ISSN 2072-5981

Magnetic Resonance in Solids

Electronic Journal

Volume ***16,***

Issue ***2***

Paper No ***14201,***

1-9 pages

2014

http://mrsej.kpfu.ru

http://mrsej.ksu.ru

Established and published by Kazan University
Sponsored by International Society of Magnetic Resonance (ISMAR)
Registered by Russian Federation Committee on Press, August 2, 1996
First Issue was appeared at July 25, 1997

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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

Resonance dc phenomena in manganite thin films†

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(Received: February 19, 2014; accepted: April 19, 2014)

The resonance spin rectification (RSR) and resonance magnetoresistance (RMR) have been studied on $La_{2/3}Sr_{1/3}MnO_3$ epitaxial thin films in the temperature range of $300 - 365$ K, including the Curie point (T_C). The RSR effect is found to be caused by anisotropic magnetoresistance under conditions of the magnetic-resonance microwave pumping; it decreases upon heating and disappears at T_C. Unlike that, the RMR is maximal at T_C and shows clear correlation with the colossal magnetoresistance (CMR) of the material under study. The interpretation implies decreasing of the sample magnetization due to resonance saturation, thus leading to an increase in electric resistivity in terms of the CMR mechanism. Quantitative agreement is demonstrated between the experimental RMR data and theory accounting for Bloch-type relaxation in the vicinity of the phase transition.

PACS: 76.50.+g, 75.47.Gk, 76.30.-v, 75.40.-s

Keywords: ferromagnetic resonance, rare-earth manganites, thin films, magnetoresistance, spin rectification, magnetic relaxation

1. Introduction

This paper is a contribution to the book dedicated to the 80 years of Professor B.I. Kochelaev. One of the authors (V.A.A) has a pleasure to know Boris Ivanovich personally for about half a century and believes to be his friend. The main subject of B.I. Kochelaev's scientific activity is the theory of spin relaxation; in this field, his papers and lectures became classical. More specifically, his recent studies on rare-earth manganites have an impact on the work of various scientific groups worldwide. In the following paper, we present some new results obtained just on the manganite thin films, including the problems related to spin relaxation. Thus, though no references are given here to specific Kochelaev's publications, the authors acknowledge great positive influence of his scientific achievements.

Non-linear dc effects arising in conducting magnetic materials under conditions of resonance microwave pumping provide clear evidences for spin-charge interplay, thus opening a way to transfer electron spin properties to changes in electrical characteristics. Theoretical consideration of these phenomena was suggested by H.J. Juretschke as early as in 1960 [1], and soon realized experimentally [2]. More intense interest to this problem was attracted, however, after several decades, in the context of contemporary nanotechnology and prospect in spintronics [3-10]. At present, two main resonant dc phenomena in ferromagnets are known: a change in electrical resistivity (resonance magnetoresistance) and resonance spin rectification, called also microwave photovoltage, spin dynamo, magnetic-resonance electromotive force, etc. Physical mechanisms of these phenomena, as suggested in Refs. [1-10], are based on anisotropic magnetoresistance (AMR) and extraordinary Hall effect, as well as on trivial bolometric effect due to resonance microwave heating. Most of the above-cited studies were performed on conventional

†This paper is originally written by authors on the occasion of eightieth birthday of Professor Boris I. Kochelaev.

ferromagnetic metals such as nickel, permalloy, etc., at temperatures far below the Curie point (T_C). Unlike that, our work is concerned with investigation of the resonant dc effects in thin films of the doped rare-earth manganites $La_{1-x}Sr_xMnO_3$ (LSMO). This material is known to demonstrate the effect of colossal magnetoresistance (CMR) [11, 12], being prospective for the nano-structure applications [13]. The T_C value of doped manganites is not far from the ambient temperature, providing possibility of studying the dc effects close to the critical point. This paper reports on the experimental studies and theoretical interpretation of both resonant spin rectification (RSR) and resonant magnetoresistance (RMR) in the manganite films in the temperature range including T_C. Some early data were published previously, see Refs. [14, 15].

2. Experimental techniques

The samples under study were thin epitaxial films of $La_{2/3}Sr_{1/3}MnO_3$ with the thickness of 50–100 nm grown by laser ablation on single crystal $NdGaO_3$ substrates; for details, see Ref. [16]. The Curie point of the films was, as a rule, in the range of $345-350$ K, so they were ferromagnetic at room temperature.

The RSR and RMR measurements were performed with a home-made EMR[†] spectrometer (working at the frequency $\omega/2\pi \sim 9.5$ GHz) which allowed access of direct electrical current and additional microwave pumping with the power P up to 1 W. The film was placed horizontally in the central maximum of the microwave magnetic field h of the TE_{102} cavity (the loaded quality factor $Q_L = 400$). Platinum contacts were sputtered at the opposite ends of the film and soldered to thin wires brought out through small holes in the narrow walls of the cavity. Static magnetic field H could be rotated in the film plane, making an angle α with the line connecting the contacts. The voltage U arising between the potential contacts was registered under sweeping the field H across the EMR condition either without the external direct current ($I = 0$, the RSR effect), or at a fixed value of I up to 30 mA. In the latter case, the 4-contact method was employed for determination of the film resistivity R. To increase sensitivity, the pumping microwave power was modulated at the frequency of 100 kHz. As a result, the voltage U to be measured was modulated as well, allowing its lock-in amplification and detection. Computer processing and accumulation were used at the final stage; as a rule, the accumulations number was about 100.

Temperature dependencies were studied in the range of $300-365$ K that included the Curie point. Electrical heater was wound on the waveguide. The temperature was determined from the manganite film resistance which was preliminary calibrated with the accuracy of 0.5 K; thus, the film under study worked as a self-thermometer.

3. Experimental results

We start with the resonance spin rectification. Typical example of the RSR effect is shown in Fig. 1. As one can see, the signal $U(H)$ amounts to the μV range; it arises just at the FMR conditions and consists of symmetric and antisymmetric contributions, $U^s(H)$ and $U^{as}(H)$, which resemble the EMR absorption and dispersion lines, respectively. The sign of the voltage depends on the direction of the external field H, so the signal inverts when H is switched over, see the traces A and B in Fig. 1(a). This feature enables one to exclude extraneous contributions invariant to the sign of H, such as parasitic microwave detection etc., by using the half-difference $U_0(H) = [U_A(H) - U_B(H)]/2$, see Fig. 1(b). The signals showed in Fig. 1 were recorded at the

[†]We use the term EMR (electron magnetic resonance) which implies both ferromagnetic and paramagnetic resonances, FMR and EPR.

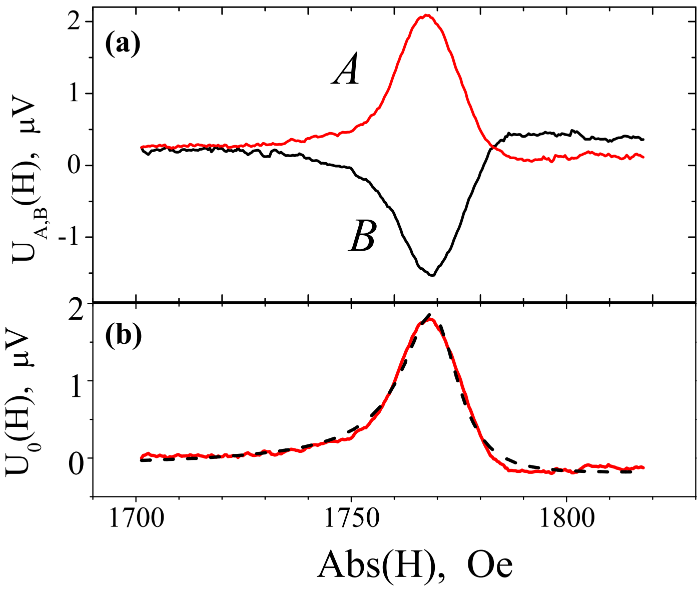

Figure 1. (a) The RSR voltage as recorded at opposite directions of external magnetic field H; $T = 308$ K, $\alpha = 45°$, $P = 400$ mW. (b) The half-difference of the traces A and B (solid line) and fitting with Eq. (3) (dashed line).

microwave power $P = 400$ mW. When P is altered, the RSR voltage $U_0(H)$ (measured at the same temperature) varies proportionally, without change in the line shape.

In Fig. 2, the amplitude U_0^s measured at $T = 308$ K is plotted against the angle α. The angular dependence looks rather specific, showing maxima at about $\pm 54°$ and zeros at 0 and 90°. Note that the data can be well fitted by the function of $\sin(\alpha)\sin(2\alpha)$, see the dashed curve in Fig. 2.

Temperature dependence of the amplitude U_0 (measured at $\alpha = 45°$) is presented in Fig. 3 by open circles (the right scale). The RSR voltage is maximal at room temperature and demonstrates a steep fall toward zero when approaching T_C.

Unlike the above-described RSR effect which was studied in the absence of external current, the measurements of magnetoresistance were based on the Ohm Law, $R = U/I$, where $I =$ const was supplied by an external source with large enough series resistor $r \gg R$. The resistance R of our manganite films was in the range of $100 - 200$ Ohm at 300 K and increased monotonically by about 3 times upon heating to 360 K, with maximum slope at T_C [14, 15]. The static field $\mathbf{H}$ was kept perpendicular to the microwave field $\mathbf{h}$ ($\alpha = \pi/2$), providing maximum EMR absorption. The RMR effect appeared as a small increase ΔU in the voltage when sweeping through the resonance conditions, corresponding to the resistance increment ΔR of the order of 10^{-4}–10^{-3} Ohm. To exclude any foreign contributions, the half-difference was used of the $U(H)$ signals recorded at opposite current directions. Typical $\Delta R(H)$ line is shown in Fig. 4, where the derivative of the FMR absorption is given for comparison. The ΔU magnitude was found to be proportional both to the current I and microwave power P.

Fig. 5(a) presents the temperature dependence of the resonance magnetoresistance for two manganite films. The data are normalized to the resonance absorption, with account made for the EMR line width and magnitude. It is seen that, in contrast with Fig. 3, the normalized resonance magnetoresistance increases when approaching the phase transition, attains its maximum at T_C and persists even in paramagnetic phase. This can be compared with the temperature

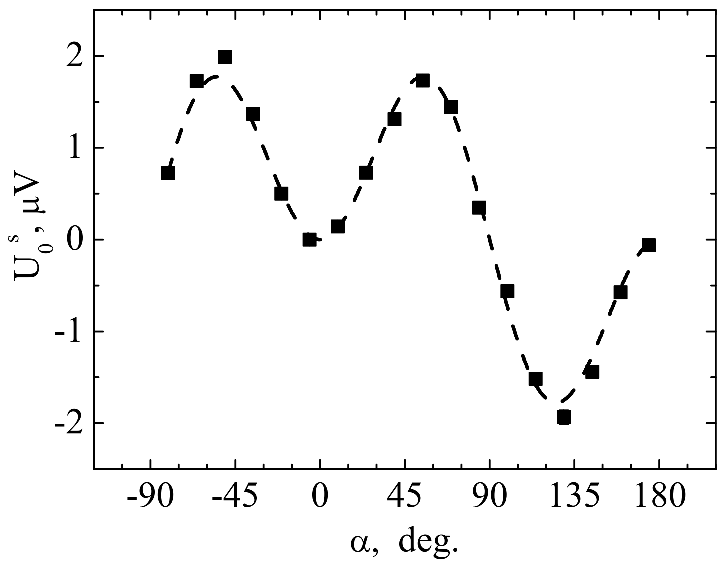

Figure 2. Angular dependence of the RSR voltage at T = 308 K. The dashed line represents the $\sin(\alpha)\sin(2\alpha)$ dependence.

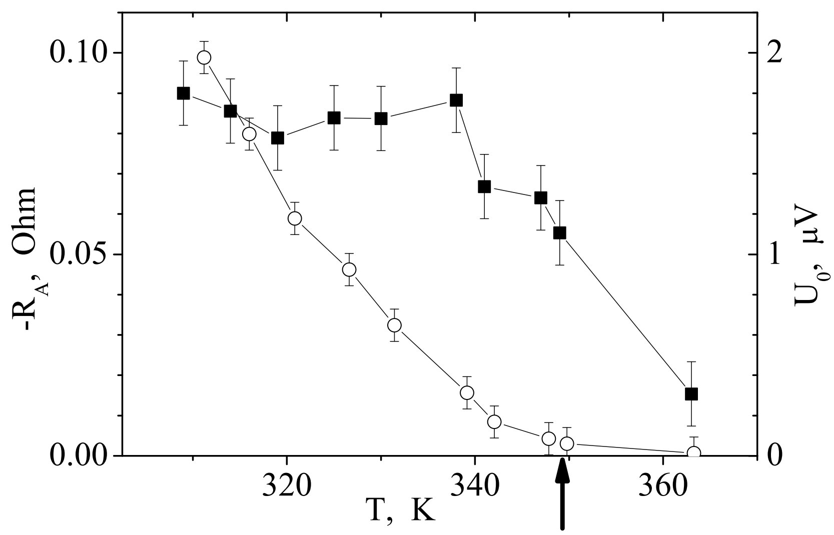

Figure 3. The RSR magnitude (open circles, right scale) and anisotropic part of the sample resistance (filled squares, left scale) as functions on temperature. The lines are the guides for eyes. The arrow indicates T_C.

dependence of the colossal magnetoresistance effect (CMR) as measured directly on the same films, see Fig. 5(b). The correlation of the two effects is evident.

Along with the above-described main set of experiments, the EMR spectra of the samples were recorded under the same conditions as in the RSR and RMR measurements. As it will be seen below, both the resonance field H_0 and the EMR line-width δH are of importance for quantitative interpretation of the data obtained. In particular, the H_0 values were used to calculate equilibrium magnetization M_0, according to the formula [17]

$$\omega^2 = \gamma^2 H_0 \left(H_0 + 4\pi M_0\right), \tag{1}$$

where γ is the gyromagnetic ratio. For the films under study, the M_0 values at 300 K are about

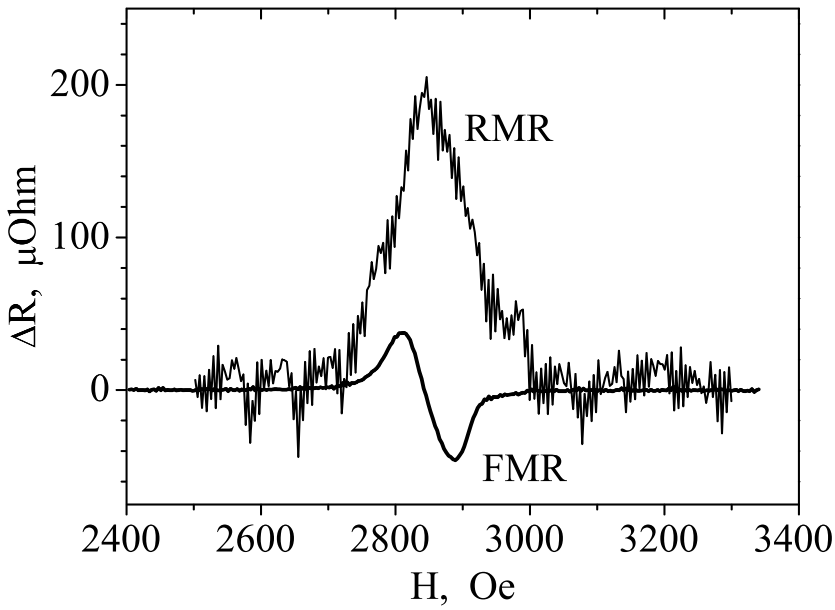

Figure 4. Resonance magnetoresistance recorded by sweeping through the FMR condition at $T = 330$ K, $P = 120$ mW. The FMR absorption derivative (in arbitrary units) is shown for comparison.

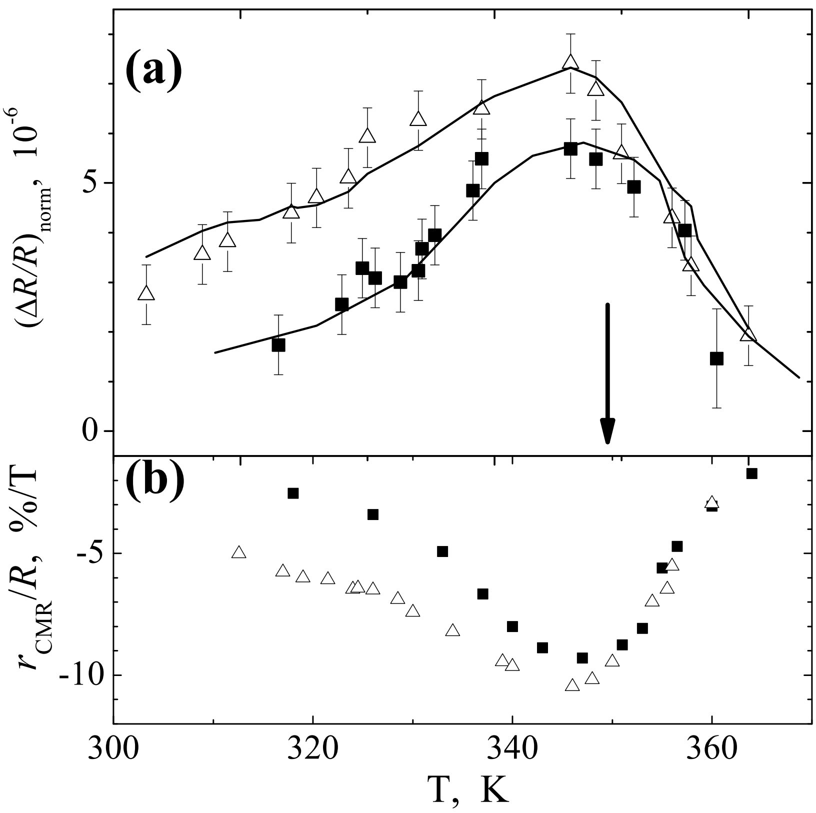

Figure 5. Temperature dependence of the normalized RMR magnitude (a) and differential CMR factor (b) measured on two manganite films. The curves are calculated with Eq. (5), see the text. The arrow indicates T_C.

300 Oe and decrease strongly when approaching T_C. As to the FMR line-width, it increases upon heating from 20-30 Oe at room temperature up to about 200 Oe near the phase transition, where δH passes through the maximum. More detailed data on EMR in the manganite films are reported in Refs. [14-16].

Further, the anisotropic part of the sample resistance R_A was determined directly as the difference between the R values measured at parallel ($\alpha = 0$) and perpendicular ($\alpha = \pi/2$) directions of H relative to the current I. At room temperature and $H \sim 2$ kOe, the negative R_A values in the range from -0.1 to -1.0 Ohm were found in various samples. At elevated temperatures, the absolute values of R_A decreased and could not be measured with proper accuracy.

4. Discussion

To interpret the data on the resonance spin rectification, we will follow the theory developed in Refs. [1, 2, 6], where two sources of the dc voltage arising under EMR conditions were considered: the anisotropic magnetoresistance (AMR) and extraordinary Hall effect (EHE). In our case, the latter mechanism should be neglected for two reasons. First, the angular dependence of EHE differs from that shown in Fig. 2; second, the shape of the ESE signal should be nearly antisymmetric, in contrast to Fig. 2, where the $U^s(H)$ contribution obviously predominates. Thus, only the AMR mechanism of the spin rectification will be analyzed.

Following the approach used in Ref. [6], the dc voltage U_0 originates from the Ohm Law applied to the microwave current $I_1 \cos(\omega t)$ (which determines the z axis) and anisotropic resistance $R = R_0 + R_A \cos^2\theta$, where θ is the angle between the current and magnetization $\mathbf{M}$. The non-linearity arises from the term

$$U_{nl}(t) = I_1 \cos\omega t \cdot R_1 \cos(\omega t + \phi), \tag{2}$$

where the oscillating resistance with the amplitude R_1 is caused by the oscillations of θ in the course of the resonant Larmor precession. The time-averaged part of Eq. (2) is just the dc voltage observed experimentally.

Thus, one should compare our data with the formulas deduced in Ref. [6] at various assumptions on the geometry of microwave currents and magnetic fields. Taking into account the $\sin(\alpha)\sin(2\alpha)$ angular dependence (Fig. 2), the only appropriate combination is

$$U_{0z}(H) = \frac{R_A I_1}{2M_0} A_{xx} \sin\alpha \sin(2\alpha) \left[-h_z^i g_a(H) + h_z^r g_d(H)\right], \tag{3}$$

where A_{xx} is the numerical factor depending on H_0, M_0 and δH; h_z^r and h_z^i are the amplitudes of the in-phase and out-of-phase components of $h_z(t)$ as related to the microwave current; $g_a(H)$ and $g_d(H)$ are the form-factors corresponding to the EMR absorption and dispersion lines, respectively. Note that, according to Eq. (3), the dc voltage U_{0z} is proportional to the microwave power, in agreement with the experiment.

The dashed curve in Fig. 1(b) presents the fitting of the observed $U_0(H)$ line with Eq. (3) (in arbitrary units, since the I_1 value is unknown). The best fit was obtained at $h_z^i/h_z^r = 3.0$, corresponding to the phase shift of 72°; this determines the ratio of the symmetric and antisymmetric components of the RSR signal.

Further, the temperature dependence $U_0(T)$ (see Fig. 3, the right scale) together with Eq. (3) enables one to calculate the temperature dependence of the anisotropic part of the sample resistance. Making use of the directly measured value of R_A at the room temperature and the EMR data obtained at different temperatures, the $R_A(T)$ plot was drawn (Fig. 3, the left scale). It is seen that the anisotropic magnetoresistance decreases steeply near the phase transition and practically disappears above T_C. Note that the negative sign of R_A in our samples was confirmed by both the direct measurement at the room temperature and relative polarities of H and $U_{A,B}$.

Passing to the results on resonance magnetoresistance, one can notice a principal difference with the above-described RSR data. In fact, the RMR effect not only does not disappear near T_C, but, on the contrary, reaches there its maximum. So, one has to look for another explanation of the observed RMR phenomenon, not reduced to the anisotropic resistance. As an alternative mechanism, the colossal magnetoresistance (CMR) may be considered, which is known to be a specific feature of the manganite materials [11, 12].

The CMR effect consists in considerable decrease of electrical resistivity due to an increase of the absolute value (the length) of the magnetization vector $\mathbf{M}$, caused, in its turn, by increasing of the external magnetic field H. We suppose that partial saturation of EMR by the resonant microwave pumping leads to M decreasing and, as a result, to an increase in resistivity. This suggestion is confirmed by correlation between the RMR and CMR in the temperature range under study: as seen in Fig. 5, both effects attain their extremal values near T_C and persist in the paramagnetic phase.

It should be emphasized that the commonly used Landau-Lifshits Equation with the Gilbert relaxation term conserves the length of $\mathbf{M}$ [17] and hence excludes applicability of the above-suggested mechanism. However, as shown by D. Garanin and his co-workers [18, 19], an additional Bloch-type relaxation term should arise due to thermal fluctuations in the vicinity of the phase transition; as a result, the so-called Landau-Lifshits-Bloch (LLB) equation was suggested. At small deviation from equilibrium and weak magnetic anisotropy, it can be represented in the form

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma \left[\mathbf{M} \times \mathbf{H}\right] - T_{1abs}^{-1} \left(M - M_0\right) \frac{\mathbf{M}}{M} - {T_2}^{-1} \frac{\left[\mathbf{M} \times \left[\mathbf{M} \times \mathbf{H}\right]\right]}{M \cdot H_0}, \tag{4}$$

where the second term describes the Bloch-type longitudinal relaxation along $\mathbf{M}$ with the characteristic time T_{1abs}. Note that $\mathbf{H}$ and H_0 designate here the full and static magnetic fields, respectively.

The LLB equation (4) was solved under conditions of partial (weak) saturation of EMR by microwave power, resulting in expression for the decrement ΔM as a function on experimental and material parameters, such as M_0, H_0, δH, h, and T_{1abs}. Then, making use of the differential CMR value $r_{CMR} = dR/dH$ and "absolute susceptibility" $\chi_{abs} = dM/dH$, one gets for the relative increment of the resistance, normalized over the resonant microwave absorption [15]:

$$\frac{\Delta R_{res}}{R} = -\frac{\gamma h^2 T_{1abs} r_{CMR}}{2\chi_{abs}} \tag{5}$$

All parameters entering Eq. (5), with the only exception for T_{1abs}, were determined by independent measurements. Besides, the ratio T_{1abs}/χ_{abs} should not change significantly near the Curie point [19], so the both quantities can be replaced by their values at a fixed temperature. The best fits of the temperature dependencies for two samples were attained at $T_{1abs}(T_C) = (2.1 \pm 0.3)$ nsec; the fitting curves are shown in Fig. 5(a) by solid lines, demonstrating a good agreement. Note that the employed value of the relaxation time is consistent with the experimental data reported in Refs. [20, 21].

In conclusion, two resonant dc effects, RSR and RMR, have been observed and studied on thin epitaxial films of $La_{2/3}Sr_{1/3}MnO_3$ in the temperature range including the Curie point. It is shown that the mechanism of the resonant spin rectification is determined by the anisotropic magnetoresistance which decreases upon heating and disappears in paramagnetic phase. Unlike this, the resonance magnetoresistance of the manganite films under study increases when approaching the critical temperature and passes through its maximum at T_C, showing strong correlation with the CMR effect. The suggested interpretation accounts for the CMR phenomenon together with partial reduction of the magnetization vector upon microwave pumping in the presence of the Bloch-type relaxation. As a result, quantitative agreement with the experiment is achieved.

Acknowledgments

The work was partially supported by the RFBR Grants 11-02-00349 and 14-02-00165. The authors are grateful to G.A. Ovsyannikov and A.M. Petrzhik for providing with the manganite films under study.

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