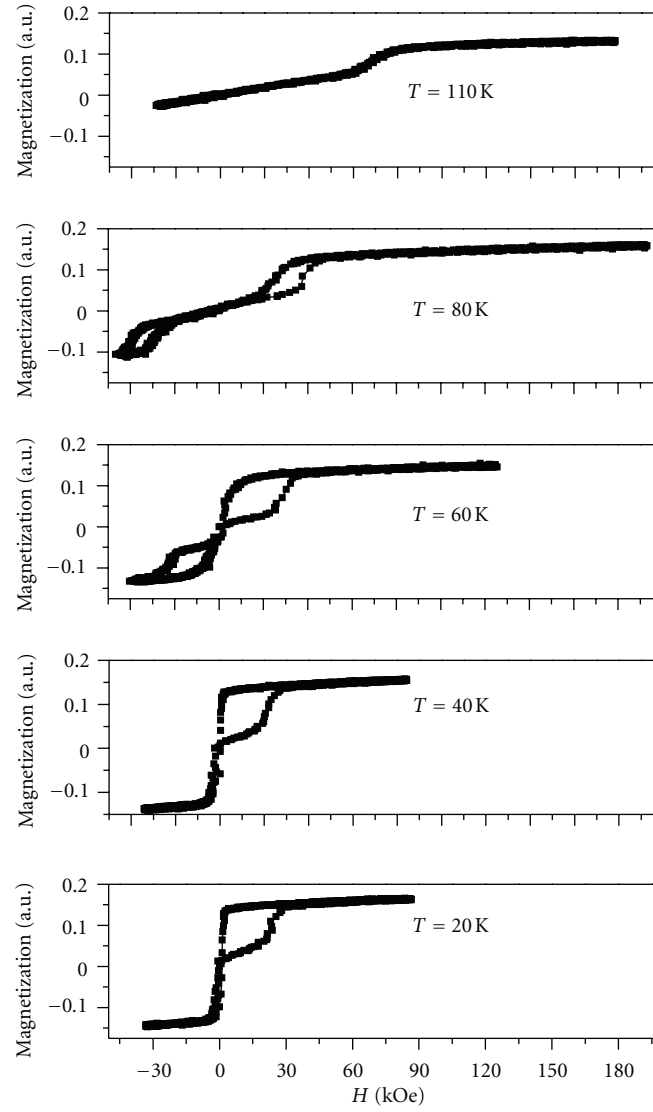


FIGURE 3: Evolution of magnetization isotherms $M(H)$ of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics in the pulse magnetic fields for temperatures of 20–110 K.

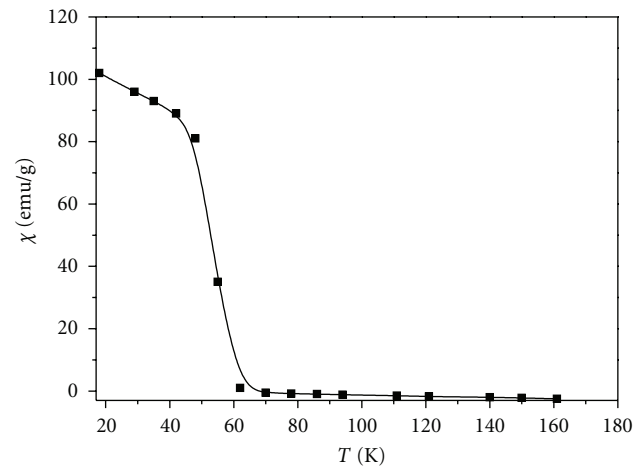


FIGURE 4: Temperature dependence of the ac magnetic susceptibility $\chi(T)$ of the ceramics $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ obtained after metamagnetic phase transition at 18 K.

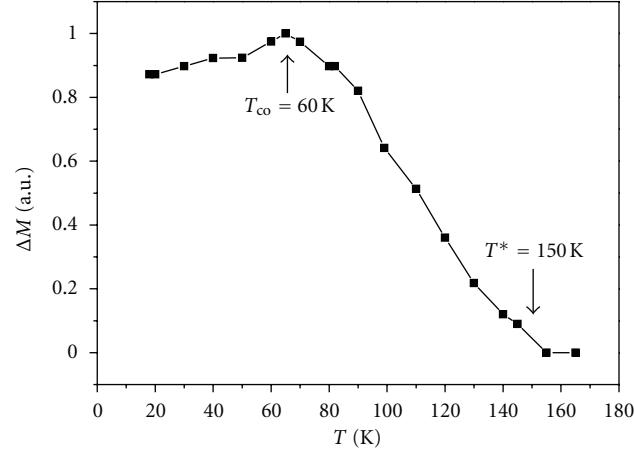


FIGURE 5: Temperature dependence of magnetization step height $\Delta M(T)$ at the metamagnetic phase transitions in the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics induced in the temperature interval of 18–165 K.

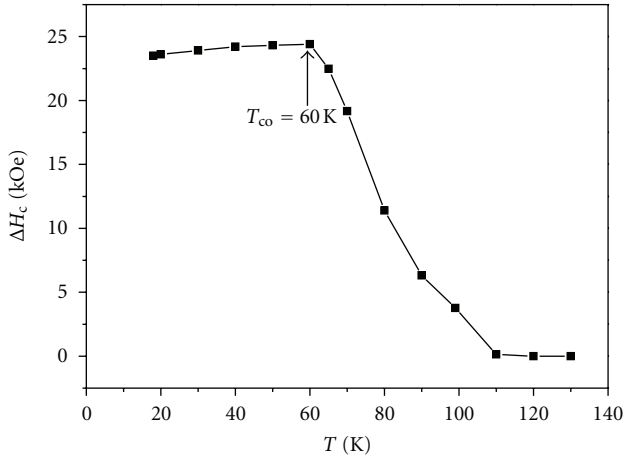


FIGURE 6: Temperature dependence of field hysteresis width ΔH_c of the metamagnetic phase transitions in the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics induced in the temperature interval of 18–165 K.

$M(H)$ curve accompanied by field hysteresis at reduction of magnetic field intensity with critical field $H_{c2} \neq 0$ below which the sample comes back to the initial paramagnetic state. The values of critical fields H_{c1} and H_{c2} practically linearly increase at temperatures above 80 K, whereas a difference of critical fields $\Delta H_c = H_{c1} - H_{c2}$ decreases with the temperature rising and reaches zero near 110 K. At the further rise in temperature, the field-induced phase transition looked like a narrow step of magnetization $\Delta M(T)$ without field hysteresis, which was displaced linearly on temperature towards stronger fields and disappeared at temperatures above 150 K (Figure 5). At temperatures above 150 K corresponding to top boundary T^* of Griffiths-like phase, we observed only linear increase in magnetization with field growth, characteristic for a usual paramagnetic phase. Unusual behaviour of magnetization isotherms at temperatures above 60 K apparently is caused by growth of local charge/orbital correlations with the temperature drop

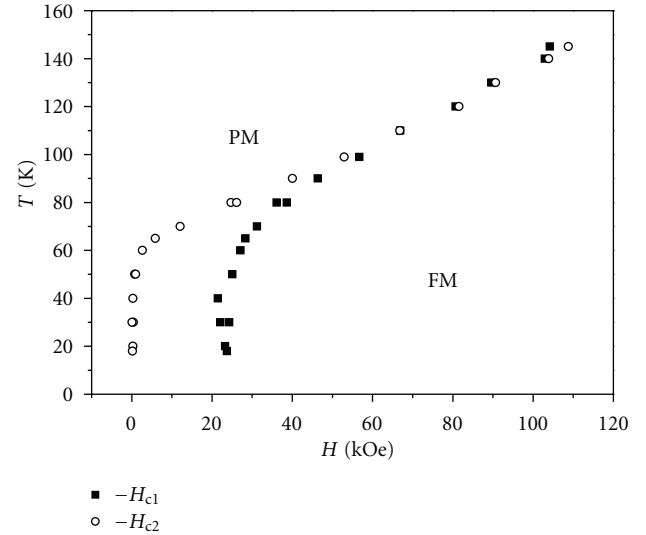


FIGURE 7: Magnetic H - T phase diagram of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics, received in strong pulse magnetic fields.

to the critical value $T_{CO} \cong 60$ K associated with transition of the sample to a state with long-range charge-ordering. The strong magnetic field destroys both long-range charge-ordering state ($T < T_{CO}$) and the state with short-range charge/orbital correlation ($T > T_{CO}$) that results in field-induced phase transitions found in this work. It is possible to assume that sharp falling to zero of width of a hysteresis ΔH_c of the critical fields of metamagnetic phase transition with growth of temperature (Figure 6) is related to destruction of local correlations of orbital order $\xi_{OO}(T)$ at temperatures above 110 K, whereas field-induced magnetization step ΔM , falling to zero in the range of temperatures of 60–150 K, implies destruction of local charge-order and reduction to zero of the correlation length $\xi_{CO}(T)$ at the temperature $T^* \cong 150$ K. Magnetic H - T phase diagramme of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics had been received in strong pulse magnetic fields (Figure 7).

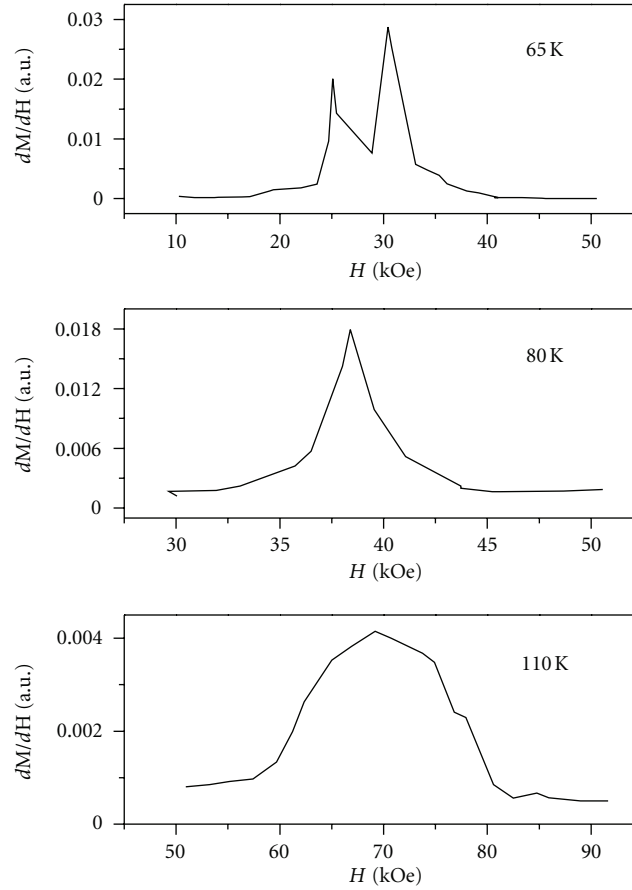


FIGURE 8: Field dependences of derivative of magnetization dM/dH received near to critical field H_{c1} of transition in a ferromagnetic state at temperatures above $T_{CO} \cong 60$ K.

Special interest is represented by additional step structure of magnetization isotherms $M(H)$ close to critical field H_{c1} where charge/orbital correlations are broken, received only in the rapid pulse fields with sweep rate of magnetic field ≥ 2 T/ μ s. In experiment, an existence of one or two small narrow steps of the magnetization was shown, which arise with growth of magnetic field near to critical value H_{c1} and are absent in hysteresis curves at field reduction close to H_{c2} . More clearly, this feature is shown in field dependences of magnetization derivative dM/dH close to H_{c1} in the form of additional narrow peaks of the derivative, having by the basis peak in width ~ 20 kOe (Figure 8). These steps of isotherms of magnetization exist only at temperatures lower 110 K that allows us to assume that the reason of their occurrence is destruction of short-range orbital correlations forming the domain structure of JT distortions of a crystal lattice for $T < 110$ K. In vicinity of $T_{CO} \cong 60$ K, there is a splitting of dM/dH single peak evidently connected with existence of close T_{CO} incommensurate lattice modulation of an elastic field JT distortions with wave number incommensurate with the lattice constant.

4. Conclusion

The results of measurement of the temperature dependences of *ac* magnetic susceptibility $\chi(T)$ of the $(\text{Sm}_{0.5}\text{Gd}_{0.5})_{0.55}\text{Sr}_{0.45}\text{MnO}_3$ ceramics illustrate formation of the mixed insulating state with a special feature of a quantum-disordered phase with dominating domains with a long-range antiferromagnetism at the temperatures below $T_N \cong 48,5$ K. In extremely weak magnetic field $H_{c1} \cong 25$ kOe, field-induced irreversible metamagnetic phase transition from a mixed insulating state to metallic ferromagnetic one was found out, being related to destroy of long-range charge/orbital correlation which amplifies the local antiferromagnetic correlations of Mn spins. From the analysis of isotherms of magnetization, the conclusion was made about spontaneous phase transition with charge/orbital ordering at temperatures below the critical value $T_{CO} \cong 60$ K. At temperatures above 60 K, there are reversible field-induced metamagnetic-like transitions in the ferromagnetic state, related to destroy of short-range charge/orbital correlations. It is supposed that nanoscale correlations of a charge exist in unusually large interval of

temperatures ~ 90 K at temperatures above T_{CO} that considerably surpasses area of existence of short-range orbital correlations ~ 50 K, creating an elastic field Jahn-Teller deformations of the crystal lattice. An additional steps structure of magnetization isotherms close to H_{c1} was found, which exist only at magnetization in very fast pulse fields with sweep rate of magnetic field ≥ 2 T/ μ s. It is supposed the existence of close to T_{CO} incommensurate lattice modulation of elastic field JT distortions with wave number incommensurate with the lattice constant.

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