08

Magnetoresistive characteristics of the $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructure

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We report on results of experimental studies of thin-film heterostructure $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ grown epitaxially on $NdGaO_3$ substrate. A transitional layer is formed at the interface between paramagnetic semimetal $SrIrO_3$, exhibiting strong spin-orbit interaction, and the spin-polarized ferromagnet $La_{2/3}Sr_{1/3}MnO_3$. Single layers of $SrIrO_3$ and $La_{2/3}Sr_{1/3}MnO_3$ were studied as well. The temperature dependences of electronic conductivity, magnetoresistance, and Hall effect responses obtained at magnetic fields $H = 0-7$ T and temperatures $T = 2-300$ K are discussed.

Keywords: heterostructure, spin-orbit interaction, strontium iridate, manganite, magnetoresistance, Hall effect.

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With the epitaxial growth of oxide films, it becomes possible to create an interface with functional characteristics radically different from those of contacting materials, for example, the formation of a two-dimensional electron gas in a heterostructure with an interface between $LaAlO_3/SrTiO_3$ insulators [1]. Recently, heterostructures consisting of magnetics and materials containing metals with high atomic weight and, consequently, with strong spin-orbit coupling (SOC) have attracted increased interest. An anomalous Hall effect was detected [2] in the boundary layer of paramagnetic strontium iridate $SrIrO_3$ due to the induced magnetic moment from manganite $La_{2/3}Sr_{1/3}MnO_3$. The authors of Ref. [3] reported the occurrence of a Dirac response in $SrIrO_3$ upon contact with an antiferromagnet $SrCuO_2$. The high energy of the SOC $SrIrO_3$ $E_{SO} \sim 0.5$ eV [4] contributes to the conversion into electric current of the spin current generated by spin pumping during ferromagnetic resonance. Thus, the $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructure has a spin Hall angle θ_{SH} obtained from the angular dependences of the spin magnetoresistance [5], turned out to be significantly higher than that of structures with a Pt film [6–8]. Considering the prospects of Ir-containing magnetic heterostructures for practical applications of spin-dependent processes, there is a request for a more detailed characterization of the heterostructure of $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ and its forming films in a wide range of temperatures and magnetic fields.

The literature data [9] based on the Hall effect in $SrIrO_3$ indicate a predominantly electronic type of conductivity, despite the mixed carrier type. In $La_{2/3}Sr_{1/3}MnO_3$ films, the Hall effect and magnetoresistance largely depend on the mismatch of the crystal structure of the film and the substrate [10]. There are also known studies of the temperature dependences of $SrIrO_3$ films on various substrates [11,12], manganite $La_{2/3}Sr_{1/3}MnO_3$ on $SrTiO_3$ substrate [13]. This paper provides the results of measurements of the temperature dependences of electrical resistance, magnetoresistance, and Hall response from $La_{2/3}Sr_{1/3}MnO_3$, $SrIrO_3$ films and the $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructure on $NdGaO_3$ substrate when cooled to temperature of $T = 2$ K and magnetic fields up to $H = 7$ T.

Epitaxial thin films of $SrIrO_3$ and $La_{2/3}Sr_{1/3}MnO_3$ with a thickness of $d = 10-50$ nm were grown on single-crystal substrates $(110)NdGaO_3$ by radio-frequency magnetron sputtering at a substrate temperature of $770-800\,^{\circ}$C in a mixture of argon and oxygen gases at a total pressure of $0.3-0.5$ mbar [14,15]. $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructures were produced by sequential deposition of $La_{2/3}Sr_{1/3}MnO_3$ and $SrIrO_3$ films in the chamber without vacuum break. The film deposition process was followed by cooling: first rapidly in a sputtering atmosphere of up to $500\,^{\circ}$C, and then in an oxygen atmosphere at a rate of $10\,^{\circ}$C/min.

The structural properties of thin-film structures were studied by X-ray diffraction using a Rigaku Smart Lab diffractometer with a rotating copper anode. It followed from the X-ray images [15] that the interplanar spacing in terms of responses (001), (002), (003) and (004) along c-direction from $La_{2/3}Sr_{1/3}MnO_3$ sputtered onto an $NdGaO_3$ substrate was $\Delta c_{LSMO} = 0.3905 \pm 0.0005$ nm, and for the $SrIrO_3$ film deposited *in situ* over $La_{2/3}Sr_{1/3}MnO_3$, $\Delta c_{SIO} = 0.404 \pm 0.001$ nm. From a cross-section image of a heterostructure on a Thermo Fisher Titan Themis Z trans-

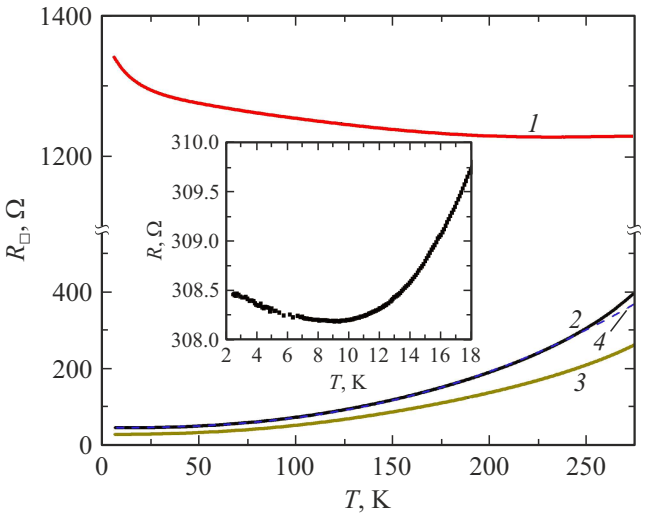

Figure 1. Temperature dependences of resistance per square ($R_\square$) of individual films and heterostructure: $SrIrO_3$ (*1*), $La_{2/3}Sr_{1/3}MnO_3$ (*2*), $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ (*3*). The dotted line (*4*) shows an approximation by a power function $\propto T^{5/2}$ of the temperature dependence of the resistance of $L = 0.65$ mm long $La_{2/3}Sr_{1/3}MnO_3$ film. Insert — low-temperature section $R(T)$ $La_{2/3}Sr_{1/3}MnO_3$.

mission electron microscope, visualizing the interface between $SrIrO_3$, $La_{2/3}Sr_{1/3}MnO_3$ films and the $NdGaO_3$ substrate, it followed that the films are grown with an epitaxial ratio $(001)SrIrO_3//(001)La_{2/3}Sr_{1/3}MnO_3//(110)NdGaO_3$, $[001]SrIrO_3//[001]La_{2/3}Sr_{1/3}MnO_3//[001]NdGaO_3$.

The electrophysical characteristics of the films and the heterostructure were studied using a 4-point measurement scheme with the „Hall“ cross geometry with a width of $W = 100\,\mu$m and a length of $L = 0.65$ mm. Metallized contact pads on the heterostructure were placed on top of the $SrIrO_3$ film. The Hall effect response and magnetic resistance voltages were measured at a temperature of $T = 2–300$ K in a cryostat with a superconducting solenoid in a magnetic field from H to ± 7 according to the method [16], which minimizes the effect of thermoelectrical effects by switching the polarity of the set measuring current I with a switching frequency set by the pulse duration (usually on the order of 1.5 s). The response voltage was recorded with a Keithley 2600B nanovoltmeter, and the temperature on the sample was recorded using a Lake Shore Cernox sensor.

Figure 1 shows the temperature dependences of the resistance of $R_\square(T) = R(T) \cdot W/L$ at $H = 0$ of $SrIrO_3$ films with $d_{SIO} = 35$ nm, $La_{2/3}Sr_{1/3}MnO_3$ with $d_{LSMO} = 40$ nm and heterostructure with $d_{SIO/LSMO} = 35$ nm ($d_{SIO} = 10$ nm, $d_{LSMO} = 25$ nm). It is possible to see the difference between $R_\square(T)$ for $SrIrO_3$ and $La_{2/3}Sr_{1/3}MnO_3$ films, whose shunting role is manifested on $R_\square(T)$ of heterostructure. In the region of relatively high temperatures of $T = 50–250$ K the dependence $R_\square(T)$ of the $La_{2/3}Sr_{1/3}MnO_3$ film is well described by the power law $R(T) \propto T^{5/2}$ as in ferromagnetic half-metals [17]. An inflection of $R(T)$ occurs in the low temperature range of $T < 20$ K on $T_{\min} = 9.5$ K at $R_\square(T)$ $La_{2/3}Sr_{1/3}MnO_3$ (inset in Figure 1), interpreted by weak localization in the three-dimensional case [18] with a root dependence of the specific conductivity $\Delta\sigma = \sigma - \sigma_0 \sim a \cdot (t)^{1/2}$, where $\sigma_0 = \sigma(T = 0)$, $t = T/T_{\min}$, a is the proportionality coefficient. The experimental dependences $R_\square(T)$ are conveniently approximated by the functions of the normalized resistivity $\Delta\rho = (\rho - \rho_{\min})/\rho_{\min}$, where $\rho = R_\square \cdot d$, $\rho_{\min} = \rho(T_{\min})$.

Figure 2 shows the experimental dependences of the normalized resistivity for $La_{2/3}Sr_{1/3}MnO_3$ and the heterostructure at $T < 20$ K and approximations of the form $b\rho(t)^{5/2} + (a(t)^{1/2} + \sigma_0)^{-1}$. In the case of film $La_{2/3}Sr_{1/3}MnO_3$ $\rho_{\min} = 1.9 \cdot 10^{-4}\,\Omega \cdot$ cm, $T_{\min} = 9.6$ K; in the heterostructure $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ $\rho_{\min} = 1.0 \cdot 10^{-4}\,\Omega \cdot$ cm, $T_{\min} = 8.7$ K. At the same time, $R_{\min} = 308.2\,\Omega$ for $La_{2/3}Sr_{1/3}MnO_3$ and the heterostructure $R_{\min} = 196.4\,\Omega$ with the same (within the error ± 0.5 nm)

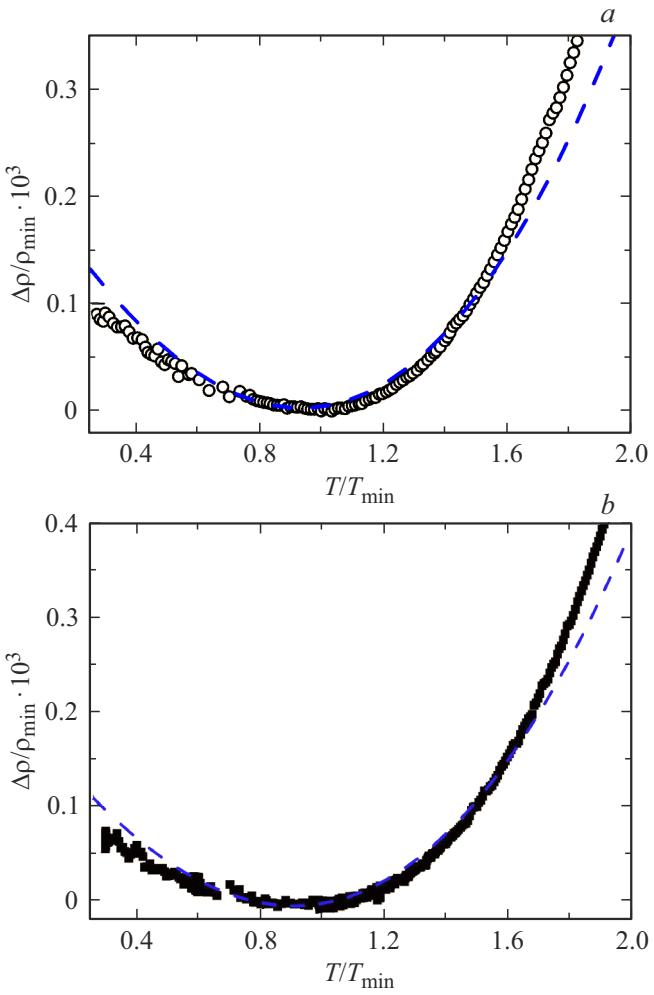

Figure 2. Normalized temperature dependences of the difference of the resistivity $\Delta\rho(t)/\rho_{\min}$: $La_{2/3}Sr_{1/3}MnO_3$ film (*a*), $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructure (*b*). The dashed lines show the approximation dependencies.

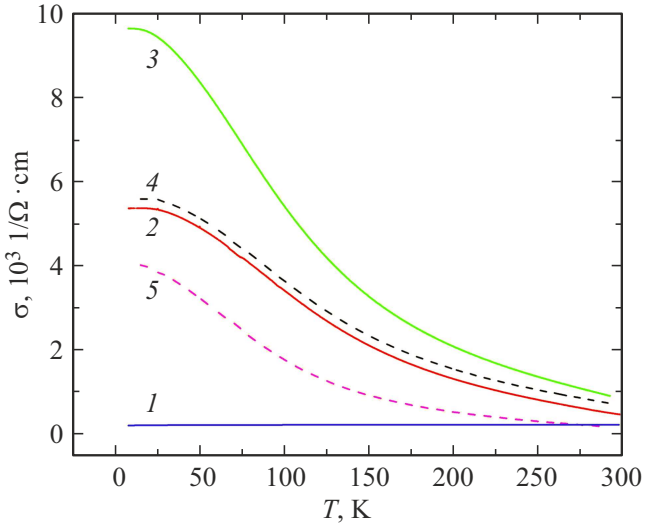

Figure 3. Experimental temperature dependences at $H = 0$ of the specific conductivities of films and heterostructures: *1* — $\sigma_1(T)$ $d_{SIO} = 35$ nm, *2* — $\sigma_2(T)$ $d_{LSMO} = 40$ nm, *3* — $\sigma_3(T)$ $d_{SIO/LSMO} = 35$ nm. Dashed lines: *4* — $\sigma_4(T) = \sigma_1(T) + \sigma_2(T)$, *5* — $\sigma_5(T) = \sigma_3(T) - \sigma_4(T)$.

thickness $d_{SIO} = d_{SIO/LSMO}$. It should be noted that the relative accuracy of the definition of $\Delta\rho/\rho_{\min}$ was at least 5 characters. Weak localization in thin films of manganite with a inflection $R(T)$ at low temperatures of $T < 20$ K was reported in Refs. [13,19].

Figure 3 shows the temperature dependences of the specific conductivities $\sigma_i(T)$ of individual films and the heterostructure: $i = 1$ ($SrIrO_3$), $i = 2$ ($La_{2/3}Sr_{1/3}MnO_3$), $i = 3$ ($SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$). As can be seen, the trivial summation of $\sigma_4(T) = \sigma_1(T) + \sigma_2(T)$ differs markedly from the $\sigma_3(T)$ of heterostructure. The figure also shows the difference curve *5*, corresponding to the contribution from the transition layer $\sigma_5(T) = \sigma_3(T) - \sigma_4(T)$. The formation of an intermediate layer at the $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ boundary was reported in Ref. [2], but its resistive parameters were not discussed.

A Hall-cross configuration was used to measure magnetoresistance and Hall response. The longitudinal current I_{xx} was set and the voltage dependences on the magnetic field were measured: transverse $V_{xy}(H)$ and longitudinal $V_{xx}(H)$. In the case of the heterostructure, the upper $SrIrO_3$ film was used to set the current I_{xx} and measure the voltages V_{xx} and V_{xy}. The magnetic field H was directed perpendicular to the film plane and ranged from 0 to $H_{\max} = 7$ T. Due to the change in the polarity of the current I_H supplied to the solenoid, the direction of the magnetic field changed.

It should be noted that the Hall responses in manganites are ambiguously dependent on temperature, in particular, in Refs. [10,20] it was reported that both a change in the sign of the Hall resistance and a response from the anomalous Hall effect were observed in the experiment. Figure 4 shows the dependences of the Hall voltage and magnetoresistance at $T \sim 10$ K, when the electrical conductivity $La_{2/3}Sr_{1/3}MnO_3$ can be considered metallic, and the Hall response $V_{xy}(H)$ demonstrates growth with a field H and negative magnetoresistance.

It can be seen from figure 4*a* that the voltages V_{xy} for all three samples linearly depend on the field H and the Hall resistance $R_H = V_{xy}d_i/\mu_0 H$ with a simplified estimate of the effective carrier concentration $n_{\text{eff}} = 1/eR_H$ (μ_0 and e — physical constants) for $T = 10$ K, we obtain for $La_{2/3}Sr_{1/3}MnO_3$ a hole $n_{\text{eff}} = 1.6 \cdot 10^{22}$ cm^{-3}, and for $SrIrO_3$ and $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ electronic n_{eff}: $1.4 \cdot 10^{21}$ cm^{-3} and $1.25 \cdot 10^{22}$ cm^{-3} respectively. It should be noted that the parameters of mobility (μ_e, μ_h) and concentration (n_e, n_h) of electrons and holes in thin $SrIrO_3$ films on the $SrTiO_3$ substrate were taken into account separately in Ref. [9]. The mobility ratio $\mu_e/\mu_h \sim 0.3$ was reported for $La_{2/3}Sr_{1/3}MnO_3$ films in Ref. [10], indicating a

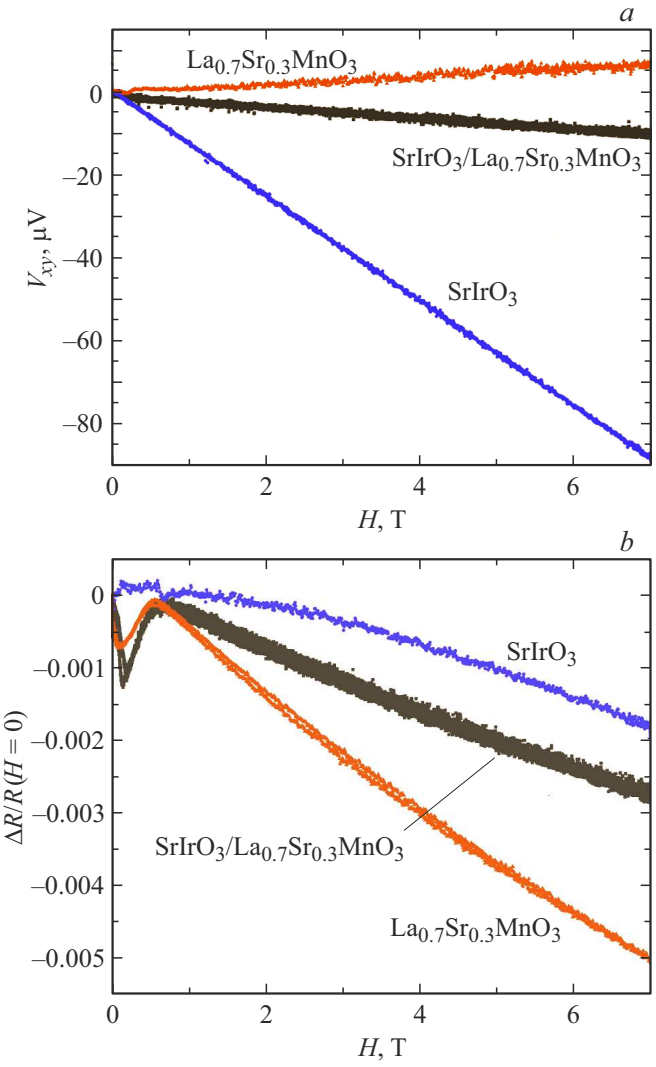

Figure 4. Dependences of Hall voltage (*a*) and magnetic resistance (*b*) $SrIrO_3$, $La_{2/3}Sr_{1/3}MnO_3$ and $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ at $T = 10$ K. The amplitude of the alternating measuring current $I_{xx} = 100\,\mu$A.

difference in the effective masses of electrons and holes in these materials.

Figure 4*b* shows the dependences of the resistance drop normalized by $R(0)$ $\Delta R = R(H) - R(0)$ on the magnetic field H. It can be seen that the $SrIrO_3$ film has a negative magnetic resistance over the entire range of fields H. Negative magnetoresistance in $La_{2/3}Sr_{1/3}MnO_3$ film and $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructure occurs at $H > 1$ T, however, in the field interval of $H = 0-1$ T, the sign of $\Delta R(H)/R(0)$ changes twice on $La_{2/3}Sr_{1/3}MnO_3$ and on $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$. The repeatability should be noted — figure 4*b* shows well-matching double tracks of magnetoresistance recording. A similar change in the sign of the magnetoresistance was reported in Ref. [21] for the ferromagnetic superlattice $SrRuO_3/La_{2/3}Sr_{1/3}MnO_3$. The authors of Ref. [21] attribute the appearance of a positive sign of magnetoresistance to weak antilocalization due to SOC, which manifests itself when varying the thicknesses of ferromagnets in the superlattice and magnetic anisotropy due to layers of $SrRuO_3$ in a superlattice grown on $SrTiO_3$. In our case, the positive sign of $\Delta R(H)/R(0)$ in the range of $H = 1.1-5.5$ kOe on $La_{2/3}Sr_{1/3}MnO_3$ film grown on $NdGaO_3$ substrate is difficult to bind with weak antilocalization, and covering $La_{2/3}Sr_{1/3}MnO_3$ with $SrIrO_3$ film with strong SOC, on the contrary, only reduced the difference $\Delta R(H)/R(0)$ approximately by 1.5 times. Almost the same ratio of 1.57 is obtained for values $R_{\min}$ for $La_{2/3}Sr_{1/3}MnO_3$ film and heterostructure.

Eventually, temperature dependences are obtained at $T = 2-300$ K of the electrical conductivity of thin films of $SrIrO_3$, $La_{2/3}Sr_{1/3}MnO_3$ and $SrIrO_3$ heterostructure on the $NdGaO_3$ substrate. From Hall measurements at $T = 10$ K and $H < 7$ T, it was found that $SrIrO_3$ exhibits an electronic type of conductivity and negative magnetoresistance. The film of ferromagnetic Half-metal $La_{2/3}Sr_{1/3}MnO_3$ has hole-like conductivity under the same experimental conditions, and the magnetic resistance at $H < 1$ T changes sign twice from negative to positive and vice versa, the same happens for the $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ heterostructure. A drop in resistance with a minimum of $T \sim 10$ K is found at $T < 15$ K on $La_{2/3}Sr_{1/3}MnO_3$ and $SrIrO_3/La_{2/3}Sr_{1/3}MnO_3$ which is described by weak localization in the three-dimensional approximation.

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Conflict of interest

The authors declare no conflict of interest.

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