1. INTRODUCTION

The development of spintronic devices based on magnetoeactive materials with the nanoscale interfaces is an important task, which requires the study of the physics of structural and phase transformations in thin films and at the interfaces. Spintronics is based on phenomena associated with the transfer of the spin moment. A “pure” spin current can be created by several

SPINTRONICS

SPIN INTERACTIONS AT THE INTERFACES FERROMAGNETIC OXIDE/FERROMAGNETIC INTERMETALLIC SUPERLATTICE

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Abstract. The magnetic properties of the heterostructures consisting of platinum Pt, epitaxially grown manganite optimally doped by strontium La_{0.7}Sr_{0.3}MnO_3 (LSMO), rare earth intermetallic superlattices consisting of exchange-coupled layers TbCo_2/FeCo (TCFC), and the epitaxial film of yttrium iron garnet Y_3Fe_5O_12 (YIG) were investigated. The TCFC material provides giant magnetostriction, large magnitude of the magnetomechanical coupling coefficient, and controlled induced magnetic anisotropy. In addition TCFC, as well as Pt, has strong spin-orbit interaction. Experimental studies have shown that the magnetic interaction of the heterostructure (TeCo_2/FeCo)n/LSMO has an antiferromagnetic character. An increase of linewidth of the ferromagnetic resonance in the Pt/LSMO structure was observed and explained by the spin current flow induced in Pt film by the LSMO film at ferromagnetic resonance. In the TCFC/YIG heterostructure, an electric voltage induced in the TCFC film was observed and explained by the inverse spin-Hall effect under conditions of ferromagnetic resonance.

Keywords: heterostructure, manganite, intermetallic superlattice, yttrium iron garnet, ferromagnetic resonance, spin current, inverse spin-Hall effect

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1. INTRODUCTION

The development of spintronic devices based on magnetoeactive materials with the nanoscale interfaces is an important task, which requires the study of the physics of structural and phase transformations in thin films and at the interfaces. Spintronics is based on phenomena associated with the transfer of the spin moment. A “pure” spin current can be created by several
mechanisms, among which the most effective are the spin-Hall effect [1] and the spin pumping in ferromagnetic resonance mode [2]. The magnetic heterostructure as a rule consists of magnetic and nonmagnetic layers. Materials with a strong spin-orbit interaction play an important role in the detection of the spin current. A pure spin current ($J_s$) can be detected using the inverse spin-Hall effect (ISHE) in a material with strong spin-orbit interaction by converting into conduction current $J_c = \theta_{SH}(\hbar/2e)(J_s \cdot \sigma)$, where $\theta_{SH}$ is the angle of spin-Hall effect, $\sigma$ - carrier polarization.

However, not only non-magnetic normal metals can be used as ISHE spin current detectors. In a number of works, it was shown that magnetic metals, such as permalloy Py (NiFe), as well as Fe, Co, Ni can be used as spin current detectors [3-5]. TbCo$_2$/FeCo structures containing Tb element with a strong spin-orbit interaction (high atomic weight $Z$) are distinguished by giant magnetostriction, a large value of the magnetomechanical coupling coefficient, controlled induced magnetic anisotropy and the ability to induce spin-orientation transitions by a magnetic field or elastic stresses [6, 7]. In turn, rare earth manganite perovskites with the structure $Re_{1-x}A_xMnO_3$ (Re are rare earth materials such as La or Nd), and A - alkaline earth metals such as Sr, Ca, Ba) exhibit a wide range of unusual electrical and magnetic properties, including high (up to 100 %) magnetic polarization, the effect of colossal magnetoresistance, etc (see review [8]). Manganite films for which the Curie temperature $T_C$ is close to room temperature is especially attractive for practical use. A number of studies on the excitation of the spin current by ferromagnetic resonance in LSMO/N structures (N is a normal metal, usually platinum) were made in [9, 10]. However, there are no data on the temperature dependences of the width of the ferromagnetic resonance (FMR) line during the generation of spin current in ferromagnets and on the magnitude of the spin conductivity of the LSMO/Pt boundary. We have investigated the magnetic properties of heterostructures consisting of platinum Pt, epitaxially grown manganite $La_{0.7}Sr_{0.3}MnO_3$ (LSMO), rare earth intermetallic superlattices consisting of exchange-coupled layers TbCo$_2$/FeCo (TCFC), and the epitaxial film of yttrium iron garnet $Y_3Fe_5O_{12}$ (YIG). Our studies are focused on the effects associated with the excitation and generation of spin current in the structures.

2. FERROMAGNETIC RESONANCE IN MANGANITE EPITAXIAL FILMS

The Hilbert damping parameter $\alpha$ is a measure of the spin precession relaxation in homogeneous ferromagnets caused by spin-orbit interaction [11]. The width of the homogeneous FMR line induced by Hilbert damping is proportional to the FMR frequency $\omega$, $\Delta H = \alpha / \gamma$ (where $\gamma = g\mu_B / \hbar$ is the gyromagnetic ratio) and describes the situation for the homogeneous case. In the ferromagnetic structure of a ferromagnet and a normal metal, the FMR line is additionally broadened due to the generation of the spin current, inhomogeneity of the magnetization of the ferromagnet, interaction with another material, two-magnon scattering and the appearance of eddy current in the ferromagnet. As a result, the FMR linewidth can be represented as a sum $\Delta H_{pp} = \Delta H_{I} + \Delta H_{2M} + \Delta H_{E}$ (1), where $\Delta H_{I}, \Delta H_{2M}, \Delta H_{E}$ are the widths of the lines for damping caused by the inhomogeneous state of a ferromagnet, two-magnon scattering and damping caused by eddy current, respectively [12, 13]. A change in the magnetic properties of materials such as its anisotropy or magnetization affects the inhomogeneous broadening of the FMR line ($\Delta H$) which is independent of frequency [12, 13]. The magnetic field of an alternating current caused by FMR induces eddy currents in a thin conductive film. These currents produce an additional change in the amplitude of the alternating current magnetic fields of the heterostructure. The influence of eddy currents on ferromagnetic resonance in a conducting ferromagnetic system can lead to
broadening of the FMR linewidth ($\Delta H_{pp}$) and to the change in the shape of the FMR spectrum at non-uniform microwave field [14-16]. The mechanism of two-magnon scattering leads to the connection of homogeneous precession ($k = 0$) with degenerate finite spin-wave modes [17, 18]. In a ferromagnet/normal metal heterostructure the precession of magnetization in a ferromagnet causes a spin current to flow through the boundary into the normal metal, which broadens the FMR line. The theory [19] predicts the spin current to flow through the boundary of the ferromagnetic and nonmagnetic layers perpendicular to the interface.

The linewidth of ferromagnetic resonance $\Delta H_{pp}$, measured by scanning the external magnetic field $H$, is defined as the difference in the field positions between the extremes $H_{p+}, H_{p-}$ of the first derivative of the $dP/dH$ microwave absorption signal (see Fig. 1). At this value, the resonance field $H_{0}$ defined as the point of transition of the signal $dP/dH$ through zero is always in the range $H_{p+} < H_{0} < H_{p-}$. Note, that the determination of the linewidth by approximating the FMR spectrum by several Lorentz lines gives an approximate 10% correction in the value of $\Delta H_{pp}$.

Experimental studies were performed on La$_{0.7}$Sr$_{0.3}$MnO$_3$ (LSMO) epitaxial films, which were deposited by magnetron sputtering onto (110) NdGaO$_3$ (NGO) single crystal substrates at $T = 820^\circ$C and oxygen pressure of 0.15-1 mbar. Pt films 10-20 nm thick were deposited ex situ immediately after cooling of the manganite film. Magnetic characteristics were measured by FMR using a Bruker spectrometer (frequency 9.51 GHz). The experimental samples were located in the microwave cavity of the spectrometer so that the sample plane was always parallel to the direction of the constant external magnetic field and the magnetic component of the microwave field (parallel orientation). The method of sample preparation and FMR measurements are described in [20]. Obtained films were investigated immediately after the deposition and cooling to room temperature (d-LSMO), and were annealed after the growth at $T = 820^\circ$C for one hour (h-LSMO). For h-LSMO films, 40 nm thick, we determined the attenuation $\alpha_0$ of the spin precession at room temperature from linewidth $\Delta H_{pp} = 28$ Oe which corresponds to $\alpha_0 = \Delta H_{pp} \gamma/\omega = 8 \cdot 10^{-3}$. The increase in attenuation $\alpha = \alpha_0 + \alpha'$ during the deposition of Pt on the LSMO can occur due to the flow of the spin current through the Pt/LSMO boundary. For Pt deposited on the h-LSMO film with thickness of 10 nm the parameter $\alpha$ increases by 10%. Using [21, 22], one can calculate the spin conductivity in the Pt/LSMO heterostructure:

$$g_{\uparrow\downarrow} = \frac{4\pi M_{s} t_{\text{LSMO}} \alpha'}{g_{\mu B}}$$

where $\gamma = 17.605 \cdot 10^{6}$ is gyromagnetic ratio for an electron, $\omega = 2 \pi \cdot 9.51 \cdot 10^{9}$ s$^{-1}$ is microwave angular frequency, $M_{s} = 300$ Oe is LSMO film magnetization, $t_{\text{LSMO}} = 40$ nm is LSMO film thickness, $\mu_{B} = 9.274 \cdot 10^{-21}$ erg/G is the Bohr magneton, $g = 2$ is the Lande factor. At room temperature, an increase in the FMR linewidth after deposition of Pt $\Delta H_{pp/\text{LSMO}} - \Delta H_{\text{LSMO}} = 4$ Oe and, therefore, $g_{\uparrow\downarrow} = 4.1 \cdot 10^{10}$ m$^2$ was obtained. This value of the spin conduction of the boundary slightly exceeds $g_{\uparrow\downarrow} \sim 10^{18}$ m$^2$, obtained from measurements of the spin current on the same Pt/LSMO structures using the inverse spin-Hall effect [23]. For comparison, $g_{\uparrow\downarrow} = 2.1 \cdot 10^{19}$ m$^2$
[24] was obtained for Py/Pt boundaries, and $g_{↑↓} = 4.8 \cdot 10^{20} \text{ m}^{-2}$ [25] for YIG/Pt.

When estimating the spin conductivity, using (2), other mechanisms of spin precession damping were not been taken into account. The effective one-dimensional spin conductivity of a normal metal layer connected in series with the spin conductivity of the gext interface contributes to the effective spin conductivity of the structure [26, 27].

$$g_{eff} = \frac{1}{g_{↑↓} + 1/g_{ext}}.$$ (3)

The expression for $g_{ext}$ is obtained by solving the spin diffusion equation with the corresponding boundary conditions. In the case of the ferromagnet/normal metal structure, the following spin conductivity expression was obtained [22]

$$g_{ext} = \frac{\tanh(d_{Pt}/\lambda_d)}{2\lambda_d},$$ (4)

where $\rho_{Pt}$, $d_{Pt}$ and $\lambda_d$ are the specific resistance, thickness and diffusion length for the Pt film, respectively. For a thickness of $d_{Pt} = 10 \text{ nm}$, exceeding $\lambda_d = 3 \text{ nm}$ [22], $\tanh(d_{Pt}/\lambda_d) \approx 1$, and the contribution to the line width from the spin conductivity in the Pt film is equal at room temperature:

$$\Delta H_{ext} = \frac{\omega}{\gamma} g \mu_B h / (2\pi e^2 M d_{Pt} \rho_{Pt} \lambda_d) \approx 6 \text{ Oe},$$ (5)

where $g = 2$, $M = 300 \text{ Oe}$, $d_{Pt} = 4 \cdot 10^{-6} \text{ cm}$, $\rho_{Pt} = 3 \cdot 10^{-5} \Omega \text{ cm}$, $\lambda_d = 3 \cdot 10^{-7} \text{ cm}$, $h/e^2 = 2.6 \cdot 10^4 \Omega$. The broadening of the FMR line due to the effective spin conductivity of the normal metal layer is equal to the contribution from the spin current. The large value of this broadening is probably caused by an error in determining the spin relaxation length $\lambda_d$. The temperature dependences of the linewidth for the b-LSMO manganite film and for the Pt/b-LSMO heterostructure are presented in Fig. 2. With a decrease in the temperature the $\Delta H_{pp}$ markedly increased. Increasing $M$ with decreasing $T$ may cause an increase in the linewidth (see expression (5)). But below $T = 200 \text{ K}$, the magnetization $M$ is saturated, and the width of the FMR line continues to grow.

![Fig. 2. Temperature dependences of the width of the FMR lines of the b-LSMO film (open squares) and Pt/b-LSMO heterostructures (filled squares). The temperature dependences of the resistance of LSMO manganite films (green line) and Pt/b-LSMO heterostructures (red line) are presented.](image)

When a Pt film is deposited on top of an LSMO film, the overall conductivity of the structure increases (see Fig. 2). The increase in $\Delta H_{pp}$, which is observed in the experiment after the deposition of Pt at low temperatures, can be fully explained by the generation of the spin current in the Pt/LSMO heterostructure. With decreasing temperature, the resistivity of the Pt film decreases in proportion to $T$, and the resistance of the LSMO film changes by more than one order of magnitude. The contribution of all layers to the resistivity of the heterostructure is explained by the fact that the LSMO film together with the Pt film acts as parallel resistors [28]. An increase in $\Delta H_{pp}$ with decreasing temperature can be caused by damping caused by eddy current, which is proportional to the conductivity of the structure.

3. FMR FOR TCFC/LSMO HETEROSTRUCTURE

The FMR spectrum of the TCFC/LSMO heterostructure presented in Fig. 3a, was measured at the frequency $\omega/2\pi = 9.74 \text{ GHz}$ at $T = 300 \text{ K}$ with a constant field lying in the plane of the heterostructure and directed along the axis of easy magnetization of the heterostructure. Three areas of ferromagnetic order are visible in the structure. The temperature dependences of the resonance fields $H_0$ for three lines of the FMR spectrum are shown in Fig. 3b. The line in the
Since both parts of LSMO-1 and LSMO-2 are located on the same substrate and have the same crystal structure, the observed difference of $\Delta H$ in peaks is most likely caused by the interaction of the TCFC superlattice and LSMO film. A similar broadening of the FMR line for structures with ferromagnet/normal metal interfaces was observed previously (see the part 2 of the article) and is explained theoretically [19] by the spin current leaving the ferromagnet in a normal metal during FMR.

Solving the Landau-Lifshitz-Gilbert equation gives two resonance relations $\omega(H_0)$, describing the FMR in TCFC and LSMO-2 films. With allowance for uniaxial and biaxial anisotropies, these relations are similar to those obtained in [29] for an autonomous LSMO film deposited on an (011) NdGaO$_3$ orthorhombic substrate, which causes the uniaxial magnetic anisotropy [20]. Taking into account the magnetic interaction in the expressions for the resonant frequency $\omega(H_0)$[20], the value of $H_0$ should be replaced by the sum of two terms $H_{01} + H_{J1}$ and $H_{02} + H_{J2}$ for the LSMO-2 film and TCFC superlattices, respectively. Here, $H_{J1} = J/(M_1d_1)$ and $H_{J2} = J/(M_2d_2)$ ($d_1$ and $d_2$ of the thickness of the LSMO and TCFC layers, respectively) give an effective interlayer exchange interaction for the LSMO-2 and TCFC films.

To determine the structure parameters, we first calculate the angular dependence of the FMR response of an autonomous LSMO film (LSMO-1). Then, using the magnitude of the magnetization obtained by fitting the dependence of the resonant field on the angle, we calculate the angular dependence of $H_0$ for the LSMO-2 film. Then, the exchange interaction constant $J$ is determined. Finally, the obtained value $J$ is used to calculate the angular dependence of $H_0$ for the TCFC film and for calculating the value $M_s$. As a result, the data obtained allow us to conclude that the TCFC/LSMO interface can be characterized by antiferromagnetic interlayer interaction with a constant $J = -0.24$ erg/cm.
4. RARE-EARTH INTERMETALLIC SUPERLATTICES

The structure in the form of a ‘sandwich’ (FeCo/TbCo$_2$/FeCo)/MgO/(FeCo/TbCo$_2$/FeCo) (TCFCM), which is promising for creating the magnetically controlled switches, was investigated [30]. The two films (in brackets) are three-layer polycrystalline films of intermetallic compounds with a thickness of 2 nm (FeCo) and 6 nm (TbCo$_2$), which are separated by a thin layer of MgO with a thickness of 3 nm. This structure was deposited on a silicon substrate with dimensions of $5 \times 5 \times 0.5$ mm$^3$. Both intermetallic films are ferromagnets at room temperature with giant magnetostriction. During deposition of intermetallic films the external magnetic field was applied in parallel to the substrate plane, keeping the same direction, but with significantly different intensities for each of compounds. In this case, it was expected that the easy axis $\mathbf{n}_u$ magnetic anisotropy of the films will lie in their plane and will be oriented along the imposed magnetic field, while their magnetic anisotropy constants will be significantly different.

Recently, ferromagnetic films with in-plane uniaxial anisotropy of the magnetic field revealed the effect of a sharp increase in the radio frequency (RF) dynamic magnetic susceptibility [23, 31–33]. It is observed when the external magnetic field $H_0$ lies in the film plane and directed perpendicular to its easy magnetization axis $\mathbf{n}_u$ (and, therefore, parallel relative to the hard axis direction) and passes through the value $H_0 = H_u$, where $H_u$ is the field of uniaxial intra-planar magnetic anisotropy of the film. In this case, the recording radio frequency magnetic field $H_0 e^{i \omega t}$ is directed perpendicular to the field $H_0$, and its frequency $\omega$ is fixed. The effect manifests itself in the form of a relatively narrow resonance-like absorption signal with a maximum at $H_0 = H_u$. A feature of this signal is that when another frequency $\omega$ is set being changed over wide limits, the field at which it is observed remains unchanged and is equal to $H_0 = H_u$. This effect, called the “magnetic pseudo-resonance” [23], is caused by the loss of stability of the magnetic system at the critical point $H_0 = H_u$ of the transition from the angular phase to the collinear one with a direction of magnetization parallel to the external field [33].

To search for a pseudo-resonance signal, the TCFC film structure was placed in a Q-meter sensor coil. The signal was recorded by the method of synchronous detection with a bi-directional sweep of the $H_0$ field, additionally modulated with the frequency of 52.3 kHz and the amplitude of 1.3 Oe. Signal registration was carried out continuously with multiple accumulations (up to 100 times). The received signal is shown in Fig. 4. Two components are clearly seen, corresponding to the two pseudo-resonance signals. We note that the shape of these signals differs markedly from the derivative of absorption in a magnetic field that is usual for EPR spectroscopy. The distortions are due to the high sensitivity of the pseudo-resonance signals to the orientation of the modulating field, which in our experimental conditions has a small transverse component. Without dwelling on the details, we confine ourselves to stating the fact that the two observed components of the RF absorption spectrum correspond to two pseudo-resonance

![Fig. 4. Radio-frequency absorption in a ferromagnetic heterostructure film (FeCo/TbCo$_2$/FeCo)/MgO/(FeCo/TbCo$_2$/FeCo) consisting of two three-layer films (in brackets), separated by a layer of MgO, depending on the external magnetic field $H_0$, oriented perpendicular to the $\mathbf{n}_u$ axis of the easy magnetization.](image)
signals, 1 and 2, with maxima at $H_0^*$ around 74 Oe and 456 Oe, respectively. Different values of $H_0^*$ for these signals mean that the magnetic anisotropy fields $H_u$ in sandwich-type films are significantly different. For signal 1, the dependence $S^{(1)}(\varphi)$ of its maximum on angle $\varphi$ was removed (inset (a) in Fig. 4). It was obtained by scanning the field $H_0$ in a relatively narrow range of 0–140 Oe at angles $\varphi$, including the values $\varphi = 90^\circ$ and $270^\circ$. It can be seen that in both cases the dependencies are almost the same. They turn out to be much smoother than a similar dependence for the LSMO film: their width at half-height reaches 40°. As a result, the average direction of the axis of difficult magnetization for film 1 corresponds to the angle $\varphi = 90^\circ$ with an accuracy of ±4°. Similar measurements for film 2 with a wider pseudo-resonance line yielded the value $\varphi = 90^\circ \pm 12^\circ$. From this it follows that in both films of the TCFCM structure the directions of the axes of the intra-plane magnetic anisotropy almost coincide.

It is natural to compare the data obtained for the described heterostructure with what gives FMR at a frequency of 9.78 GHz. Inset (b) is Fig. 4 shows the FMR signal from this structure, which is a derivative of the FMR absorption signal. It was obtained by the method of synchronous detection when sweeping the $H_0$ field under conditions of its modulation with a frequency of 100 kHz and amplitude of 10 Oe and using 4-fold accumulation. It can be seen that the signal is very broad, there is no resolved structure in it, corresponding to two films with significantly different values of the anisotropy field $H_u$. Experimentally, it was not possible to identify the maximum of the pseudo-resonance in a magnetic field. In complex structures such as TCFCM, interlayer exchange interactions usually occur, which can lead to renormalization of the anisotropy fields and the corresponding shift of the maximum of the pseudo-resonance in a magnetic field.

5. SPIN CURRENT IN TCFC/Y$_3$Fe$_5$O$_{12}$ STRUCTURE
Films from Y$_3$Fe$_5$O$_{12}$ (YIG) are very attractive for spintronic structures because of the small magnetic attenuation and the fact that they are an insulator. It was reported that the spin current can be excited in YIG and detected with a Py film [3-5]. Fig. 5 presents the results of experimental studies of the inverse spin-Hall effect (ISHF) in the TCFC/YIG heterostructure. The YIG epitaxial film with a thickness of 5 μm was grown on a (111) Gd$_3$Ga$_5$O$_{12}$ (GGG) substrate. The [TbCo$_2$(5nm)/Cu(0.4nm)]
FeCo(5nm)/Cu(0.4nm)]_{32} \text{nm thick superlattice was deposited. On the surface of the heterostructure, contact pads were formed to measure the potential difference caused by the ISHE effect. The sample was placed on a strip microwave line located in the gap of the electromagnet, with which FMR was excited in the YIG film. When the magnetizing field was changed, the potential difference ISHE and the intensity of the FMR signal were recorded. Fig. 5 shows the FMR spectrum of a YIG film, taken in a microstrip configuration at a frequency of 1615 MHz at $T = 300$ K, a generator power of 2 mW and a magnetic field that lies in the plane of the substrate and directed along the hard axis of magnetization. At $H = 169 \pm 0.2$ Oe, a peak is observed, caused by FMR in the YIG film. There is a slight asymmetry of the peak position relative to a change in the polarity of the magnetic field. The peak half-width is 24 Oe. The second peak caused by ferromagnetism of TCFC is not observed due to the small thickness of the layer.

Fig. 5 also shows the dependence of the voltage of the inverse spin-Hall effect measured on the TCFC$_c$ film on the magnetic field. A strong asymmetry of the ISHE peak of $\sim 7$ Oe is observed. The ISHE peak width was 60 Oe. When the direction of the magnetizing field was changed, a change in the ISHE sign of voltage was observed, the value of which reached 80 $\mu$V.

The implementation of ISHE in ferromagnetic metals significantly expands the types of materials that could be used to study the spin current, including the rare-earth materials of the lanthanum group with extremely large spin-Hall angles, such as manganites, ruthenates, and etc.

6. CONCLUSION
Experimental studies of the magnetic properties of heterostructures consisting of epitaxially grown manganite LSMO and rare-earth intermetallic superlattices consisting of exchange-coupled TCFC layers showed that the magnetic interaction in the heterostructure has an antiferromagnetic character. After the deposition of a thin Pt film on top of the LSMO manganite film, an increase in the ferromagnetic resonance line width was observed due to the spin current flowing in Pt, which occurs in the LSMO film at resonance. An electric voltage caused by the inverse spin Hall effect in a TCFC film was experimentally observed under conditions of ferromagnetic resonance in an yttrium iron garnet.

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