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Penetration of electromagnetic field through the La-Er-Ba manganite far above the magnetic phase transition temperature

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Abstract. Using the technique of penetration of electromagnetic waves through the bulk samples of $La_{0.60}Er_{0.07}Ba_{0.33}MnO_3$ manganite, it is shown that in this class of strongly correlated materials the local dynamic magnetic ordering is preserved at the temperature by 66 K above the Curie temperature where the static long-range magnetic order is absent. In this temperature range the dynamic magnetic permeability exceeds unity and is frequency dependent. The average lifetime of local ordered states is estimated.

PACS. 75.47.Lx Manganites – 75.47.-m Magnetotransport phenomena; materials for magnetotransport – 75.47.Gk Colossal magnetoresistance

1 Introduction

Due to the strong correlation between the charge, spin, orbital, and lattice degrees of freedom in doped lanthanum manganites investigations of the magnetic subsystem remains one of the most important tasks in physics of this class of oxide materials. Relative importance of different mechanisms, including double exchange, phase separation, charge-lattice coupling, and others, is under intensive investigation now [1]. One of the problems, which is important for understanding the physical mechanisms of magnetic interactions in manganites, is the role of magnetic fluctuations and its influence on magnetic and transport properties. Since the intrinsic "colossal" magnetoresistance in high quality single crystals and thin films of doped manganites is observed near the temperature of magnetic phase transition, the contribution from magnetic fluctuations must be considered as the important factor which in a complicated way influences the behavior of the system. The method of penetration of electromagnetic waves was found to be effective in investigations of the dynamic electromagnetic properties of doped manganites [2,3]. The electromagnetic waves are excited by one coil and the signal in the second (receiving) coil, which is separated from the first one by a manganite sample, is measured. Its value depends on the peculiarities of penetration of electromagnetic waves through a manganite sample, which acts as a screening element placed across the direction of their propagation. This method permits not only to investigate the temperature and frequency variations of resistivity and magnetic permeability, but also to study the frequency characteristics of magnetic fluctuations. Thus, investigations of doped manganites by the method of penetration of electromagnetic waves open the principal possibility to determine the time of existence of short-range dynamic magnetic ordering in the wide temperature range, including the temperatures both below and above the Curie temperature T_c.

It was shown [2], that the *dc* magnetic field and frequency dependences are the most informative in the penetration experiments. The coefficient of penetration of electromagnetic waves through a manganite sample D can be defined as the ratio between the values of electromagnetic fields in the receiving and exciting coils. Variations of the penetration coefficient in a *dc* magnetic field r_m (see definition below) are determined by two physical characteristics: the magnetic permeability and electrical resistivity as well as by their magnetic field dependence. If measurements are carried out in the ferromagnetic temperature range, upon application of the *dc* magnetic field the reversible differential dynamic magnetic permeability decreases. This results in increase of the penetration coefficient. It was shown [1–3], that for $T < T_c$, except for the highest frequencies of the order of hundreds megahertz, variation of the penetration coefficient r_m in applied magnetic field is positive owing to variations of the magnetic permeability. If the frequency of the electromagnetic field is so high that magnetic permeability substantially decreases owing to its frequency dispersion, variations of r_m in doped manganites are determined by "colossal" magnetoresistance. Since in manganites resistivity decreases upon application of the *dc* magnetic field, in this frequency range r_m is negative.

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The anomalies of the coefficient of penetration of radiofrequency electromagnetic field near the Curie temperature are most pronounced if the frequency is chosen according to the following reasons. First, the frequency must not be too high in order to avoid the frequency dispersion of magnetic permeability. Second, the condition $d < \delta$, where d is the thickness of a manganite sample and δ is the skin depth, must be fulfilled. This puts the lower limit for the frequency of the electromagnetic field. Under this condition the comparison of experimental results with theoretical considerations becomes easier. The magnetic field dependence of penetration coefficient in manganites was found not only for the ferromagnetic state, but also for the paramagnetic state at temperatures slightly (by 10−15 K) above T_c [3], which can be considered as manifestation of preservation of local dynamic short-range magnetic ordering. The question about the upper limit of the temperature range in which such short-range dynamic magnetic ordering exists in doped manganites remains open.

The aim of this work is to investigate the dynamic magnetic properties of doped manganites far above the Curie temperature using the method of penetration of electromagnetic waves. The $La_{0.60}Er_{0.07}Ba_{0.33}MnO_3$ manganite was studied since it has the T_c value only slightly above the room temperature. Doping the La-Ba manganite with erbium, as well as with other rare-earth elements, permitted to observe and investigate the magnetic pair-breaking effect [4]. It is known that upon doping manganites with rare-earth elements additional magnetic interaction, which, in particular, lowers the Curie temperature between the rare-earth and manganese ions, is introduced. It is important to note that in the penetration experiments it is possible to investigate variations of the dynamic magnetic ordering above T_c, which can not be studied by the standard *dc* or low-frequency magnetization techniques. In this work the results obtained in the experiments with penetration of electromagnetic waves of different frequencies will be compared to the *dc* properties (magnetization and resistivity).

2 Experimental technique

The powder of nominal composition $La_{0.60}Er_{0.07}Ba_{0.33}$ MnO_3 was prepared by the co-precipitation technique, which permits to obtain the samples with improved homogeneity [4]. In the final stage the powder was pelletized and annealed in flowing oxygen for 12 h at 1200 °C. The disk-shaped samples had the diameter of 10 mm and the thickness d of 1.5 mm. The structural characteristics and homogeneity of the samples were investigated by X-ray diffraction and electron-probe microanalysis.

The *dc* magnetic properties were studied using extraction magnetometer in the temperature range 5−400 K with magnetic field values up to 60 kOe. The value of the Curie temperature $T_c = 304.6$ K was determined from the magnetization curves using the Arrott-Belov plots. The *dc* magnetoresistance $MR = [\rho(H) - \rho(0)]/\rho(0) \cdot 100\%$, where $\rho(H)$ and $\rho(0)$ are the values of electrical resistivity in the magnetic field H and zero magnetic field, respectively,

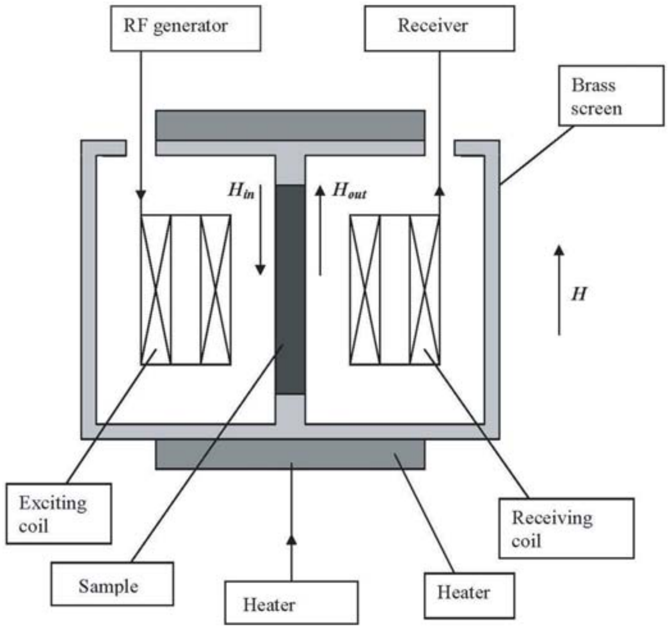

Fig. 1. The scheme of the radiofrequency measurements.

was measured by the standard four-probe method. The value of $\rho(0)$ at $T = 304.6$ K was found to be 0.183 Ω cm. The maximum value of magnetoresistance measured in the field of $H = 10$ kOe in the temperature range including T_c was −7.6%.

The plate-shaped bulk manganite sample was used as a screening element placed between two inductively coupled coils. One coil was used for excitation of the *ac* electromagnetic field, which passed through a manganite sample, and was recorded by the receiving coil, see Figure 1. In all experiments the *ac* electromagnetic field was oriented parallel to the *dc* magnetic field, both lying in the plane of a sample. The modulus of the penetration coefficient can be defined as $|D| = |\vec{H}^{out}_{\sim}|/|\vec{H}^{in}_{\sim}|$, where $|\vec{H}^{in}_{\sim}|$ and $|\vec{H}^{out}_{\sim}|$ are the amplitudes of the electromagnetic fields of the exciting and receiving coils, respectively. Experimentally the *ac* voltage in the receiving coil U_{out} was measured when the exciting coil was powered by small (typically about 0.1 V) *ac* voltage U_{in}. Since $U_{out} \approx \omega|\vec{H}^{in}_{\sim}|DnS$, where $\omega = 2\pi f$ is the angular frequency, n is the number of turns in the receiving coil, and S is its cross-section, $|D| \approx U_{out}/U_{in}$. Variations of D with frequency, *dc* magnetic field, or temperature were measured. The frequency range from 0.05 to 30 MHz was investigated. Typical values of the signal in the receiving coil without a sample were about 100 mV. Placing a metallic sample resulted in substantial screening of the receiving coil and decrease of the signal to the level of ∼0.3 μV. The signals, recorded with a manganite samples, were at the level of 10 μV. Thus, the signal from leakage around a sample was below 0.3 μV. Variations of the penetration coefficient in the magnetic field are more convenient to characterize by the parameter $r_m = [D(H) - D(0)]/D(0) \cdot 100\%$ where $D(H)$ and $D(0)$ are the values of the penetration coefficient in the magnetic field H and zero magnetic field, respectively. The values of skin depth δ can be calculated for different frequencies assuming the value of the magnetic permeability μ equal to unity. For the frequency of $f = 0.05$ MHz $\delta_1 = 98$ mm while for the frequency of $f = 30$ MHz $\delta_1 = 4$ mm. Thus, the condition $d \ll \delta_1$ was satisfied for the whole frequency range investigated in this work. The magnetic field dependences were reproducible within less than 1% while the temperature dependences – within 2%.

3 Results and discussion

The results of measurements of the *dc* magnetization isotherms for the $La_{0.60}Er_{0.07}Ba_{0.33}MnO_3$ manganite recorded at different temperatures near the Curie temperature are shown in Figure 2a. It is important that magnetic saturation is not reached even for the highest value of the applied magnetic field (60 kOe). Figure 2b shows the temperature dependence of resistivity. Its shape is typical of bulk deoxygenated polycrystalline manganites: the broad peak around 240 K [5], the small peak near T_c, and semiconductor-like behavior of resistivity above T_c.

Figure 3 shows the results of investigations of the *dc* magnetic field dependences of penetration coefficient. The data of Figure 3a were recorded at $T = 293$ K, i.e., they correspond to the ferromagnetic state of the sample. Measurements were made at several frequencies. At the lowest frequency of $f = 0.05$ MHz r_m monotonically increases with the *dc* magnetic field. At higher frequencies a maximum appears, the position of which corresponds to the value of magnetic field approximately equal to the anisotropy field. Variations of r_m increase with frequency. The abovementioned features point to the fact that at frequencies below 30 MHz the frequency dispersion of the magnetic permeability is not important. This circumstance is characteristic of the $La_{0.60}Er_{0.07}Ba_{0.33}MnO_3$ composition since for other compositions, in particular for the Pb doped manganites, the frequency dispersion of the magnetic permeability is clearly seen at frequencies of the order of 1 MHz. In the ferromagnetic state the *dc* magnetic field variations of the penetration coefficient are mostly due to the dynamical differential magnetic permeability which changes in low *dc* fields strongly. Because of that fact, there is no reason to expect that the field dependences of the penetration coefficient and *dc* magnetization which is governed by the *dc* permeability, will look similar each other, see Figures 2a and 3a.

The results of measurements of the *dc* magnetic field dependences of penetration coefficient at the temperature of $T = 371$ K are shown in Figure 3b. This temperature is by 66 K above the Curie temperature. These results of the *dc* magnetic field measurements show that the long-range magnetic order is absent in the sample at this temperature. Nevertheless, substantial variations of the penetration coefficient are observed in the wide frequency range. At the frequency of $f = 0.05$ MHz r_m monotonically increases reaching the value of 60% in the magnetic field of 10 kOe. The dependences measured at higher frequencies have maxima and negative values of r_m in strong field.

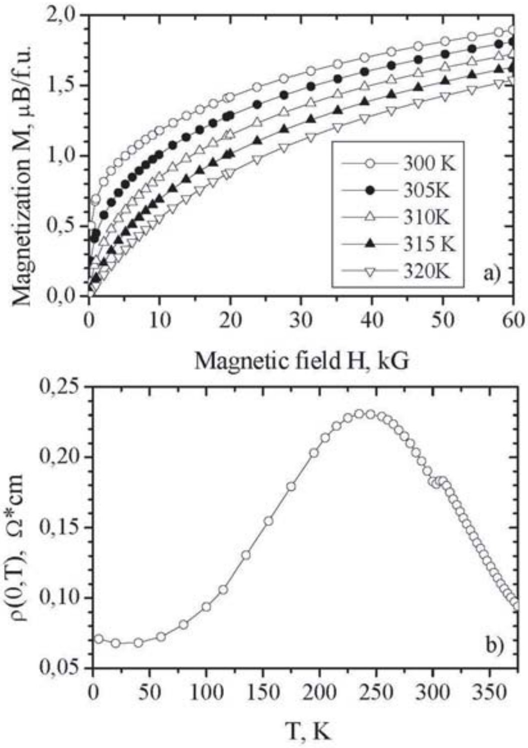

Fig. 2. The *dc* magnetization curves for the $La_{0.60}Er_{0.07}Ba_{0.33}MnO_3$ manganite near the Curie temperature (a) and the temperature dependence of resistivity (b).

Note that no full saturation is observed for the $r_m(H)$ dependences in the fields from 6 to 10 kOe.

The following formula is valid for the penetration coefficient of electromagnetic waves through thin ferromagnetic screen under the $d \ll \delta_1$ condition [2]:

$$D = \frac{2\rho}{Z_0 \mu d}, \tag{1}$$

where Z_0 is the wave impedance of the free space ($Z_0 = 120\pi\Omega$). The penetration coefficient is proportional to resistivity and inversely proportional to magnetic permeability and thickness of a sample. The following formula, which relates the ratio of penetration coefficients in zero magnetic field and in saturating field with the initial effective magnetic permeability $\mu(0)$ and magnetoresistance $M R$, can be obtained from (1):

$$\left| \frac{D(H \to \infty)}{D(0)} \right| = \mu(0)(1 + MR). \tag{2}$$

The formula (2) is valid when $|MR \ll 1|$. We can use it to estimate the initial effective dynamic magnetic permeability using the data of Figure 3b corresponding to the frequency of $f = 0.05$ MHz since at this frequency variation of penetration coefficient in the magnetic field is positive, i.e., it is caused mostly by the magnetic permeability. At higher frequencies variations of r_m are negative and are caused mostly by magnetoresistance. From Figure 3b $r_m(H = 10 \text{ kOe}) = 0.6$ which means that

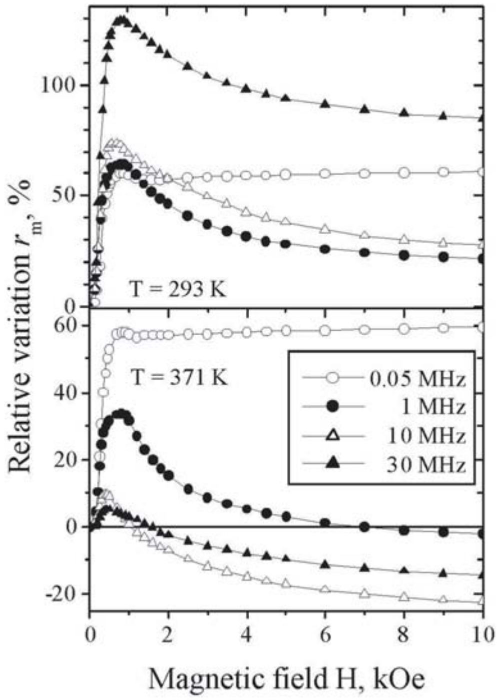

Fig. 3. The *dc* magnetic field dependences of the normalized penetration coefficient below the Curie temperature at $T = 293$ K (a) and above the Curie temperature at $T = 371$ K (b).

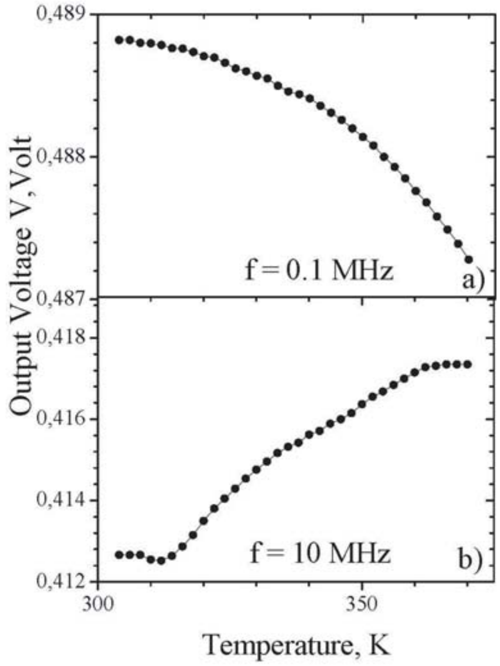

Fig. 4. Temperature dependence of the signal in the receiving coil for the *dc* magnetic field of $H = 0.8$ kOe: frequency $f = 0.1$ MHz (a); frequency $f = 10$ MHz (b).

$|\frac{D(H=10\ \text{kOe})}{D(0)}| = 1.6$ and, according to formula (2) $\mu(0) \approx 1.7$, with an account for $MR(10\ \text{kOe}) = -7.6\%$. Thus, the value of the dynamic initial effective magnetic permeability substantially differs from unity at temperature by 66 K above T_c, which means that short-range magnetic order still exists in some parts of a sample while the long-range magnetic order is absent. This result is in accordance with the phase separation mechanism of colossal magnetoresistance in doped lanthanum manganites [6]. Formation of ferromagnetic clusters in the temperature range by 100 K above T_c was experimentally observed in [7].

The temperature dependence of the signal in the receiving coil measured at the frequency of $f = 0.1$ MHz is shown in Figure 4a. Measurements were made upon heating. Figure 4b shows similar dependence measured at a frequency of $f = 10$ MHz upon heating in the *dc* magnetic field of $H = 0.8$ kOe which approximately corresponds to the anisotropy field. The shape of temperature dependence is substantially different: the amplitude of the measured signal decreases with temperature in Figure 4a, while in Figure 4b it increases. According to the abovementioned considerations, this difference can be explained by domination of contributions from different factors: the magnetic permeability at low frequencies (Fig. 4a) and the colossal magnetoresistance at high frequencies (Fig. 4b).

The principal result of this work is in experimental proof, using the electromagnetic field penetration technique, of the fact that in the $La_{0.60}Er_{0.07}Ba_{0.33}MnO_3$ manganite the local dynamic magnetic ordering is preserved in the temperature range at least by 66 K above the Curie temperature. At these temperatures the magnetic permeability is the highest at the frequencies of tenths kilohertz, which corresponds to the average lifetime of local ordered states of about 10−100 μs. The results of the *dc* magnetic measurements show that the static long-range magnetic order is absent in this temperature range.

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