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RESEARCH ARTICLE

Specific features of temperature dependence of colossal and tunneling magnetoresistance in manganite films

Tatiana N. Bakhvalova¹, Alexey N. Yurasov¹, Maxim M. Yashin^{1, @},
Valentina A. Bessonova²

¹ MIREA – Russian Technological University, Moscow, 119454 Russia

² Mikheev Institute of Metal Physics of the Ural Branch of the Russian Academy of Sciences, Yekaterinburg, 620108 Russia

@ Corresponding author, e-mail: ihkamax@mail.ru

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Abstract

Objectives. This work aims to theoretically and experimentally investigate the specific features of magnetoresistance temperature dependence in nanostructured films of doped manganites. The temperature dependence of electrical resistance for $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ manganite films, grown by laser ablation on various dielectric substrates, is investigated over a wide temperature range.

Methods. Epitaxial $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films with a thickness of 80 nm were grown by pulsed laser ablation using an ArF excimer laser (a laser wavelength of 247 nm) on single-crystalline SrTiO_3 and $\text{ZrO}_2(\text{Y}_2\text{O}_3)$ substrates. The magnetoresistance properties were measured using a two-probe DC method. The measurements were conducted in magnetic fields up to 8 kOe applied in the film plane, across a temperature range of 80–350 K. To accomplish the research objectives, an empirical magnetoresistance model was applied in two distinct temperature regions: near the magnetic phase transition temperature and in the ground-state region.

Results. Empirical relations for temperature dependence of magnetoresistance for nanostructured $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films were established, encompassing both the Curie temperature region and the ground-state regime. Our studies revealed that the magnetoresistance of epitaxial single-crystalline $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films exhibits a sharp peak exclusively near the Curie temperature while remaining negligible in other temperature ranges. Conversely, $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films with a variant structure demonstrate significant low-temperature magnetoresistance. This effect arises from magnetic-field-induced modifications of the high-frequency conductivity, which results from spin-polarized electron tunneling across structural domain boundaries. A unified empirical model to describe various mechanisms of magnetoresistance in doped manganites is proposed.

Conclusions. For the first time, an empirical model to describe both the colossal and tunneling magnetoresistance in thin films of doped manganites has been developed. This model demonstrates excellent agreement between experimental and calculated data for $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ films with and without a variant structure. The simulation results agree well with experimental data. The findings elucidate the understanding of magnetoresistance mechanisms, contribute to the development of the magnetorefractive effect theory for thin-film manganites, and inform new approaches for controlling charge carrier dynamics in strongly correlated magnetic oxides.

Keywords: manganites films, variant structure, colossal magnetoresistance, spin-dependent tunneling, magnetorefractive effect, structural domains

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НАУЧНАЯ СТАТЬЯ

Особенности температурных зависимостей колоссального и туннельного магнитосопротивления в пленках манганитов

Т.Н. Бахвалова¹, А.Н. Юрасов¹, М.М. Яшин^{1, @}, В.А. Бессонова²

¹ МИРЭА – Российский технологический университет, Москва, 119454 Россия

² Институт физики металлов им. М.Н. Михеева УрО РАН, Екатеринбург, 620108 Россия

@ Автор для переписки, e-mail: ihkamax@mail.ru

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Резюме

Цели. Целью работы является экспериментальное и теоретическое исследование особенностей температурных зависимостей магнитосопротивления в наноструктурированных пленках легированных манганитов. В широком температурном диапазоне изучено поведение электросопротивления пленок манганитов состава $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$, выращенных методом лазерной абляции на различных диэлектрических подложках.

Методы. Для достижения поставленной цели методом лазерной абляции с использованием импульсного эксимерного были выращены эпитаксиальные пленки $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ толщиной 80 нм на монокристаллических подложках SrTiO_3 и $\text{ZrO}_2(\text{Y}_2\text{O}_3)$. Магнитосопротивление измерялось двухконтактным методом на постоянном токе в полях до 8 кЭ в плоскости образца и температурном интервале 80–350 К. Для достижения поставленной цели применялась эмпирическая модель магнитосопротивления в двух температурных областях: вблизи температуры магнитного фазового перехода и в области основного состояния.

Результаты. Построены эмпирические температурные зависимости магнитосопротивления для наноструктурированной пленки $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$, охватывающие как область температуры Кюри, так и область основного состояния. Показано, что в эпитаксиальной монокристаллической пленке $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ магнитосопротивление имеет выраженный максимум вблизи температуры Кюри и пренебрежимо мало в других областях. В пленке $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ с вариантной структурой имеется сильный низкотемпературный вклад в магнитосопротивление, связанный с изменением высокочастотной проводимости пленки во внешнем магнитном поле из-за процессов туннелирования спин-поляризованных электронов через границы структурных доменов. Предложена единая эмпирическая модель для описания различных механизмов магнитосопротивления в легированных манганитах.

Выводы. Впервые в рамках одной эмпирической модели проведено описание колоссального и туннельного магнитосопротивления для пленок легированных манганитов. Показано, что такая модель дает хорошее согласие экспериментальных и расчетных данных в пленке $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ с вариантной структурой. Результаты моделирования хорошо согласуются с экспериментальными данными. Полученные данные могут способствовать пониманию механизмов магнитосопротивления и развитию теории магниторефрактивного эффекта для тонкопленочных манганитов, а также разработке новых подходов к управлению динамикой носителей заряда в сильно-коррелированных магнитных оксидах.

Ключевые слова: пленки манганитов, вариантная структура, колоссальное магнитосопротивление, туннелирование, магниторефрактивный эффект, структурные домены

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INTRODUCTION

In modern physics, special attention is paid to the study of fundamental magnetic and transport properties of composite or nanostructured functional thin-film nanomaterials. Such materials form the basis of many current technological applications, from information carriers to sensor devices [1]. Doped lanthanum manganites are a promising class of substances exhibiting unusual properties [2–5] and possessing a number of unique characteristics. These include high spin polarization of charge carriers and magnetization, high sensitivity of static and optical conductivity to structural and magnetic phase transitions and external fields [2–5], e.g., metal-insulator transition, colossal magnetoresistance (MR) effect, giant magnetotransmittance, magnetorefractive effect (MRE), etc. [6–8]. These properties render doped lanthanum manganites potential candidates for use in spintronics and magneto-optical devices in the infrared (IR) range operating at room temperatures.

The magnetorefractive effect consists in a change in optical parameters (refractive indices, or more precisely, reflection and transmission coefficients) in a magnetic field. In heavily doped manganites, this is a high-frequency analogue of colossal MR in the IR range of the spectrum [2]. In manganite films, the MRE reaches gigantic values (10–20%); at the same time, its spectral and temperature dependence is quite intricate due to the contribution of not only delocalized charge carriers but also the magnetic and charge uniformity of the films, their thickness and MS value, as well as size effects, resonance phenomena, and defects ([2] and the references therein).

The physical properties of manganite films are determined by their composition and growth conditions, as well as by the type of substrate. Thus, the study [9] investigated $\text{La}_{2/3}\text{Ba}_{1/3}\text{MnO}_3$ films on $\text{ZrO}_2(\text{Y}_2\text{O}_3)$ substrates and found the formation of a variant (equivalent) structure during epitaxial growth of the film on a single-crystal substrate with significantly different crystal lattice parameters. This structure is formed by structural domains (variants) with nanoscale crystallites in the film bulk,

separated by coherent high-angle boundaries with a thickness of ~ 0.4 nm. It should be noted that the fixed orientation of crystallites with a limited set of angles is a fundamental difference between films with a variant structure and polycrystalline samples with disoriented crystallites of different sizes in the film bulk. The nature and features of the variant structure were discussed in detail in [10].

Consequently, the domain structure of such films determines the appearance of an additional mechanism underlying the MR caused by the tunneling processes of spin-polarized charge carriers at the domain boundaries, which are most intense when the temperature approaches absolute zero. A comparative analysis of the optical, electrical, and magneto-optical properties of $\text{La}_{2/3}\text{Ba}_{1/3}\text{MnO}_3$ films with and without a variant structure allowed us to study the main features of the physical mechanisms responsible for the temperature and field dependence of MR and MRE in these films [2, 7–9]. To describe MRE as a response to colossal MR in manganites, the effective medium approximation [11], previously proposed for the analysis of tunneling processes at optical frequencies in various metallic nanocomposites and granular alloys [12], was successfully applied. However, the high susceptibility of the electrical properties of manganites to internal and external factors leads to difficulties in the theoretical description of the MR and MRE mechanisms. Indeed, the developed theory is applicable to purely metallic and tunnel conduction and is capable of considering the redistribution of the high- and low-conductivity phases; however, it fails to take into account the effects associated, e.g., with changes in the magnetic field concentration of charge carriers, absorption edges, effective mass of polarons [2], etc. Furthermore, there is currently no single model describing the coexistence of two MR mechanisms in films of doped manganites with significantly different temperature regions of existence.

This article presents the results of simulating the temperature dependence of MR in a $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ (LBM) manganite film with a variant structure, in comparison with a film without a variant structure. It is shown that

the proposed empirical approach enables the main features to be taken into account, thus being useful when developing a general theory of MR in strongly correlated magnetoresistive magnetics.

EXPERIMENTAL

LBM epitaxial films with a thickness of $d = 80$ nm were grown by laser ablation using an excimer pulsed argon fluoride laser (ArF laser), with a laser wavelength of 247 nm and a frequency of about 9 Hz, on single-crystal SrTiO₃ (hereinafter referred to as STO) and ZrO₂(Y₂O₃) (hereinafter referred to as YSZ (Y₂O₃ stabilized ZrO₂)) substrates, at a substrate temperature of 730 K and an oxygen pressure of 0.4 mbar. The thickness of the films was determined by exposure duration. According to scanning electron microscopy (SEM) data, the surface of the LBM/YSZ film has a multi-grain structure with an average crystallite size of 40–70 nm. This structure is formed by columnar oriented crystallites growing through the thickness of the film, which is fundamentally different from the island-like growth of thin LBM/STO epitaxial films. The synthesis method and the results of film sample testing are detailed in [9, 13].

The magnetoresistance of the films $\frac{\Delta\rho}{\rho} = [\rho_H - \rho_0] / \rho_0$, where ρ_H and ρ_0 are the values of the specific resistance in the presence and absence of a magnetic field, was measured using a two-contact method at direct current in fields up to 8 kE in the plane of the sample and in the temperature range of 80–50 K. The experimental temperature and field dependencies of the MR of the films are shown in Fig. 1. The magnetoresistance in epitaxial films of LBM manganites (without a variant structure) demonstrates a field and temperature dependence similar to that of bulk single crystals of the same composition. In the LBM/STO film, a pronounced maximum in the temperature dependence of the negative MR effect in the region of Curie temperature $T_C = 305$ K was observed. The presence of this maximum is associated with the suppression of magnetic moment fluctuations by the field, which are maximal in the region of magnetic phase transition. This maximum is characteristic of manganites with colossal MR [2, 14]. The field dependence of MR in this region shows a linear dependence without hysteresis and saturation in fields up to 8 kE (Fig. 1c). At the same time, for the LBM/YSZ film, the MR dependence on T shows only a shoulder near T_C against the background of a continuous increase in MR with decreasing temperature (Fig. 1a). The continuous increase in MR is associated with the tunneling of spin-polarized electrons through the boundaries of structural domains.

The maximum value of the tunnel MR obtained was approximately 27% at $T = 13$ K (not shown in the figure). Estimation of the degree of electron spin polarization P at $T \sim 0$ K using the expression $\frac{\Delta\rho}{\rho} = 2P^2 / (1 - P^2)$ gives a value of $P \approx 0.36$, which is close to the value of P for La_{0.8}Ag_{0.1}MnO_{3+ δ} /ZrO₂(Y₂O₃) films with a variant structure [15]. The field dependence of low-temperature MR differs significantly (Fig. 1b). Thus, the negative MR sharply reaches saturation and gains almost 25% in magnetic fields greater than 2 kE, resulting in a butterfly-wing hysteresis characteristic of materials with tunnel MR with weak positive maxima in the fields of about 0.5 kE. The arrow in the figure indicates a feature associated with the colossal MR in the LBM/YSZ film. The hatched areas schematically reflect the regions of predominant contribution of the tunnel (I) and colossal (II) MR mechanisms, respectively. More detailed results of the analysis of the temperature and field dependence of MR and MRE in LBM films can be found in [9, 13, 16].

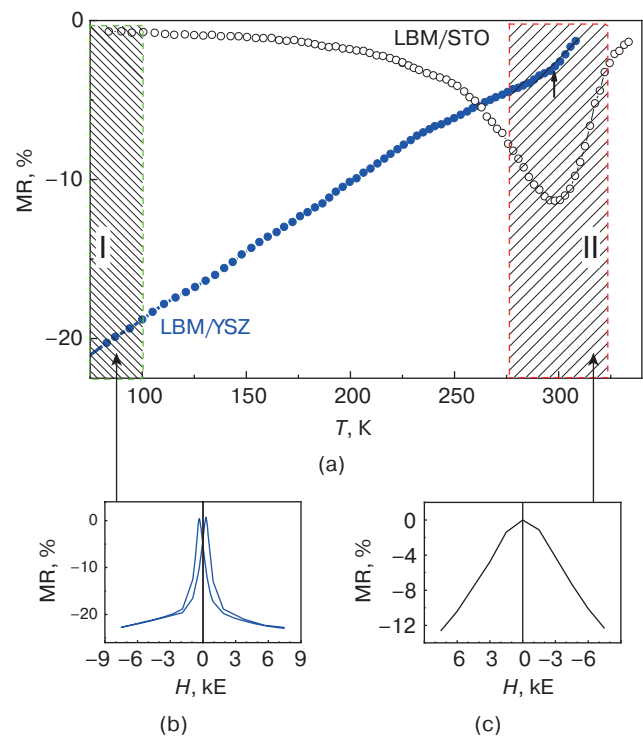


Fig. 1. Experimental characteristics: (a) temperature dependence of MR for LBM/STO and LBM/YSZ films at $H = 7.5$ kE; (b) and (c) are field dependencies of MR for LBM/YSZ and $T = 80$ K and LBM/STO films at $T = T_C$, respectively

SIMULATION RESULTS

The nanostructured LBM/YSZ film (with a variant structure) shows the presence of conductive ferromagnetic structural domains separated by weakly

conductive boundaries. When a constant external magnetic field is applied in the T_C region, a peculiarity associated with colossal MR in the domain volume is observed in the MR. At low temperatures, tunnel MR associated with charge carrier tunneling at domain boundaries is observed. In this regard, the peculiarities of the temperature behavior of MR in LBM/YSZ were considered in two main areas: near and well below the ferromagnetic ordering temperature (Curie temperature). At low temperatures, the $MR(T)$ dependence has the form of a quasi-linear function $T_{cr} \sim 250$ K. Upon an increase in temperature, it takes the form of a film without a variant structure (LBM/STO) and is described by a second-order polynomial function (or, in the first approximation, a linear function). To account for these two contributions, the proposed mathematical model is complemented with the coefficients of α and β . In terms of its physical meaning, the coefficient α takes into account the contribution of low-temperature (tunnel) MR: $\alpha = 1$ at $T < T_{cr}$ and $\alpha = 0$ at $T > T_{cr}$. This condition is satisfied by the expression of the form:

$$\alpha = \frac{1}{1 + e^{\frac{T - T_{cr}}{T_{cr}}}}, \quad (1)$$

where T_{cr} is the critical temperature at which a change in the MR mechanism is observed. Similarly, we can obtain a formula for the coefficient β of the high-temperature (colossal) MR:

$$\beta = \frac{1}{1 + e^{\frac{T_{cr} - T}{T_{cr}}}}. \quad (2)$$

A similar approach was used to describe the MR in the LBM/STO film (without a variant structure). The low-temperature region of the MR has a weakly linear section with a zero output near $T = 0$ K. Above 250 K, the $MR(T)$ dependence is described by a Gaussian function. A similar pattern was observed for $La_{0.7}Ca_{0.3}MnO_3$ films in [17].

The low-temperature region of the MR films was approximated as a quasi-linear section. The best match between the experimental and calculated curves is achieved by approximation with a second-order polynomial function (Fig. 2). However, the use of a polynomial significantly complicates the calculations; hence, a linear approximation was used. As a result, the general formula for describing the temperature dependence of MR ($\Delta\rho/\rho$) can be represented as:

$$\frac{\Delta\rho}{\rho} = \alpha A_1 T + \beta \left(A_1 T_{cr} + B_1 e^{-\frac{(T - T_C)^2}{2\sigma^2}} \right), \quad (3)$$

where A_1 is a coefficient describing the slope of the temperature dependence curve and determined by the growth conditions and composition of the sample (the magnitude is small and amounts to ≈ 0.01 , which is associated with a slight change in MR in the low-temperature region); B_1 is the MR value for a given film; σ is the root mean square deviation.

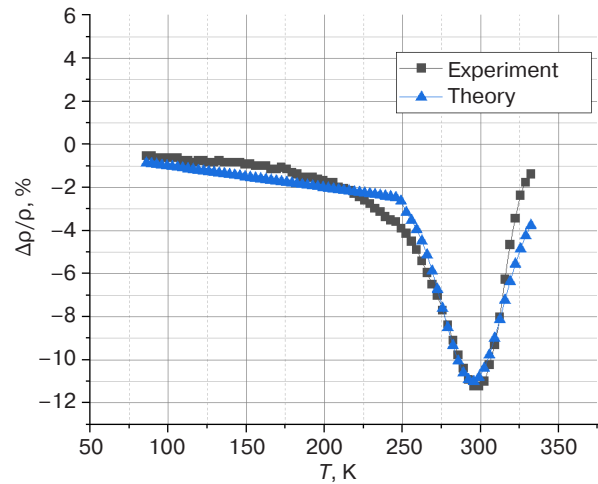


Fig. 2. Temperature dependence of MR ($\Delta\rho/\rho$) for LBM/STO film: experiment and calculation

In LBM films with a variant structure, the differences between low-temperature and high-temperature regions are not extremely significant (Fig. 3). The presence of structural domains in the film leads to a noticeable change in the $MR(T)$ curve and the absence of pronounced features near T_C . At the same time, the value of MR increases with a decrease in temperature, in contrast to films without a variant structure. A similar MR dependence was also observed for $(La_{0.65}Sr_{0.35})_{0.8}Mn_{1.2}O_{3\pm\delta}$ films with a variant structure [18]. In the low-temperature region, the dependence $\Delta\rho/\rho(T)$ is linear, becoming close to quadratic above the critical temperature (Fig. 3). In a first approximation, this feature near T_C can be considered insignificant. Then the dependence $\Delta\rho/\rho(T)$ will be quasi-linear. As a result, similar to the case considered earlier, we obtain a simplified expression for $\Delta\rho/\rho(T)$ for the LBM/YSZ film:

$$\frac{\Delta\rho}{\rho} = (\alpha + \beta)(A_0 + A_2 T), \quad (4)$$

where coefficient A_0 is the MR of the sample in terms of modulus (at $T = 0$ K), A_2 is the coefficient describing the slope of the curve (~ 0.086).

Thus, in doped LBM manganites, it became possible to account for the high- and low-temperature components of MR within a single empirical model. A simplified linear model that takes into account the critical temperature and characteristic values of colossal and

tunnel MR enables a good description of experimental data for both films with and without a variant structure. This model can be applied to explain the temperature characteristics of MR in a wide class of manganites of other compositions. The next step is to develop a theory of MRE in polycrystalline and composite manganite samples, taking into account the development of optical calculations of magnetic reflection [19, 20] and the results of this work. It is important to note that MRE, compared to the linear equatorial Kerr effect, is an even magnetization magneto-optical effect. In the IR region of the spectrum, taking into account interference effects [21, 22], the former can significantly exceed the latter, which makes MRE the most promising intensity effect from the point of view of fundamental and applied problems.

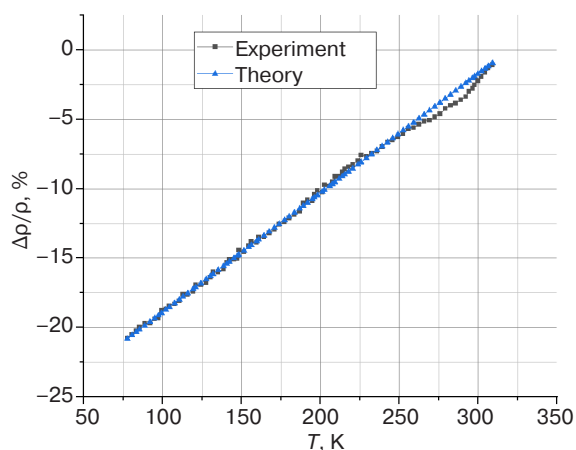


Fig. 3. Temperature dependence of MR ($\Delta\rho/\rho$) for LBM/YSZ film: experiment and calculation

CONCLUSIONS

The work shows that in the LBM/STO film, the MR has a pronounced maximum near the Curie temperature and is negligible in other areas. In the LBM/YSZ film, a strong low-temperature contribution to MR is observed, associated with a change in the high-frequency conductivity of the film in an external magnetic field due to the tunneling of spin-polarized electrons through the boundaries of coherent structural domains. For the first time, a unified empirical model that takes into account different MR mechanisms is proposed. Such an approach provides good agreement between experimental and calculated data for doped manganites. The proposed empirical approach can be used to model spin transport processes in strongly correlated magnetoresistive magnets, including at optical frequencies in the form of MRE.

It should be noted that the wide temperature window of MR existence in a film with a variant structure is important from the point of view of possible practical applications.

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Authors' contributions

T.N. Bakhvalova—literature review, computer modeling, discussion of results, writing the text of the article.

A.N. Yurasov—model development, discussion of results, writing the text of the article, editing the article.

M.M. Yashin—computer modeling, discussion of results, writing the text of the article.

V.A. Bessonova—conducting the experiments.

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About the Authors

Tatiana N. Bakhvalova, Lecturer, Department of Nanoelectronics, Institute for Advanced Technologies and Industrial Programming, MIREA – Russian Technological University (78, Vernadskogo pr., Moscow, 119454 Russia). E-mail: bahvalova@mirea.ru. ResearcherID ITW-2747-2023, Scopus Author ID 35145196400, <https://orcid.org/0000-0001-7595-785X>

Alexey N. Yurasov, Dr. Sci. (Phys.-Math.), Professor, Department of Nanoelectronics, Institute for Advanced Technologies and Industrial Programming, MIREA – Russian Technological University (78, Vernadskogo pr., Moscow, 119454 Russia). E-mail: alexey_yurasov@mail.ru. ResearcherID M-3113-2016, Scopus Authors ID 6602974416, RSCI SPIN-code 4259-8885, <https://orcid.org/0000-0002-9104-3529>

Maxim M. Yashin, Cand. Sci. (Phys.–Math.), Associate Professor, Department of Nanoelectronics, Institute for Advanced Technologies and Industrial Programming, MIREA – Russian Technological University (78, Vernadskogo pr., Moscow, 119454 Russia). E-mail: ihkamax@mail.ru. ResearcherID G-6809-2017, Scopus Author ID 57210607470, RSCI SPIN-code 2438-6135, <https://orcid.org/0000-0001-8022-9355>

Valentina A. Bessonova, Junior Researcher, M.N. Mikheev Institute of Metal Physics of Ural Branch of Russian Academy of Sciences (IMP UB RAS) (18, S. Kovalevskaya ul., Yekaterinburg, 620108 Russia). E-mail: valentina.a.bessonova@mail.ru. Scopus Author ID 55510248600, ResearcherID K-5887-2013, RSCI SPIN-code 7874-2518, <https://orcid.org/0000-0002-2163-4913>

Об авторах

Бахвалова Татьяна Николаевна, преподаватель, кафедра нанoeлектроники, Институт перспективных технологий и индустриального программирования, ФГБОУ ВО «МИРЭА – Российский технологический университет» (119454, Россия, Москва, пр-т Вернадского, д. 78). E-mail: bahvalova@mirea.ru. ResearcherID ITW-2747-2023, Scopus Author ID 35145196400, <https://orcid.org/0000-0001-7595-785X>

Юрасов Алексей Николаевич, д.ф.-м.н., профессор, кафедра нанoeлектроники, Институт перспективных технологий и индустриального программирования, ФГБОУ ВО «МИРЭА – Российский технологический университет» (119454, Россия, Москва, пр-т Вернадского, д. 78). E-mail: alexey_yurasov@mail.ru. ResearcherID M-3113-2016, Scopus Authors ID 6602974416, SPIN-код РИНЦ 4259-8885, <https://orcid.org/0000-0002-9104-3529>

Яшин Максим Михайлович, к.ф.-м.н., доцент, кафедра нанoeлектроники, Институт перспективных технологий и индустриального программирования, ФГБОУ ВО «МИРЭА – Российский технологический университет» (119454, Россия, Москва, пр-т Вернадского, д. 78). E-mail: ihkamax@mail.ru. ResearcherID G-6809-2017, Scopus Author ID 57210607470, SPIN-код РИНЦ 2438-6135, <https://orcid.org/0000-0001-8022-9355>

Бессонова Валентина Анатольевна, младший научный сотрудник, ФГБУН «Институт физики металлов имени М.Н. Михеева Уральского отделения Российской академии наук» (ИФМ УрО РАН) (620108, Россия, Екатеринбург, ул. Софьи Ковалевской, д. 18). E-mail: valentina.a.bessonova@mail.ru. Scopus Author ID 55510248600, ResearcherID K-5887-2013, SPIN-код РИНЦ 7874-2518, <https://orcid.org/0000-0002-2163-4913>

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