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Giant magnetic-field changes in radio-frequency absorption in $La_{0.67}Sr_{0.33}MnO_3$ near the Curie temperature

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Abstract

The DC transport properties of and the radio-frequency (RF) wave absorption (at 2.525 MHz) in a sample of $La_{0.67}Sr_{0.33}MnO_3$ prepared by floating-zone method are measured. The Curie temperature, T_c, of the sample is about 374 K. Giant temperature and magnetic-field variations in RF absorption are found in the vicinity of T_c. Relative change of the RF absorption in magnetic field (magnetoabsorption) is about 67% in field 2.1 kOe and about 55% in field 1 kOe. This giant magnetoabsorption effect can be used to develop RF devices controlled by temperature and low magnetic field. A weak temperature dependence of magnetoabsorption for the sample studied in the range from room temperature to about 350 K makes it especially attractive for practical use. The RF study supplemented with transport, magnetoresistive and magnetic measurements enables us to discuss the optimal properties of manganite samples for observation of giant magnetoabsorption in low field.

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Keywords: Manganites; Colossal magnetoresistance; Magnetically ordered materials; RF absorption

1. Introduction

In the last decade much interest is being shown in mixed-valence manganites of the type $La_{1-x}A_xMnO_3$, where A is a divalent alkaline-earth element like Ca, Sr, Ba [1–3]. In these compounds, a huge negative magnetoresistance [so called, colossal magnetoresistance (CMR) effect] has been found near the temperature, T_c, of paramagnetic–ferromagnetic transition. This effect offers applications in advanced technology. The amplitude of CMR in manganites depends essentially on magnitude of applied magnetic field and the value of Curie temperature T_c [1,2]. For

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example, in optimally doped $La_{1-x}A_xMnO_3$ with $x \approx 0.3$, high-field (60–100 kOe) magnetoresistance (MR), defined as $-\Delta R(H)/R(H)$, can be about 5 at $T \simeq T_c$ in Ca manganites with $T_c \simeq 250$ K; whereas in Sr manganites with T_c about 370 K the MR is by the order of magnitude smaller [1,4].

In spite of enormous efforts, a clear understanding of CMR is not yet available, though some numerical simulations can describe experimental data with an acceptable accuracy [1,3]. Beside this, the requirement of high magnetic field hampers a possible application of the CMR manganites to practical use. It is desirable, therefore, for application purposes to look for some other "non-magnetic" properties of manganites which are dependent, however, on their magnetic properties to the extent that it can ensure a sensitivity to rather low magnetic field. A well known property of this type is the high-frequency complex resistance (impedance) which can show a high sensitivity to low magnetic field in soft ferromagnetic (FM) conductors. Generally this phenomenon is called magnetoimpedance. In the case that a considerable relative change (several tens of percent) in the impedance takes place in low external magnetic field this phenomenon is referred to as giant magnetoimpedance [5].

The impedance of a sample is determined by its resistance and reactance both of which are influenced by absorption of applied electromagnetic waves. It is known that in FM metals this absorption is influenced not only by the conductivity, but by the magnetic permeability, μ_r, of absorbing media as well. The skin depth in FM conductor is given by

$$\delta = \sqrt{\frac{2\rho}{\omega\mu_0\mu_r}}, \tag{1}$$

where ρ is the resistivity, $\omega = 2\pi\nu$ the wave angular frequency, and μ_0 the magnetic permeability of vacuum. The application of an external magnetic field leads to a considerable decrease in the permeability, causing increase in the skin depth, and hence a giant change in the high-frequency wave absorption. This effect is, however, fairly large only for optimal frequency range. On the one hand, the frequency must be high enough to assure that δ is smaller than the sample dimensions [5]. On the other hand, the magnitude of μ_r in zero magnetic field should be high enough to produce a giant magnetoabsorption (or magnetoimpedance) effect. Really, it is known that the value of μ_r in FM metals decreases with increasing frequency going close to unity for $\nu \geqslant 100$ MHz [6]. At high enough frequency, therefore, an FM metal behaves like a non-magnetic one. For this reason, the optimal frequency for observation of giant changes in high-frequency absorption should be of the order of 1 MHz, that is, in the radio-frequency (RF) range. For example, a giant RF magnetoabsorption is found in cobaltite $La_{0.5}Sr_{0.5}CoO_3$ [7] which is a FM perovskite-like oxide related to manganites but with a far smaller MR. The relative change in magnetoabsorption in this compound was found to be about 38% near the Curie temperature (about 250 K) in a rather weak magnetic field 2.1 kOe at frequency 1.33 MHz [7].

In this article, we report a study of RF magnetoabsorption effect in a bulk sample of $La_{1-x}Sr_xMnO_3$ ($x = 0.33$) prepared by floating-zone method. For this Sr concentration, according to the known phase diagram [2], manganite samples are in metallic state both below and above T_c. The T_c was about 374 K in the sample studied. The previous RF studies of magnetoimpedance in Sr manganites (with about the same composition, $x = 0.3$ and 0.35) were done [8] only at room temperature, that is rather far below T_c. A rather minute magnetoimpedance effect (a few percent) is found in these samples. Temperature dependence of the complex permeability in $La_{0.7}Sr_{0.3}MnO_3$ and the effect of crystal-lattice disorder on this were studied in Refs. [9,10]. All those studies were done on polycrystalline ceramic samples. In this study, RF (2.525 MHz) absorption in $La_{0.67}Sr_{0.33}MnO_3$ sample was studied in rather large temperature range (from room temperature up to about 400 K), which includes T_c. In contrast to Ref. [8], we found a really giant magnetoabsorption below T_c (including room temperature) with maximum amplitude about 67% in low magnetic field 2.1 kOe. The effect remains to be giant in smaller fields as well. For example, in field 1 kOe the magnetoabsorption is about 55%. This

makes it attractive for practical use. RF study was supplemented with transport, magnetoresistive and magnetic measurements of the sample which make it possible to discuss the optimal properties of manganite samples for observation of giant magnetoabsorption in low field.

2. Experimental

A crystal of nominal composition $La_{0.67}Sr_{0.33}MnO_3$ was grown by the floating zone method at the Institute of Physics, Warsaw. The appropriate amounts of starting materials La_2O_3, $SrCO_3$ and MnO_2 were calcined at temperature 1000°C, then mixed, compacted into pellets and sintered at 1400°C. After that the pellets were milled, and resulting powder was pressed to form the feed rod with a diameter of 8 mm and a length of 90 mm. This rod was further sintered at 1470°C during 12 h in air. An optical furnace (type URN-2-3Pm made by Moscow Power Engineering Institute) with two ellipsoidal mirrors and 2500 W xenon lamp as the heat source was applied for crystallization. The feed rod and the growing crystal were rotated in opposite directions to make heating uniform and to force convection in the melting zone. The growth rate was 1 mm/h. An additional afterheater was used for lowering the temperature gradients in growing crystal. It is known [11] that manganite crystals produced by the described technique are like single crystals,[1] and, in this respect, they have far better crystal quality and far less porosity than the samples prepared by a solid-state reaction technique. On the other hand, the crystals have twin domain structure. X-ray powder diffraction pattern has testified a single-phase state of the sample.

The magnetization M of the sample studied was measured in a Faraday-type magnetometer. The DC resistance, as a function of temperature and magnetic field H (up to 16 kOe), was measured using a standard four-point probe technique. The RF technique employed is essentially the same as that used by some of us for ceramic cobaltites [7]. The sample for RF measurements (with diameter about 8 mm and length about 10 mm) was placed in an induction coil (9.7 mm in diameter and with a height about 22 mm). This coil is a part of an LC tank circuit. The magnitude of RF magnetic field in the induction coil was not more than 0.1 Oe. The measurements of the quality factor, Q, of the LC tank with and without the sample inside the coil were taken. The measurements of the Q-factor depending on temperature or magnetic field were taken at fixed frequency $\nu = 2.525$ MHz. The available cryostat with electromagnet makes it possible to measure the Q-factor in DC magnetic fields up to 2.5 kOe. The DC and RF fields were mutually perpendicular. The temperature and magnetic-field dependences of the Q-factor were recorded mainly on heating after the sample had been cooled down in zero field.

Consider shortly the physical properties which can be derived from the Q-factor measurements of the RF LC tank employed. The sample behavior in an electromagnetic field can be described, using complex permittivity $\varepsilon_c = \varepsilon' - i\varepsilon'' = \varepsilon'[1 - i\tan(\delta_\varepsilon)]$ (where $\varepsilon' = \varepsilon_r\varepsilon_0$, $\tan(\delta_\varepsilon) = \sigma_{RF}/(\varepsilon'\omega)$ is the dielectric loss tangent, σ_{RF} is the RF conductivity of the sample) and complex magnetic permeability $\mu_c = \mu' - i\mu'' = \mu'[1 - i\tan(\delta_\mu)]$, where $\mu' = \mu_r\mu_0$ and $\tan(\delta_\mu) = \mu''/\mu'$. For a plane electromagnetic wave, the complex resistance (impedance) of the sample is given by relation

$$Z_c = \sqrt{\frac{\mu_c}{\varepsilon_c}} = R_c + iX_c, \tag{2}$$

where resistance R_c defines the energy loss in the sample, and X_c is the sample reactance.

Once the sample with the wave resistance Z_c is inserted into the coil of the RF circuit, arranged from resistance, inductance and capacitance elements R_0, L_0 and C_0, a complex resistance is added to the circuit, given by $Z_{sp} = kZ_c$ where k is a geometrical factor determined by the sample and coil dimensions, which was about 0.35 in this study. The Q-factor of the circuit without the sample is expressed as $Q_0 = \omega L_0/R_0$. It is often more convenient to use the damping factor (or

[1] For example, our X-ray diffraction study of related manganite crystal, $La_{0.67}Ca_{0.33}MnO_3$, prepared in Institute of Physics, Warsaw, in the same floating-zone equipment has revealed that this crystal is very close to a single-crystal state although with twins which are inevitable arisen in manganites grown by floating-zone method.

decrement) $d = Q^{-1}$. The decrement of the circuit without the sample is, therefore, $d_0 = R_0/\omega L_0$. On inserting the sample into the coil the circuit decrement changes to $d = (R_0 + kR_c)/[\omega(L_0 + kL_c)]$. In the case that the sample causes merely small changes in the circuit inductance ($kL_c \ll L_0$) and the circuit quality without the sample is fairly high ($\omega L_0/R_0 \gg 1$), the expression for the difference $d - d_0 = P_A$ can be written as

$$P_A = k\frac{R_c}{\omega L_0} = k\frac{R_c}{R_0 Q_0}. \quad (3)$$

It is seen from it that P_A is determined by the loss in the sample. For the further analysis of the situation it is necessary to make suggestions about the amplitudes of dielectric and magnetic loss tangents. Taking $\tan(\delta_\varepsilon) \gg 1$ and $\tan(\delta_\mu) \ll 1$, the Eq. (3) can be presented in the form

$$P_A = k_0\left(1 + \frac{1}{2}\frac{\mu''}{\mu'}\right)R_s, \quad (4)$$

where $k_0 = k/(\omega L_0)$,

$$R_s = (\omega\mu'/2\sigma_{RF})^{1/2} = (\omega\mu'\rho_{RF}/2)^{1/2} \quad (5)$$

is the surface resistance, ρ_{RF} is the RF resistivity. According to Refs. [9,10], $\tan(\delta_\mu)$ is close to unity in polycrystalline samples of $La_{0.7}Sr_{0.3}MnO_3$ at the frequency used in this study. The same can be expected for the sample studied as well. In the case $\tan(\delta_\varepsilon) \gg 1$ and arbitrary magnitude of $\tan(\delta_\mu)$, the expression for P_A takes the form

$$P_A = k_0\left[\frac{\cos(\delta_\mu/2) + \sin(\delta_\mu/2)}{\sqrt{\cos(\delta_\mu)}}\right]R_s. \quad (6)$$

It is seen that in any case the loss behavior is determined mainly by that of the surface resistance R_s.

3. Results and discussion

The temperature dependence of DC magnetization, M, of the sample studied is shown in Fig. 1. The value of $T_c \approx 374$ K can be found if T_c is defined as the temperature of the inflection point in the $M(T)$ curve. The value of T_c and general behavior of $M(T)$ agree well with previous studies of $La_{1-x}Sr_xMnO_3$ samples with $x = 0.3 - 0.35$ [4,12,13]. The inset in Fig. 1 shows the magnetic-field dependence of the magnetization at $T = 290$ K. It is seen that the magnetization is close to saturation already at rather low field about 1 kOe. Taking into account a saturation value of M at higher fields, we have obtained the magnetic moment per formula unit at $T = 290$ K to be equal to $\mu_{fu} = 2.95\,\mu_B$, where μ_B is the Bohr magneton. Extrapolating $M(T)$ to low temperature limit ($T \to 0$) for higher fields enables us to obtain $\mu_{fu}(0) = 3.8 \pm 0.1\,\mu_B$. For nominal composition $La_{1-x}Sr_xMnO_3$ ($x = 0.33$) of the sample studied, taking into account that spin of Mn^{+3} is $S = 2$ and that of the Mn^{+4} is $S = 3/2$, μ_{fu} should be equal to $(4 - x)\mu_B$, that is to $3.67\,\mu_B$. This value agrees rather well with the estimated value, indicated above.

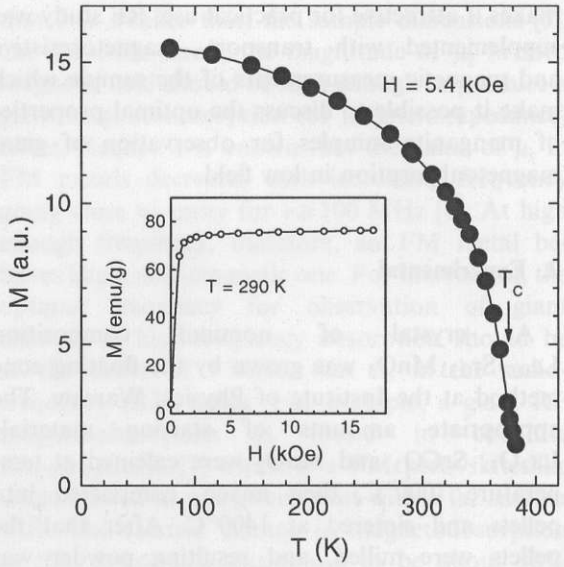

Fig. 1. Temperature dependence of the DC magnetization of the sample $La_{0.67}Sr_{0.33}MnO_3$ in magnetic field $H = 5.4$ kOe. The dependence was recorded with temperature increasing after the sample was cooled down to liquid nitrogen temperature, $T \approx 77.3$ K, in a field close to zero. The inset shows the magnetic field dependence of the magnetization at $T = 290$ K.

The temperature dependence of DC resistivity, $\rho(T)$, is shown in Fig. 2. The $\rho(T)$ behavior is metallic ($d\rho/dT > 0$) over the whole temperature range investigated, below and above T_c. The ρ magnitude decreases enormously (by a factor of 40) when going from 400 to 77 K (Fig. 2). The ρ

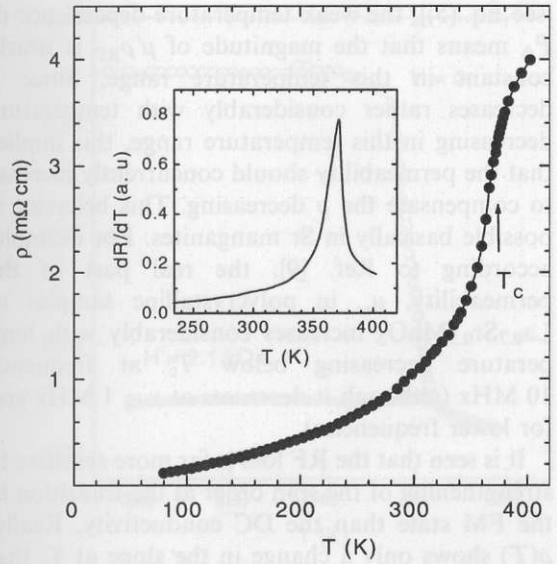

Fig. 2. Temperature dependence of the DC resistivity and its derivative $d\rho/dT$ (inset) of the sample studied.

values in low temperature range are fairly low (about 10^{-4} Ω cm). These are even less than those found in Ref. [12] in $La_{1-x}Sr_xMnO_3$ ($x = 0.3$) samples prepared (the same as in this study) by floating zone method, but are equal to those found in single-crystal samples of the same composition [13]. This is an evidence of excellent crystal perfection of the sample studied. The $\rho(T)$ dependence exhibits a change of slope at $T \approx T_c$ (see inset in Fig. 2) due to a contribution from the electron scattering on the spin disorder (in addition to the usual contributions from crystal lattice defects and electron–phonon scattering) [6]. This "magnetic" contribution to the resistivity, $\Delta\rho_m$, depends on the magnetization. The external magnetic field enhances the spin order (that is, the magnetization), which leads to a decrease in the resistivity, that is to negative MR.

The magnetic-field dependences of resistance of the sample studied are shown in Fig. 3. The temperature dependences of the MR at different amplitudes of H are presented in Fig. 4. Both these figures show that the MR depends strongly on temperature and is maximal near T_c. It goes down rather steeply for temperature deviating to either side from T_c. This temperature behavior of the MR is expected for FM manganites of fairly good

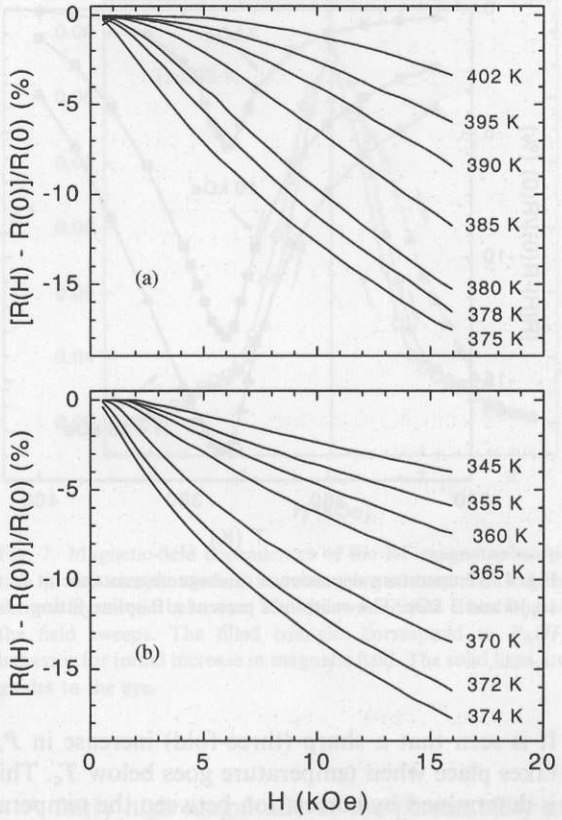

Fig. 3. Magnetoresistance curves of the sample studied for different temperatures above T_c (a) and below T_c (b).

crystal perfection. Indeed, the MR amplitude is determined by the ability of an external magnetic field to increase the magnetization. It is obvious that at low temperature ($T \ll T_c$), when nearly all spins are already aligned by the exchange interaction, this ability is minimal. For increasing temperature and, especially, at temperature close to T_c, the magnetic order becomes weaker (the magnetization goes down) due to thermal fluctuations. In this case the possibility to strengthen the magnetic order with an external magnetic field increases profoundly. This is the reason for maximal MR magnitude near T_c. Above T_c, the spin arrangement becomes essentially random, the magnetization is zero, and, therefore, the MR is close to zero as well.

Let us now take a look at behavior of the RF loss P_A in the sample studied. We begin with the temperature behavior $P_A(T)$ in zero field (Fig. 5).

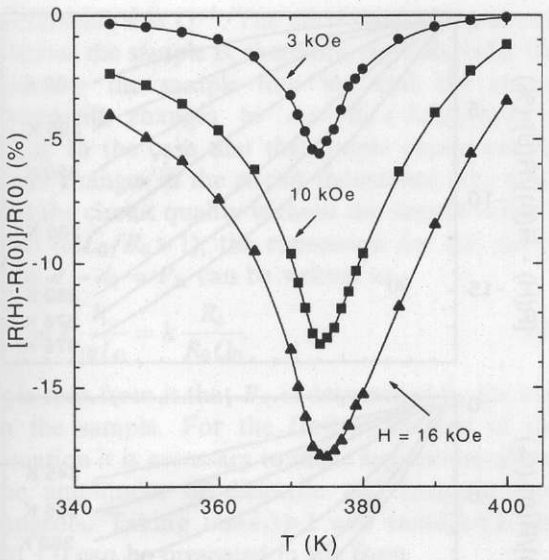

Fig. 4. Temperature dependence of magnetoresistance at $H =$ 16, 10 and 3 kOe. The solid lines present a B-spline fitting.

It is seen that a sharp (three-fold) increase in P_A takes place when temperature goes below T_c. This is determined by correlation between the temperature behavior of the loss and that of the permeability μ_r [5,7]. It is clear that far enough above T_c the relations $\mu_r = 1$ and $\mu'' = 0$ are true. In this case [see Eqs. (4)–(6)] the loss depends only on the conductivity, that is P_A should decrease with temperature decreasing. In mixed-valence manganites, the FM fluctuations (or developing of small FM regions in the paramagnetic matrix) can be evident even fairly far above T_c for extrinsic and intrinsic sources of magnetic inhomogeneity [3,14]. In that event, as T_c is approached from above, the slight increase in the permeability can occur even in the range above T_c. This will cause a corresponding increase in the loss. The drastic increase in P_A occurs, however, only quite near or somewhat below T_c when transition of the most of the sample volume to the FM state takes place inducing a sharp increase in μ'.

A decrease in temperature below T_c down to room temperature leads only to very weak changes in P_A (Fig. 5). In the case that the loss is determined mainly by the surface resistance R_s [see Eq. (5)], the weak temperature dependence of P_A means that the magnitude of $\mu'\rho_{RF}$ is nearly constant in this temperature range. Since ρ decreases rather considerably with temperature decreasing in this temperature range, this implies that the permeability should concurrently increase to compensate the ρ decreasing. This behavior is possible basically in Sr manganites. For example, according to Ref. [9], the real part of the permeability, μ_r, in polycrystalline samples of $La_{0.7}Sr_{0.3}MnO_3$ increases considerably with temperature decreasing below T_c at frequency 10 MHz (although it decreases at $\nu = 1$ MHz and for lower frequencies).

It is seen that the RF loss is far more sensitive to strengthening of the spin order at the transition to the FM state than the DC conductivity. Really, $\rho(T)$ shows only a change in the slope at T_c that means the steeper decrease in resistivity with temperature decreasing below $T < T_c$ (Fig. 2). In contrast to this, the RF loss increases sharply (by a factor of three) in the vicinity of T_c in zero field (Fig. 5). It is the change in the permeability that causes the large variations in the loss near T_c.

From measurements of the loss, P_A, at different magnetic fields we have obtained the values of the magnetoabsorption (MA), which has been defined as $[P_A(H) - P_A(0)]/P_A(0) = \Delta P_A(H)/P_A(0)$. Under suggestion, that the loss is determined primarily by the surface resistance R_s, and taking into account Eq. (5), a simplified expression for MA can be written as

$$\Delta P_A(H)/P_A(0) \approx \frac{R_s(H)}{R_s(0)} - 1 = \sqrt{\frac{\mu'(H)\rho_{RF}(H)}{\mu'(0)\rho_{RF}(0)}} - 1. \quad (7)$$

This shows the correlation between the $P_A(T)$ and $\mu'(T)$. It is clear from Eqs. (5) and (7) that owing to decrease in ρ_{RF} and μ' in an applied magnetic field, the MA should be negative, as it was really found in this study. We discovered an extremely high sensitivity of the RF loss to applied magnetic field (Figs. 5–7). It is remarkable that the strong magnetoabsorption effect shows itself in rather low magnetic fields. Not far from T_c the MA is about 67% in field $H = 2.1$ kOe (Fig. 6). By

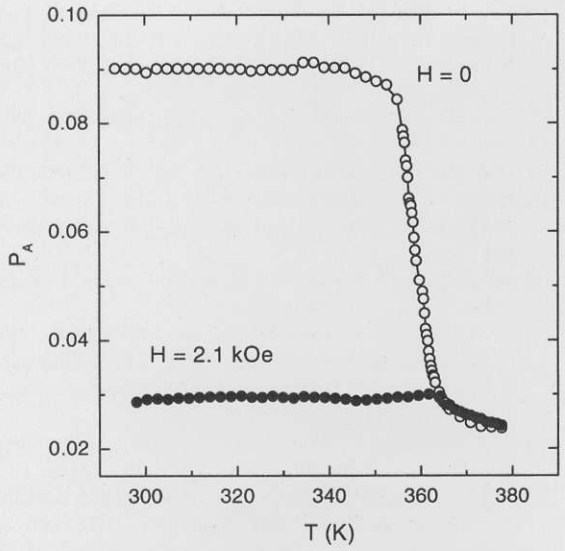

Fig. 5. Temperature dependences of RF absorption ($\nu = 2.525$ MHz) in the sample studied in zero field and in the field $H = 2.1$ kOe. $P_A = d - d_0$, where d and d_0 are the decrements of the used LC circuit with the sample in an inductance coil and without the sample in it, respectively.

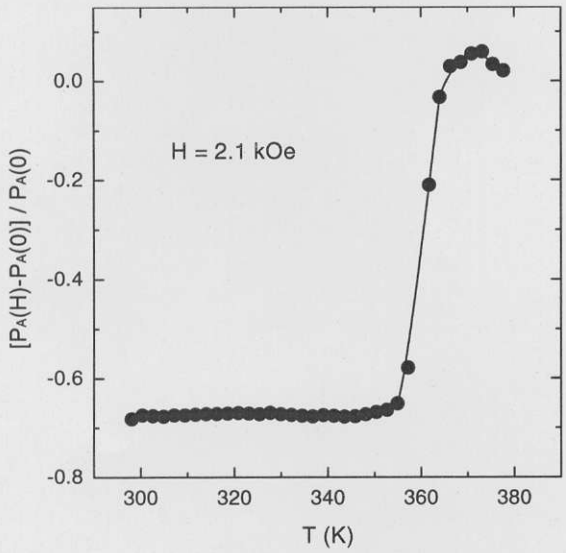

Fig. 6. Temperature dependence of the RF magnetoabsorption in the sample studied in field $H = 2.1$ kOe.

contrast, the DC MR is of only about 4% near T_c in the above-mentioned field (Fig. 3). The MA remains to be very high even for lower field. For example, in field 1 kOe the MA is about 55%.

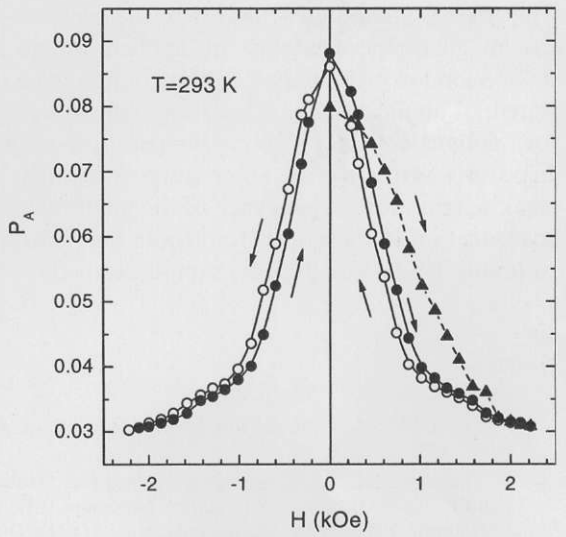

Fig. 7. Magnetic-field dependences of the RF magnetoabsorption in the sample studied at room temperature. The curves show a hysteretic behavior. The arrows indicate directions of the field sweeps. The filled triangles correspond to $P_A(H)$ behavior for initial increase in magnetic field. The solid lines are guides to the eye.

A fairly high magnitude of the permeability at zero field is a necessary condition to ensure giant MA (or magnetoimpedance) effect. In FM oxides this can be achieved only under the condition of their high crystal perfection. It is known, for example, that the permeability of ferrites decreases dramatically with increasing in porosity and decreasing in grain size [15]. The same is true for manganites. According to Ref. [10], the permeability of ceramic $La_{0.7}Sr_{0.3}MnO_3$ samples reduces severely with grain size decreasing. In this case high MA magnitude cannot be expected. For this reason, obviously, rather low (about 3%) magnetoimpedance was found in ceramic samples of $La_{0.67}Sr_{0.33}MnO_3$ prepared by solid-state reaction method. By contrast, in the sample studied prepared by floating-zone method, which has much better crystal perfection and negligible porosity, the MA is found to be really giant (close to 70%, as was indicated above).

In conclusion, we found the giant negative RF magnetoabsorption effect in bulk sample of $La_{0.67}Sr_{0.33}MnO_3$ under low DC magnetic fields.

The effect is attributed primarily to the decrease in the magnetic permeability in applied magnetic field. Such low-field magnetoabsorption may be of potential application in RF devices controlled by low magnetic field in room temperature range. Especially attractive for some purposes can be a weak temperature dependence of the giant magnetoabsorption in the range from room temperature to about 350 K found in the sample studied.

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