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## Magnetoresonance EMF in Thin Manganite Films

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**Abstract**—The effect of magnetoresonance emf (MREMF) in thin epitaxial films of rare-earth manganites  $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$  is detected and investigated. The effect is manifested in the occurrence of a constant voltage under the action of microwave pumping in magnetic fields corresponding to ferromagnetic resonance conditions. The MREMF signal includes symmetric and antisymmetric components and changes its polarity upon switching of the external magnetic field. The temperature dependence of the effect (including the range in the neighborhood of the phase transition) is analyzed. The experimental data including the shape of the signal and its dependence of the field orientation are in good agreement with the results obtained in the theoretical model based on the mechanism of anisotropic magnetoresistance. It is shown that the magnetoresistance anisotropy in the manganite under investigation is negative and sharply attenuates as the temperature approaches the Curie point, almost vanishing in the paramagnetic phase.

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### 1. INTRODUCTION

The interaction between the spin (magnetic) and charge (transport) properties and phenomena is one of the central problems in contemporary solid state physics. Among the most significant advances in this field, we can mention the discovery and numerous applications of the effects of giant and colossal magnetoresistance, the development of nanosize devices for magnetic control over current, and the conception and development of spintronics. Magnetoresonance methods occupy an important position in this vast field. The effect of magnetoresonant pumping on the properties of conducting ferromagnets was first predicted theoretically and discovered experimentally in the 1960s [1, 2]. In recent years, the intensity of such investigations has grown significantly due to advances in nanotechnology and prospects of spintronics [3–15].

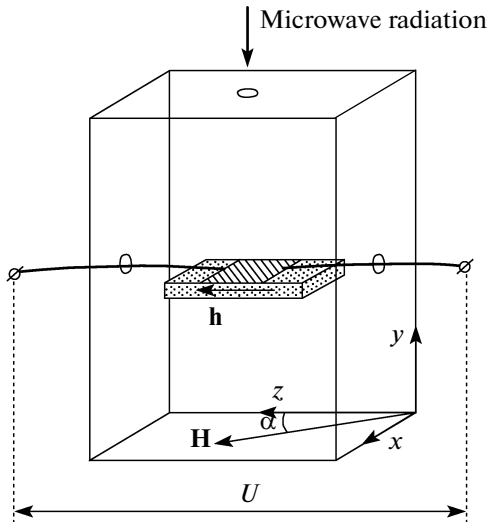
This work is devoted to one of the spin–charge phenomena, viz., the occurrence of a constant potential difference under the action of microwave radiation under ferromagnetic resonance (FMR) conditions. From here on we refer to this effect as the magnetoresonance electromotive force (MREMF); it is also called the microwave photoeffect, spin rectification, spin dynamo, etc. The physical nature of MREMF in conducting ferromagnets was discussed in detail even in the first publications [1, 2], where two mechanisms of the occurrence of this effect were considered: anisotropic magnetoresistance and the anomalous Hall effect. Later, the so-called inverse spin Hall effect [3] associated with the occurrence of the spin current through the interface between ferromagnetic and nor-

mal metals under FMR conditions (“spin pumping”) was added [4]. A characteristic feature of all these mechanisms is the change in polarity of the MREMF upon switching of the direction of a constant magnetic field. Subsequently, the theoretical description of these phenomena was modified and matched with experimental conditions [6, 12]. The effect found a number of applications from characterization of the microwave field parameters [10] to detection of magnetic resonance in spintronic devices [15].

In contrast to previous investigations of MREMF performed on standard ferromagnetic metals, this study was carried out on samples of doped rare-earth manganites possessing unique properties and a peculiar phase diagram (see reviews [16, 17]). This study is aimed at clarifying the physical nature of the effect and especially the manifestation and characterization of anisotropic magnetoresistance of manganite films in the temperature range that includes the magnetic phase transition (Curie point  $T_C$ ). This problem is especially topical in connection with the role played by anisotropic magnetoresistance in physics and applications of film nanostructures, including the application of  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$  manganites, for which  $T_C$  exceeds room temperature only slightly [17–19].

### 2. EXPERIMENTAL

We studied epitaxial films composed of  $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$  with a thickness from 50 to 140 nm, grown by laser ablation on various single-crystal substrates [20]. At room temperature, the films were in the



**Fig. 1.** Diagram of arrangement of sample in rectangular cavity and system of coordinates used. Vectors  $\mathbf{H}$  and  $\mathbf{h}$  indicate directions of constant and microwave fields.

ferromagnetic phase; Curie temperature  $T_C$  was 345–350 K.

The experimental geometry is illustrated in Fig. 1, which also shows the Cartesian system of coordinates that will be used below. The film on the substrate (the area of the surface was  $5 \times 5$  mm) was arranged horizontally at the central antinode of microwave magnetic field  $\mathbf{h}$  in a rectangular cavity with the  $TE_{102}$ -type mode (the  $Q$  factor of the cavity loaded with the sample was  $Q \approx 400$ ). The potential difference was measured along the  $z$  axis parallel to the wide wall of the cavity using platinum contacts sputtered on the opposite edges of the film. The cavity was placed between the poles of an electromagnet that could be rotated about the vertical axis so that constant magnetic field  $\mathbf{H}$  remained in the plane ( $xz$ ) of the film, forming angle  $\alpha$  with the  $z$  axis. Voltage  $U$  between the contacts was led out by thin horizontal wires emerging from the cavity through small holes in its narrow walls. Nominally, microwave field  $\mathbf{h}$  for the given mode in the central part of the cavity should also be parallel to the  $z$  axis. It will be shown below, however, that the presence of the ferromagnetic film with contact wires attached to it introduces some distortions into the geometry. The electromagnet had a switch for reversing polarity, which was used to record the signal for opposite directions of the constant magnetic field.

The thickness of the films was much smaller than the skin depth at the resonant pumping frequency  $f = \omega/2\pi \approx 9$  GHz, which ensured uniformity of field  $\mathbf{h}$  in the bulk of the sample. Microwave pumping (up to 400 mW) was ensured by Gann diodes or a magnetron. The power supplied to the cavity was modulated by a meander with a modulation frequency of  $f_m = 100$  kHz and a modulation depth of about 100%. Voltage  $U$  from the potential contacts of the film was fed to the

input of an SR844 lock-in amplifier, which selected the varying component of the voltage at frequency  $f_m$ . After lock-in detection with respect to a reference voltage of the same frequency, the signal accumulated upon periodic sweep of magnetic field  $H$  through the FMR region (with a period of 16 s and a number of accumulations from 10 to 1000).

Along with the main measurements, the standard FMR spectra of the samples were also recorded under the same conditions. The position of the center ( $H_0$ ) and halfwidth ( $\Delta H$ ) of the FMR line were compared with MREMF signals and used to interpret the results. In particular, the equilibrium magnetization  $M_0$  was calculated for each preset temperature  $T$  from the shift of the resonance line due to the demagnetizing field of the film in accordance with the well-known formula [21]

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

where  $\gamma$  is the gyromagnetic ratio.

It should be noted that the use of this formula presumes the collinearity of vectors  $\mathbf{H}$  and  $\mathbf{M}_0$  lying in the plane of the film. This means that we disregard the axial magnetic anisotropy fields ( $H_u \approx 100$ – $200$  Oe) occurring during film growth [20, 22], as well as weaker fields of the crystal (cubic) anisotropy as compared to demagnetizing field  $4\pi M_0$ . The use of a more accurate formula [22] taking into account the above-mentioned small corrections gives approximately the same results.

The temperature of the sample was controlled by a wire electrical heater wound on the microwave cavity. The temperature sensor was the experimental film itself, whose resistance was preliminarily calibrated to within  $\pm 1$  K in the temperature range 295–365 K. Typical  $R(T)$  and  $M_0(T)$  dependences for such films are given in [11, 20].

### 3. EXPERIMENTAL RESULTS

The main batch of measurements was taken on two samples with relatively narrow FMR lines recorded at room temperature. This indicated the high quality of the films and ensured the best MREMF signal-to-noise ratio. Some parameters of the experimental films are given in the table. The results for both samples did not differ qualitatively, but the effect was considerably stronger for sample no. 1 with a narrower line.

Figure 2a shows typical MREMF signals obtained upon passage of the magnetic field through resonance. The signals recorded for opposite directions of the constant magnetic field are denoted  $A$  and  $B$ . It can be seen that after reversal of the field polarity, the signal changes its sign, as expected for the MREMF effect. More detailed analysis shows, however, that the shape of the signal slightly changes upon its inversion. This points to the admixture of a signal independent of the direction of field  $\mathbf{H}$ . In particular, this can be due to a

certain nonlinearity of measuring contacts, leading to trivial rectification of the microwave signal and, as a consequence, to conventional detection of magnetic resonance line. Evidently, this “parasitic” contribution cannot change upon switching of the constant magnetic field polarity, so it can be easily eliminated by using the half-difference of signals *A* and *B*,  $U_0(H) = (A - B)/2$  (see Fig. 2b). This “purified” signal was used for further analysis.

Figure 2 shows that the MREMF signal is asymmetric and resembles the superposition of magneto-resonant absorption and dispersion lines. The deconvolution of  $U_0(H)$  into symmetric and antisymmetric components  $U_0^s(H)$  and  $U_0^{as}(H)$  was performed using the formulas based on the Lorentz approximation of the line shape (see Section 4 below). As a rule, the  $U_0^s(H)$  component dominated.

Analysis of the dependence of signal  $U_0(H)$  on microwave pumping power *P* revealed that its shape remains unchanged, while amplitude  $U_0$  is proportional to the irradiation power.

Figure 3 shows the angular dependences of amplitudes  $U_0^s$  for two samples. It can be seen that the amplitude passes through maxima at  $\alpha \approx \pm 55^\circ$  and vanishes at  $\alpha = 0$  and  $90^\circ$ . The angular dependence is successfully approximated by a function proportional to  $\sin \alpha \sin(2\alpha)$  (curves in Fig. 3).

The temperature dependence of amplitude  $U_0^s$  is shown in Fig. 4. It can be seen from the graph that as the temperature approaches the critical value ( $T_C = 347$  K), the MREMF signal sharply decreases and almost vanishes in the paramagnetic phase. It will be shown in the next section that these data can be recalculated into the temperature dependence of the anisotropic part of the magnetoresistance (see Fig. 4).

It should be noted that the anisotropic part  $R_a$  of the magnetoresistance of the samples, which is defined as the difference between the resistances measured for the parallel and perpendicular orientations of external magnetic field **H** relative to the measuring current, was estimated directly at room temperature. The values of  $R_a$  for both samples turned out to be negative; the magnitude of ratio  $R_a/R$  did not exceed tenths of a percent. In view of the considerable error, such measurements were not taken at higher temperatures.

4. DISCUSSION

Let us consider the mechanism of occurrence of MREMF due to anisotropic magnetoresistance using the simple approach proposed in [6]. It should be recalled that the phenomenon of anisotropic magnetoresistance is manifested in the dependence of resistance *R* of the material on angle  $\theta$  between magnetiza-

tion vector **M** and the direction of current **I** in accordance with the formula [1, 2]

$$R = R_0 + R_a \cos^2 \theta, \tag{2}$$

where  $R_0$  is the isotropic part of the resistance. In our experimental conditions including resonant irradiation at frequency  $\omega$ , the potential difference across the measuring sample region is given by

$$U(t) = I(t)R(t), \tag{3}$$

where  $I(t) = I_1 \sin \omega t$  is the microwave current in the sample and  $R(t) = R_0 + R_1 \sin(\omega t + \varphi)$  is the resistance consisting of the constant and oscillating terms. The latter is due to the anisotropic term in formula (2) because angle  $\theta$  under FMR conditions oscillates due to precession of vector **M** about the magnetic field direction. Formula (3) implies that  $U(t)$  contains con-

Sample no.	Substrate*	Film thickness, nm	<i>R</i> , Ω	<i>M</i> <sub>0</sub> , Oe	Δ <i>H</i> , Oe
1	NGO	80	70	303	10
2	LSAT	140	101	319	55

\* NGO = NdGaO<sub>3</sub>; LSAT = (LaAlO<sub>3</sub>)<sub>0.3</sub> + (Sr<sub>2</sub>AlTaO<sub>6</sub>)<sub>0.7</sub>.

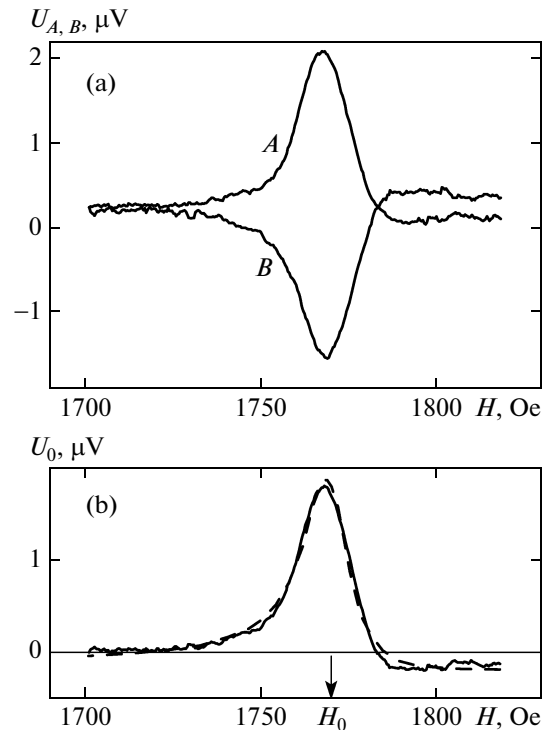
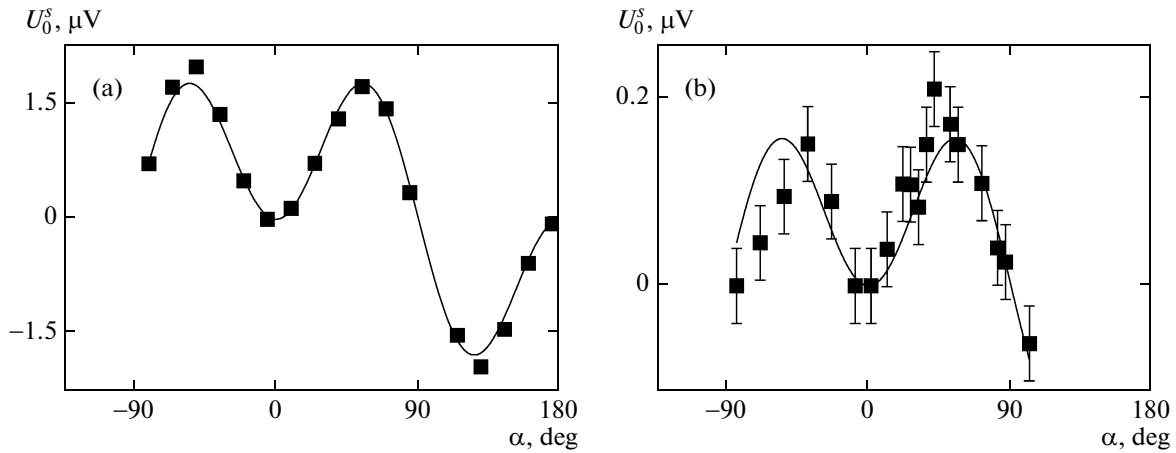


Fig. 2. (a) MREMF signals (*A* and *B*) recorded for opposite directions of constant magnetic field; (b) their half-difference (dashed curve corresponds to the approximation based on formula (4)). The arrow indicates position of the FMR line. Sample no. 1,  $T = 309$  K,  $P = 400$  mW,  $\alpha = 45^\circ$ .



**Fig. 3.** Dependence of the amplitude of the symmetric part of MREMF signals in sample nos. 1 (a) and 2 (b) on the angle between the constant magnetic field and the direction of measurement of the potential difference at  $T = 310$  K,  $P = 400$  mW. The curves describe a dependence of the type  $\sin\alpha\sin(2\alpha)$ .

stant component  $U_{dc} = \langle U(t) \rangle$ , where angle brackets indicate averaging over time.

In publications by different authors [1, 2, 6, 12], the magnitude and shape of MREMF signals was calculated in detail taking into account the mechanisms of anisotropic magnetoresistance, anomalous Hall effect, and spin pumping for various versions of experimental geometry. Comparing the theoretical formulas in these publications with experimental data shown in Fig. 3, we can see that the only version that ensures the observed angular dependence of the form  $\sin\alpha\sin(2\alpha)$  is associated with the mechanism of anisotropic magnetoresistance with the microwave magnetic field directed along the  $z$  axis. The corresponding expression for the dc voltage occurring along the same axis

under FMR conditions in a thin film [6] can be written in the form

$$U_{dc}(H) = \frac{R_a I_1 (H_0 + M_0)}{2(2H_0 + M_0)} \sin\alpha \sin(2\alpha) \times g(H) \left( -h_z^i + h_z^r \frac{H - H_0}{\Delta H} \right), \tag{4}$$

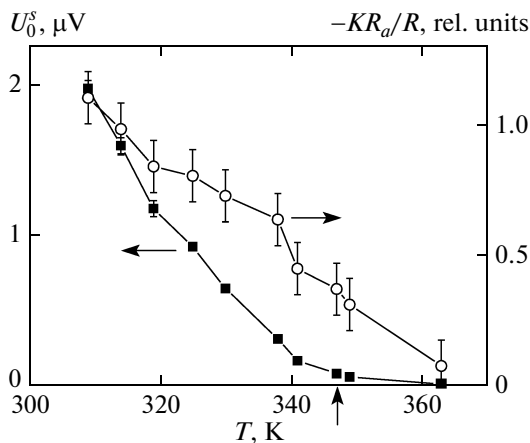
where  $h_z^r$  and  $h_z^i$  are the amplitudes of the real (syn-phase) and imaginary (out-of-phase relative to the current) parts of the  $z$  projection of the microwave magnetic field and

$$g(H) = \frac{\Delta H}{\Delta H^2 + (H - H_0)^2}$$

is the Lorentz form factor of the FMR absorption line.

It should be emphasized that allowance for other microwave field components leads to an essentially different angular dependence of the effect. For example, for the  $h_x$  projection, factor  $\sin\alpha$  on the right-hand side of expression (4) is replaced by  $\cos\alpha$ , which is in striking contrast with the results depicted in Fig. 3. At the same time, the existence of the  $z$  component of the microwave current does not match the structure of the  $TE_{102}$  mode if we disregard the distortions introduced by the sample under study and connecting wires. We assume that the leading role in the formation of the  $z$  component of the microwave current in our samples is played by horizontal segments of the copper wire leading constant voltage  $U$  from the potential contacts (see Fig. 1). In this case, microwave currents are induced in the wires due to perturbations in the structure of the resonant mode, and the circuit is closed by the capacitors formed when the wires pass through small holes in the cavity walls.

Let us continue the comparison of expression (4) with experimental data. It should be noted above all



**Fig. 4.** Temperature dependences of the amplitude of the symmetric part of the MREMF signal (■) and of the relative anisotropic magnetoresistance (○;  $K$  is an arbitrary positive coefficient), Sample no. 1,  $P = 400$  mW,  $\alpha = 45^\circ$ . The arrow indicates the Curie point.

that products  $I_1 h_z^i$  and  $I_1 h_z^r$  on the right-hand side of this expression are proportional to the microwave pumping power. Such a dependence of  $U_0$  on the power was confirmed in experiment. Further, formula (4) contains the symmetric and antisymmetric contributions with relative weights determined by the phase shift of field  $h(t)$  relative to the microwave current. Approximation of the shape of the  $U_0(H)$  signal with the help of expression (4), shown in Fig. 2b, corresponds to the ratio  $h_z^i/h_z^r = 3.0$  (phase shift  $72^\circ$ ). It can be seen from the figure that it is in good agreement with experiment. It should be noted that the relative contribution of the antisymmetric component was not strictly constant in our experiments and varied slightly depending on angle  $\alpha$  and temperature. It cannot be ruled out that this is due to manifestations of other microwave field components, but the smallness of the effect does not allow us to draw a final conclusion.

The specific angular dependence  $\sin\alpha\sin(2\alpha)$  is convincing evidence in favor of the anisotropic magnetoresistance mechanism as the main factor responsible for the occurrence of MREMF in our samples. The absence of the contribution from the anomalous Hall effect, which is characterized by an angular dependence of another type and predominance of the antisymmetric component of the resonant signal [1, 2], is obviously associated with suppression of the  $y$  component of vector  $\mathbf{M}$  due to the strong demagnetizing field of the thin film [12]. As regards the spin pumping effect [3, 4, 12], it is manifested only in a ferromagnet/normal metal two-layer structure. The role of such a structure could be played by platinum contacts sputtered onto the surface of the manganite film; however, the area of these contacts is small and their thickness (on the order of 100 nm) considerably exceeds the optimal values [12]. Nevertheless, this question requires further investigation.

Let us now consider the temperature dependence of MREMF. For this purpose, we compare our experimental data on the temperature dependence of  $U_0^\circ$  (see Fig. 4) with formula (4). Since parameters  $M_0$ ,  $H_0$ , and  $\Delta H$  appearing in expression (4) are known from experiment, it is possible to calculate the temperature dependence of product  $R_a I_1$ . Unfortunately, the absolute values of  $R_a$  cannot be determined in this way because amplitude  $I_1$  of the microwave current remains unknown. We can assume, however, that it varies with temperature in inverse proportion to the total resistance  $R$  of the film, so that product  $R_a I_1$  is proportional to the relative anisotropic magnetoresistance  $R_a/R$ . The temperature dependence of this quantity calculated to within a constant factor  $K > 0$  using formula (4) is also shown in Fig. 4. Taking into account the experimental estimate of  $R_a$  at room temperature, we can conclude that the magnetoresistance anisotropy in the ferromagnetic phase of the manganite

is negative and does not exceed tenths of percent; it decreases as the temperature approaches the Curie point and almost vanishes when the substance is transformed into the paramagnetic state.

## 5. CONCLUSIONS

Thus, the occurrence of a constant potential difference in thin manganite films under the action of microwave pumping under ferromagnetic resonance conditions (MREMF effect) has been detected and investigated. It was found that the experimental data are in good agreement with the theoretical model based on the dependence of the sample resistance on the projection of magnetization onto the direction of electric current (anisotropic magnetoresistance). The shape of the MREMF signal and the dependence of its amplitude on the orientation of the external magnetic field are in conformity with the theory. The temperature dependence of the effect has been investigated for the first time. The results have demonstrated a sharp decrease in the coefficient of the anisotropic magnetoresistance in the vicinity of the phase transition.

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