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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEM

Resonant Magnetoresistance in the Vicinity of a Phase Transition

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Abstract—The change in the electrical conductivity of manganite films upon microwave pumping in the magnetic resonance conditions is investigated. The temperature dependence of the effect correlates with the temperature variation of colossal magnetoresistance (CMR), passing through a maximum at the Curie point. The results are interpreted using a model that assumes a decrease in the absolute value $|\mathbf{M}|$ of the magnetic moment of the sample under the action of magnetoresonant saturation, which leads to an increase in resistance in accordance with the CMR mechanism. Theoretical analysis based on the Landau–Lifshitz–Bloch equation confirms the correctness of this model and ensures good agreement with experiment.

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1. INTRODUCTION

The interrelation between spin (magnetic) and transport (electric) phenomena is one of the central topics of contemporary studies in the physics of condensed media. The development of this trend has led in recent years to important discoveries and applications, such as the giant and colossal magnetoresistance (GMR and CMR) effects, nanosize devices for magnetic control over current, and spintronics elements. Magnetoresonance effects and methods occupy a special place in this rapidly developing field of science (we will henceforth use the generalized concept "electronic magnetic resonance" (EMR) including ferromagnetic (FMR) and paramagnetic (PMR) resonances). The first publications devoted to the effect of magnetoresonant pumping on the electric properties of ferromagnets appeared in the 1960s [1, 2]. At that time, researchers realized that such results can be interpreted as electric detection of magnetic resonance (EDMR). A number of spin–electric phenomena have been discovered and investigated by now; their mechanisms include anisotropic magnetoresistance of ferromagnets, the Hall effect in a resonant microwave field, and the elementary bolometric effect associated with additional heating of the sample due to resonant absorption of microwave power [3–8]. The first attempt at EDMR due to colossal (isotropic) magnetoresistance in rare-earth manganites was undertaken in [9], where the observation of significant changes in the voltage across a ceramic current-carrying sample under ferromagnetic resonance conditions was reported. However, the interpretation of these results was subsequently questioned, and the EDMR due to the CMR effect was reliably observed on a thin $La_{0.67}Sr_{0.33}MnO_3$ epitaxial film [10]. Clear correlation between the temperature dependences of both effects was demonstrated, and a physical interpretation was proposed associated with the emergence of the Bloch relaxation mechanism in the vicinity of the Curie point of the ferromagnetic material. The goal of our paper is the further development of this research (in particular, a more comprehensive and correct comparison of experimental data with the result of applying the Landau–Lifshitz–Bloch (LLB) equation in the modern theoretical interpretation [11, 12] and reliable separation of the bolometric effect and other mechanisms of electrical detection of EMR).

2. THEORETICAL PREMISES

It has been established that the peculiar properties of rare-earth manganites doped by bivalent metals A^{2+} (the general formula is $La_{1-x}A_xMnO_3$) are mainly due to so-called double-exchange between Mn^{3+} and Mn^{4+} ions with intervention of the oxygen ion separating them [13, 14].[1] The probability of an electron jump between the exchange participants has been found to depend on the mutual orientation of the spins of manganese ions; this probability attains its maximal value when the spins have the same direction. As a result, the electric conductivity of manganites increases significantly with the external magnetic field, which enhances spin polarization. This is precisely the CMR effect, which is observed in the ferromagnetic as well as in the paramagnetic state and is manifested most clearly in the vicinity of the phase transition (Curie point T_C). The CMR effect in rare-earth manganites was studied by many researchers (see, e.g., reviews [16–18], monograph [19], and references therein).

[1] More complex mechanisms can be associated with phase separation, oxygen vacancies, etc. (see, e.g., [15] and references therein).

The idea of employing the CMR under magnetic resonance conditions is based on the assumption that (even partial) saturation of resonance with a microwave field must lead to a certain decrease in the absolute value of the magnetic moment of the sample (length of vector **M**). This means that the deviation from collinearity of the spins at adjacent manganese ions occurs, which leads to a decrease in the electric conductivity via the double exchange mechanism. In the paramagnetic phase, such an interpretation is obviously valid: according to the Bloch equations (see, e.g., [20]), saturation of electron paramagnetic resonance (EPR) indeed suppresses the spin polarization and reduces the value of $|\mathbf{M}|$. It can easily be shown that in the case of weak saturation, the relative change in the modulus of the magnetic moment is given by

$$-\frac{\Delta|\mathbf{M}|}{M_0} \equiv s = (\gamma H_1)^2 T_2^2\left(a - \frac{1}{2}\right), \qquad (1)$$

where M_0 is the equilibrium magnetization, γ is the gyromagnetic ratio for the electron, H_1 is the half-amplitude of the resonant microwave film, and $a = T_1/T_2$ is the ratio of the longitudinal and transverse spin relaxation times. It should be emphasized that saturation factor $s \ll 1$, which was introduced by relation (1), characterizes the decrease in the length of vector **M** and not its projection M_z onto the direction of external magnetic field $\mathbf{H}_0$, as is usually assumed in the literature (in this case, the term $-1/2$ in parentheses in Eq. (1) is absent).

The situation in the ferromagnetic phase is more complicated. At low temperatures, all spins of the sample are oriented in the same direction and form the unified ferromagnetic moment **M** precessing about effective field $\mathbf{H}_e$. This situation is reflected in the Landau–Lifshitz equation with the Hilbert relaxation term [21], which can be written in the form

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma[\mathbf{M}\times\mathbf{H}_e] - \gamma\alpha\frac{[\mathbf{M}\times(\mathbf{M}\times\mathbf{H}_e)]}{M^2}, \qquad (2)$$

where α is the relaxation parameter. The dissipative term in this relation is perpendicular to vector **M** and, hence, does not affect its length. Supplementing constant field $\mathbf{H}_e$ with a small alternating field $2\mathbf{H}_1\exp(i\omega_{\mathrm{FMR}}t)$ orthogonal to it and oscillating at the FMR frequency, as well as confining the solution of Eq. (2) to the first approximation ($H_1 \ll H_e, M, \alpha$), we can easily see that FMR saturation is only reduced to an increase in the precession angle, but it does not affect the length of vector **M** [22]. This means that the spins belonging to individual ions preserve mutual parallelism (precess in phase) so that the electrical conductivity should not change and EDMR effect is not expected.

The situation changes when the temperature of the ferromagnet increases and approaches the phase-transition temperature ($T \longrightarrow T_C$). Thermal fluctuations violate the strict order in the mutual orientation of spins; modulus $|\mathbf{M}|$ decreases as compared to its limiting low-temperature value and becomes a function of external magnetic field H_0. This gives rise to the CMR effect. In these conditions, resonant magnetoresistance can appear (i.e., the conductivity can change upon partial saturation of FMR by the field of microwave pumping).

Let us estimate the magnitude of the expected effect using the equation of motion proposed in [11, 12]. Random varying fields produced by thermal fluctuations are included in Eq. (2) in these publications; after theoretical analysis based on the Fokker–Planck equation in the mean field approximation, this leads to the so-called Landau–Lifshitz–Bloch (LLB) equation. At temperatures differing from the Curie point insignificantly, this equation can be written in the form [12]

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma[\mathbf{M}\times\mathbf{H}_e] + \gamma\alpha_{\parallel}\frac{(\mathbf{M}\cdot\mathbf{H}_e)\mathbf{M}}{M^2} - \gamma\alpha_{\perp}\frac{[\mathbf{M}\times(\mathbf{M}\times\mathbf{H}_e)]}{M^2}, \qquad (3)$$

where $\alpha_{\parallel}$ and $\alpha_{\perp}$ are the parameters characterizing the longitudinal and transverse relaxation, respectively. The expressions for these parameters and cumbersome formulas for effective field $\mathbf{H}_e$ obtained in [11, 12] are not given here for the sake of brevity. Presuming small deviations from equilibrium and weak anisotropy, we can reduce Eq. (3) to the form

$$\frac{\partial \mathbf{M}}{\partial t} = -\gamma[\mathbf{M}\times\mathbf{H}_0] - \frac{\gamma\alpha_{\parallel}}{\chi_{\mathrm{abs}}}(M - M_0)\frac{\mathbf{M}}{M} - T_2^{-1}\frac{[\mathbf{M}\times(\mathbf{M}\times\mathbf{H})]}{M\cdot H_z}, \qquad (4)$$

where **H** is the external magnetic field (its constant component $\mathbf{H}_0$ determines the direction of the z axis), T_2 is the transverse relaxation time defined in the standard manner, and

$$\chi_{\mathrm{abs}} = \frac{\partial|\mathbf{M}|}{\partial H_0} \qquad (5)$$

is the "absolute" magnetic susceptibility reflecting the change in the magnetic moment modulus under the action of a constant external field. Henceforth, we will omit subscript "0" in this expression to simplify notation. It should be noted that the second (Bloch) term on the right-hand side of Eq. (4) describes the variation of the modulus of magnetic moment **M** with characteristic relaxation time

$$T_{1\mathrm{abs}} = \frac{\chi_{\mathrm{abs}}}{\gamma\alpha_{\parallel}}. \qquad (6)$$

Including the resonant microwave field in **H** and solving Eq. (4) in the first approximation in saturation factor $s \ll 1$, we obtain

$$s = -\frac{\Delta|\mathbf{M}|}{M_0} = (\gamma H_1)^2 T_{1\mathrm{abs}}T_2. \qquad (7)$$

Let us now derive the relations suitable for direct quantitative estimation of the magnitude and temperature dependence of the resonance magnetoresistance. The sought increment ΔR_{res} of the sample resistance upon resonant saturation can be written in the form [10]

$$\Delta R_{res} = \frac{\partial R}{\partial H}\left(\frac{\partial|\mathbf{M}|}{\partial H}\right)^{-1}\Delta|\mathbf{M}| = -\frac{r_{CMR}}{\chi_{abs}}\frac{\gamma H_1^2 T_{1abs} M_0}{\delta H}, \tag{8}$$

where the second equality was obtained using relation (7). Here, the first factor on the right-hand side, which will be henceforth denoted by r_{CMR}, is the negative differential magnetoresistance associated with the CMR effect; it can be directly measured in experiment. The halfwidth δH of the magnetic resonance absorption line, which is substituted into Eq. (8) for T_2^{-1}/γ, is also a measurable quantity. Knowing the Q factor of the resonant cavity and the microwave power, we can easily calculate the value of H_1. Thus, it remains for us to estimate the ratio T_{1abs}/χ_{abs}, which is completely determined by quantity $\alpha_{\|}$ in accordance with relation (6).

It was shown in [12] that the temperature dependence $\alpha_{\|}(T)$ has no singularities near T_C; therefore, at temperatures close to the phase-transition temperature, the value of $\alpha_{\|}$ can be assumed approximately constant (i.e., we can set $T_{1abs} \propto \chi_{abs}$). This is in complete agreement with the physical picture. In particular, upon a decrease in temperature, both these quantities tend to zero, which agrees with the behavior of an ideal ferromagnet: the value of $|\mathbf{M}|$ is fixed and is restored almost instantaneously upon any deviation from equilibrium. On the other hand, the values of T_{1abs} and χ_{abs} are "fitted" upon a transition through T_C with longitudinal relaxation time T_1 and longitudinal susceptibility typical of the paramagnetic phase. Thus, assuming that ratio T_{1abs}/χ_{abs} is constant, we can replace temperature-dependent parameters T_{1abs} and χ_{abs} appearing in relation (8) by their values at a certain fixed temperature. We will use this approach when comparing the results with experimental data.

3. EXPERIMENTAL TECHNIQUE

We studied $La_{0.67}Sr_{0.33}MnO_3$ epitaxial films 50–100 nm in thickness, which were grown by laser ablation on substrates made of $NdGaO_3$ single crystals in the (110) orientation [23]. Detailed investigations showed [23, 24] that mechanical stresses appearing during the growth of the films lead to changes in the unit cell parameters and to axial magnetic anisotropy with characteristic values 100–200 Oe of the anisotropy field. At room temperature, the films were in the ferromagnetic phase; the Curie temperature T_C was 345–350 K. The resistance of the films was measured under resonant pumping using four-contact and two-contact circuits (with identical results) at a fixed direct current I. The film was placed at the maximum of the microwave field of a rectangular resonant cavity of the TE_{102} type with $Q = 400$; it was connected to the circuit of a homemade EPR spectrometer (with frequency $\omega/2\pi = 9.5$ GHz) permitting variation of the sample temperature in the required range. Constant magnetic field $\mathbf{H}_0$ and high-frequency field $\mathbf{H}_1(t)$ perpendicular to it were in the film plane; the measuring current I was approximately collinear with $\mathbf{H}_1$. It should be noted that the film thickness was much smaller than the skin depth at frequency ω, which ensured uniformity of field $\mathbf{H}_1(t)$ in the bulk of the sample. Microwave pumping (up to 500 mW) was provided by a magnetron. The power fed to the resonator was modulated by a meander at a frequency of f_m = 100 kHz and a modulation depth of about 100%. Voltage U across the potential contacts of the film (sputtered platinum) was applied to the input of the SR844 RF lock-in amplifier, which selected and detected the alternating voltage component at frequency f_m. This signal was accumulated and recorded in computer storage during periodic passage of magnetic field H_0 through the EMR region (period was 16 s, and the number of passages was up to 200).

The same contacts were used for calibration measurements of the resistance as a function of temperature and magnetic field. The $R(T)$ dependence was then used in the main set of experiments for measuring the film temperature (with an error of ±0.2 K). The $R(H_0)$ dependence characterizing the CMR effect made it possible to determine the differential parameter r_{CMR} required for computations (see relation (8)).

Apart from main measurements, standard EMR spectra of the object under investigation were also recorded on the same setup (as well as on commercial Bruker spectrometer). The examples can be found in [10]. The shift of the resonance line relative to the Larmor field $H_{00} = \omega/\gamma$ due to the demagnetizing field of the film made it possible to calculate equilibrium magnetization M_0 using the standard formula [21]. The resonance shifts due to crystal magnetic anisotropy and the anisotropy induced by mechanical stresses were much smaller [23, 24] and were disregarded. The width of the FMR line at room temperature varied from sample to sample over a wide range (20–100 Oe), indicating the extent of inhomogeneity of the films. When the temperature approaches T_C, these differences were suppressed to a considerable extent due to additional broadening attaining its maximal value (up to 200 Oe) at the transition point.

4. RESULTS AND DISCUSSION

The experiments were mainly performed on two samples denoted conditionally as S422 and S670 (the designation is associated only with the serial number

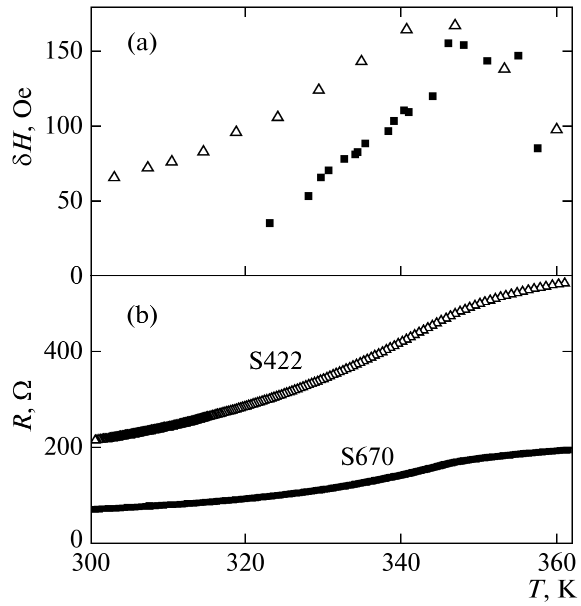

Fig. 1. Temperature dependences of (a) the EMR line halfwidth and (b) resistance of samples S422 (△) and S670 (■).

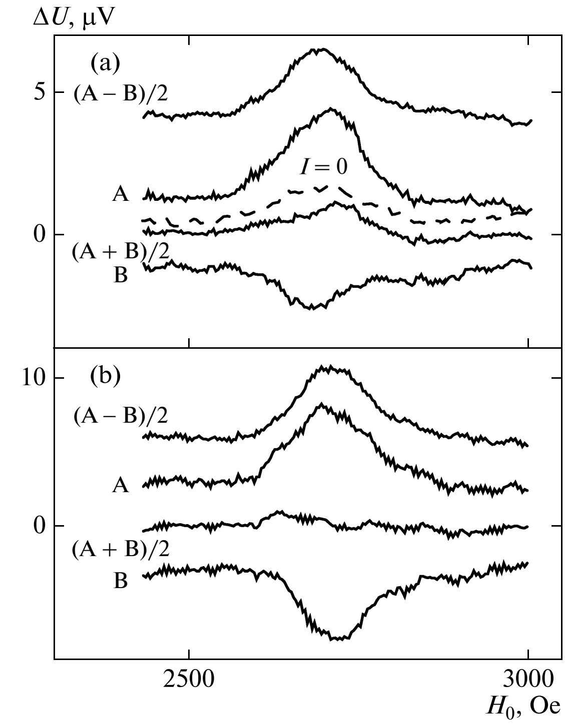

Fig. 2. Signals of electric detection of magnetic resonance in sample S670 at $T = 329$ K: (a) $I = 12$ mA, $P = 320$ mW; (b) $I = 20$ mA, $P = 93$ mW. Letters A and B correspond to opposite directions of the current. The dashed curve is the signal for $I = 0$. The signals are displaced along the vertical for better visualization.

and is otherwise meaningless). The choice of these films from a large batch described in [23, 24] was dictated by the requirement of the highest structural homogeneity characterized by the minimal values of the magnetic resonance linewidth. The temperature dependences of the halfwidth of the EMR line and the film resistance are shown in Fig. 1. In both samples, the value of δH passes through the maximum near the critical temperature $T_C \approx 348$ K. It can be seen from the figure that sample S670 is characterized by lower values of δH and R, indicating a higher homogeneity and a larger film thickness. Magnetization M_0 of both films, measured from the shift of the EPR line, decreased monotonically from 250–300 Oe at 300 K to 10–20 Oe in the phase transition region.

The typical signals demonstrating additional voltage ΔU emerging in the film when it passes through resonance under microwave pumping are shown in Fig. 2 (sample S670). The signals recorded for the opposite directions of current I are denoted by A and B. It can be seen from the figure that when the direction of the current is reversed, the signal changes its sign, but its amplitude is not preserved, the difference being the stronger the smaller the current (cf. Figs. 1a and 1b). This means that the dependence $\Delta U(I)$ deviates from the Ohm law. Analysis shows that signal ΔU can be written as the sum of two components,

$$\Delta U = \Delta U_R + \Delta U_0, \quad (9)$$

where the first (Ohmic) term $\Delta U_R = I\Delta R$ is proportional to the current and characterizes the increase in the film resistance under resonant pumping, while the second (static) term is almost independent of I. In particular, signal ΔU_0 is also observed when $I = 0$ (dashed curve in Fig. 2a). Both these components of the signal are proportional to the microwave power P.

To suppress the effect of the static contribution, the experiments were mainly performed at the maximal admissible currents I; to eliminate this effect completely, each experiment was repeated for opposite values of the current, after which the sought value of ΔU_R was determined as the half-difference of the recorded signals (see Fig. 2). The half-sum of the same signals gives the value of ΔU_0, which almost coincides with the signal recorded at zero current (see Fig. 2a). Detailed analysis and discussion of the origin of the ΔU_0 signal is beyond the scope of this article; some preliminary results will be considered at the end of this section.

Figure 3 shows a typical signal of resonance magnetoresistance in comparison with the EMR line recorded for the same conditions by the standard modulation method. The measured value of ΔU_R is recalculated to the relative variation of film resistance, $\Delta R/R$, which attains a value of 2.8×10^{-6} at exact resonance.

For convenience of comparison with the theory, all such signals were reduced to the same value of the microwave power and then normalized to the resonance absorption coefficient $\chi''_{res} = M_0/(2\delta H)$. As noted above, the values of M_0 and δH can be deter-

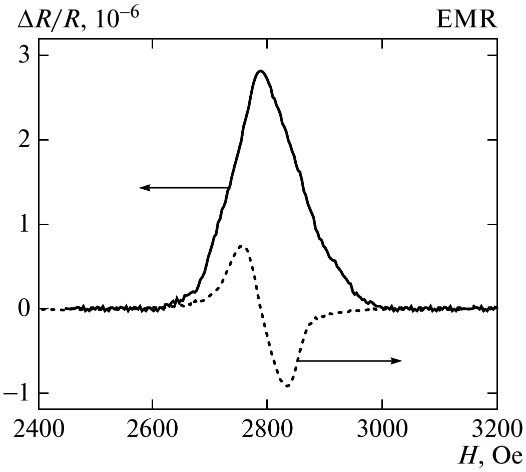

Fig. 3. Resonance magnetoresistance signal for $I = 23$ mA and $P = 116$ mW (solid curve) and conventional EMR line (dotted curve) recorded on sample S670 at $T = 331$ K.

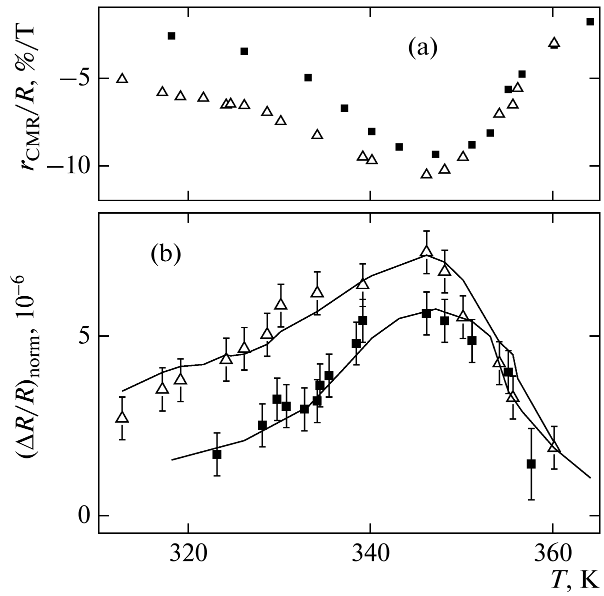

Fig. 4. Temperature dependences of (a) differential CMR and (b) normalized resonance magnetoresistance on sample S422 (△) and S670 (■). Solid curves correspond to calculation by formula (11) for $H_1 = 0.6$ Oe (S422) and 0.45 Oe (S670).

mined from the EMR spectra (see Fig. 1). The normalized values

$$\left(\frac{\Delta R}{R}\right)_{\rm norm} = \left(\frac{\Delta R}{R}\right)_{\rm res}\frac{2\delta H}{M_0} \tag{10}$$

are plotted in Fig. 4b as functions of the temperature. It can be seen that for both samples the effect is observed on both sides of the critical temperature, passing through the maximum near T_C. Figure 4a shows for comparison the temperature dependences of the relative differential CMR effect ($r_{\rm CMR}/R$) in percent per tesla.

To compare the results with theoretical predictions, solid curves in Fig. 4b show the results of computations based on the formula derived from relations (8) and (10):

$$\left(\frac{\Delta R}{R}\right)_{\rm norm} = -2\gamma\frac{T_{\rm 1abs}(T_C)}{\chi_{\rm abs}(T_C)}\frac{r_{\rm CMR}H_1^2}{R}, \tag{11}$$

where the temperature dependent quantities $T_{\rm 1abs}$ and $\chi_{\rm abs}$ are replaced by their fixed values measured near T_C. Such a substitution is possible owing to the approximate proportionality of the calculated values of these quantities (see Section 2).

The theoretical curves are plotted using the data from Fig. 4a and the values of H_1 computed for experimental values of microwave power (given in the figure caption). The value of $\chi_{\rm abs}(T_C) \approx 0.03$ was estimated from the slope of the magnetization curve (see [10]). Relaxation time $T_{\rm 1abs}$ passing into conventional longitudinal relaxation time T_1 at the critical temperature remained the only free parameter, which was determined from the considerations of the best fitting of the theoretical curve to experimental data. The values of $T_{\rm 1abs}(T_C)$ determined in this way were 1.8 and 2.4 ns, respectively, for films S422 and S670; we can assume that these values coincide with an error of 20%. These values are in good agreement with times T_1 measured directly in the vicinity of the critical temperature by longitudinal detection on a batch of ceramic samples of manganites with different compositions [25–27]. It should be noted that the saturation factor calculated by formula (7) for both samples under our experimental conditions is on the order of 10^{-4}; therefore, condition $s \ll 1$ is obviously satisfied. Thus, we can speak of satisfactory quantitative agreement between experimental data and the results of theoretical calculations based on the model of colossal magnetoresistance in combination with the LLB equations.

Let us now consider the reasons for the emergence of "static" signals ΔU_0. It was proposed in [10] that these signals appear due to nonlinearity of the current–voltage characteristics of current contacts during their detection. However, this mechanism is hardly compatible with the independence of ΔU_0 of current I determining the bias at the contacts. On the other hand, conducting ferromagnets exhibit fundamental nonlinearities associated with anisotropic magnetoresistance and manifestations of the Hall effect [1–8]. It is well known that these nonlinearities may lead to detection (rectification) of the microwave power under FMR conditions (including zero bias current). Without going into details, we note that the emf sign in this case is determined by the direction of the static magnetic field; therefore, signal ΔU_0 must reverse upon switching of $\mathbf{H}_0$. Such a behavior was indeed observed for some of our samples. Analysis of this effect forms the subject of a separate publication.

Another side effect that can be superimposed on the resonance magnetoresistance signals and that can lead to an incorrect interpretation is associated with additional heating of the film due to resonant absorption of the microwave field. Such a bolometric detection of FMR was described in detail in [4, 5]. Analysis of heat balance based on the heat conduction equation shows [10] that in our experimental conditions, the characteristic times of transient processes in the film exceed the period of the meander modulating the microwave power. It follows hence that with increasing modulation frequency, the amplitude of temperature oscillations at this frequency must decrease and their phase must increasingly lag behind the phase of saturating pulses. Our measurements performed in the modulation frequency range (50–200 kHz) have demonstrated the constancy of the amplitude and phase of the resonance magnetoresistance signals. Therefore, the influence of the bolometric effect can be disregarded in this case.

5. CONCLUSIONS

Thus, the experiments on epitaxial films of rare-earth manganites confirmed the increase in the resistance of the material under the action of microwave pumping under the EMR conditions. It is found that experimental data are in quantitative agreement with the results of theoretical model based on the dependence of the resistance on the absolute value of the magnetic moment (CMR effect). We used the Landau—Lifshitz—Bloch equation, which makes it possible to take into account the effect of the "Bloch" relaxation contribution, including its temperature dependence in the vicinity of the phase transition. Thus, our results can be treated not only as a new method of electric detection of EMR, but also as an experimental confirmation of the LLB model proposed in [11, 12]. It should be noted in this connection that the specific temperature dependences and the absolute values of parameters χ_{abs} and T_{1abs} predicted from theoretical considerations in these publications differ from the dependences observed in our experiments. This is apparently due to the simplified analysis of the LLB equation in the mean field approximation as well as with peculiarity and complexity of phase transitions in rare-earth manganites; the theory of these phase transitions has not been completely developed as yet.

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