

03,05,13

Modification of the functional characteristics of InGaAs/GaAs/Al₂O₃/CoPt spin light-emitting diodes

© E.I. Malysheva, P.B. Demina, M.V. Ved[✉], M.V. Dorokhin, A.V. Zdoroveishchev, A.V. Kudrin, N.V. Baidus, V.N. Trushin

Research Institute for Physics and Technology, Lobachevsky State University of Nizhny Novgorod, Nizhny Novgorod, Russia

E-mail: ved@nifti.unn.ru

Received November 22, 2023

Revised December 13, 2023

Accepted December 14, 2023

Spin light-emitting diodes based on InGaAs/GaAs heterostructures and CoPt_x ferromagnetic contacts, where the composition varied within ($1 \leq x \leq 2.5$), were formed and studied. It is shown that varying the composition of the ferromagnetic contact provides control over the type of magnetic field dependence of the circular polarization degree of electroluminescence. Research has shown that control is carried out by modulating the magnetic characteristics of the films when varying the composition. The obtained result shows the possibility of controlling the magnetization of the contacts of a spin light-emitting diode using a magnetic field, which is useful from a practical point of view.

Keywords: magnetic thin films, spin injection, spin LEDs, III-V semiconductors.

DOI: 10.61011/PSS.2024.02.57913.261

1. Introduction

Spin light-emitting diodes (SLEDs) based on semiconductor heterostructures A^{III}B^V with a ferromagnetic (FM) metal injector is one of the basic elements of spintronics, in which spin-dependent phenomena such as electrical injection of spin-polarized carriers from magnetized FM electrode into a semiconductor structure, spin relaxation of carriers during transfer in the semiconductor material [1–3], spin precession in the magnetic field of a non-uniformly magnetized contact [4,5], as well as radiative recombination with the emission of partially circularly polarized light are realized. To date, the basic functions of the spin light-emitting diode were realized at room temperature in a series of papers [4–8]. Further development is associated with the search for ways of practical application of SLEDs, their integration into existing or new integrated optoelectronic circuits [9]. One of the problems solved within the framework of this aim is the search for ways to control such important functional characteristics of SLEDs as residual polarization (polarization in a zero magnetic field due to the residual magnetization of the injector) and switching magnetic field (from a state with circular polarization along right circle to a state with circular polarization along the left circle and vice versa). Control of these characteristics is achieved by modulating the properties of the ferromagnetic injector. The most common types of injectors based on CoFeB alloys have a fixed composition and a small number of degrees of freedom [8,10]. In contrast to them, in injectors based on alloys CoPt_x, considered in papers [4,5,11], the composition x can vary within wide limits, while the above parameters of the hysteresis loop also vary.

In this paper, the spin light-emitting diodes with injector based on alloys CoPt_x are considered, where the composition varied within the limits ($1 \leq x \leq 2.5$). It is shown that composition varying of the ferromagnetic contact provides control of the type of magnetic field dependence of the circular polarization degree of electroluminescent radiation, while the maximum value of the circular polarization degree (obtained in the maximum magnetic field) does not undergo significant changes. The obtained result seems to be important in view of constructing SLEDs with controlled parameters of operating magnetic fields.

2. Methods of creation and investigation of samples

The semiconductor part of the light-emitting diodes, which is a heterostructure with a quantum well In_{0.18}Ga_{0.82}As/GaAs, was grown by vapor-phase epitaxy from metal-organic compounds at atmospheric pressure of hydrogen on substrates n -GaAs. The distance from the quantum well to the surface of the structure (thickness of the coating layer) was 30 nm. After the end of epitaxial growth a thin 1 nm layer of dielectric — Al₂O₃, and metal film based on alloy CoPt_x were deposited on the surface of structures using electron beam evaporation in high vacuum in a single technological process. Thus, a Schottky contact was formed based on the ferromagnetic metal/tunnel-thin dielectric/semiconductor system. The metal contact was formed by alternate depositing of Co and Pt layers with varied thickness amounting to 10 periods. Technologically, the contact was a multilayer structure [Co(a)/Pt(b)]₁₀, where a and b — thickness of each layer in nm. The layer composition was specified by setting technological thickness

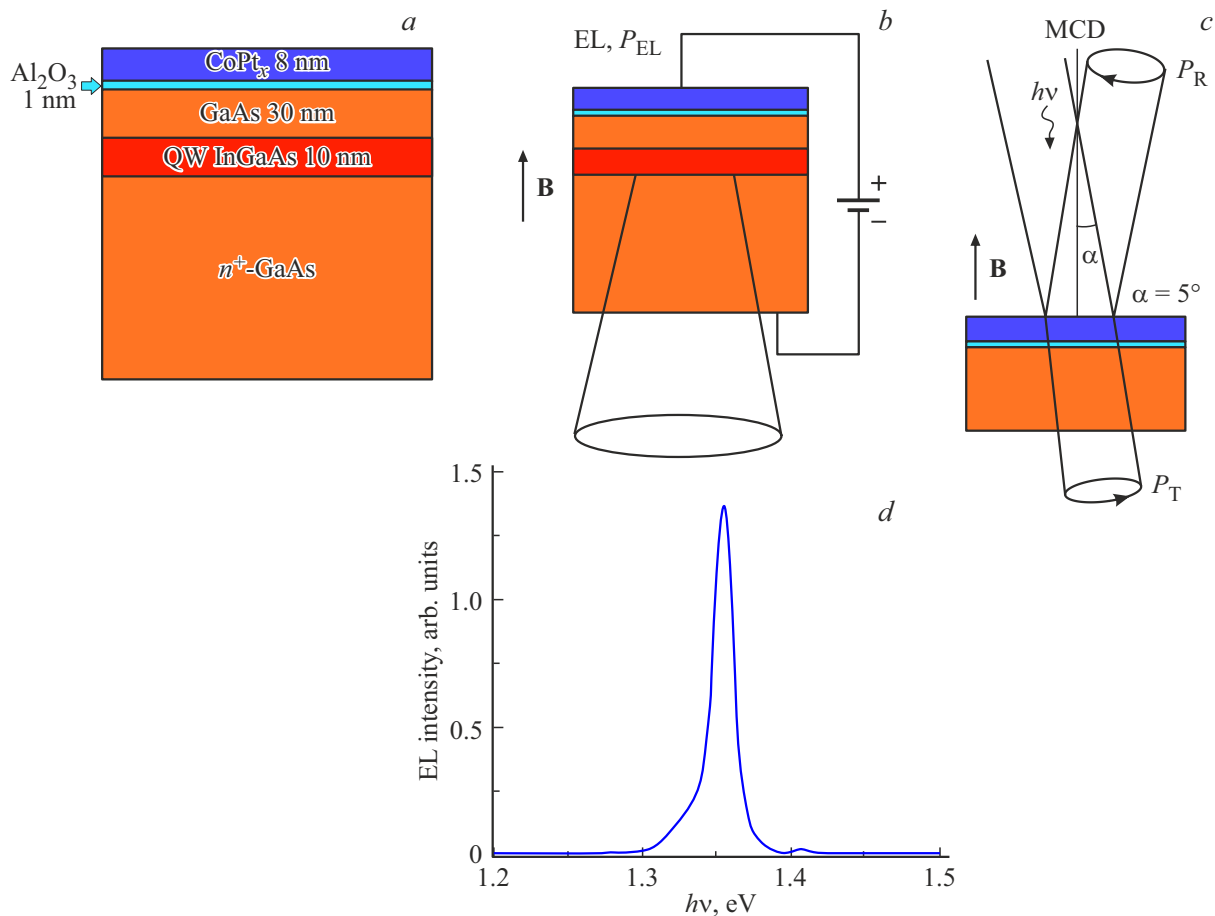


Figure 1. (a) Diagram of the sample for study; (b) geometry of measurements of the circular polarization degree of electroluminescence; (c) geometry of measurements of the circular polarization degree of light passed through the layer CoPt_x (P_T) and light reflected from the surface at close to normal incidence (P_R); (d) electroluminescence spectrum of spin LED measured at a temperature of 10 K and a current of 10 mA (current density 50 mA/mm^2).

values $x = b/a$, thickness was controlled using a quartz thickness sensor built into the technological equipment. The layer deposition temperature was 200°C , the total film thickness was $(a + b) \cdot 10 \text{ nm}$. To study magneto-optical effects on the formed films the polished surface of semi-insulating GaAs substrate was used as semiconductor part. The diagram of the samples for study is presented in Figure 1, a. A total of four samples were formed, with different compositions: sample 1 ($a = 2$, $b = 5$); sample 2 ($a = 3$, $b = 5$); sample 3 ($a = 4$, $b = 5$) and sample 4 ($a = 4$, $b = 4$). The composition was selected in accordance with previously obtained results, which showed the presence of perpendicular magnetic anisotropy [12] in films with such a composition, which is a necessary condition for obtaining circularly polarized electroluminescence [1].

The paper carried out studies of the phase composition of the formed films, as well as measurements the magnetic field dependences of the circular polarization degree of the electroluminescence of the formed spin light-emitting diodes. The phase composition and crystal structure of

the formed films were studied by X-ray diffraction using a Bruker D8 Discover X-ray diffractometer. The crystal structure of the GaAs substrates on which the multilayer metal film was formed corresponds to a single-crystal and is not considered in this paper. Phase analysis of the films was performed using the built-in software of the (DIFFRAC.EVA) device, using the database PDF-2. The magnetic field dependences of the polarization characteristics were measured in the magnetic field range 0–2000 Oe at a temperature of 10 K. To excite electroluminescence radiation a direct electrical bias was applied to the samples, and electroluminescence was recorded from the substrate side. The electroluminescence spectrum is presented in Figure 1, d. A peak with an energy of 1.35 eV was recorded in the spectrum, corresponding to the main transition in quantum well with a given content of In. When LED is introduced into magnetic field directed perpendicular to the surface, its radiation becomes partially circularly polarized due to the spin-polarized injection of carriers from the ferromagnetic CoPt contact into the quantum well. In this paper we measured the magnetic field dependences

of the circular polarization degree of electroluminescent radiation (P_{EL}).

To analyze the mechanisms of electroluminescence polarization, as well as to study the magnetic properties of the formed films, the circular polarization was measured upon transmission and reflection of unpolarized light from a CoPt film (the experimental diagram is shown in Figure 1, *c*). For measurements we used standard structures formed on *i*-GaAs substrates. During measurements the samples were irradiated with unpolarized light with wavelength corresponding to the main transition in the InGaAs quantum well. The magnetic field dependence of the circular polarization degree of radiation passed through a translucent film CoPt_x and transparent for the used wavelength substrate GaAs (P_T), as well as the circular polarization degree of P_R of radiation components reflected from the layer CoPt_x at light incidence close to normal (Figure 1, *c*).

The values of the circular polarization degree for all geometries were calculated using the formula

$$P_{EL(T,R)} = \frac{I(\sigma^+) - I(\sigma^-)}{I(\sigma^+) + I(\sigma^-)} \cdot 100\%, \quad (1)$$

where $P_{EL(T,R)}$ — the circular polarization degree of EL, transmitted or reflected radiation in percent, $I(\sigma^+)$ and $I(\sigma^-)$ — intensity of σ^+ and σ^- of polarized radiation. To calculate P_T and P_R , the relative intensity of the monochromatic radiation of a halogen lamp was measured. To calculate P_{EL} the intensity of the diode radiation was used; in this case, the intensities were obtained by integrating the part of the spectrum corresponding to the main transition in InGaAs quantum well.

3. Experimental results and discussion

Figure 2 shows the X-ray diffraction spectra of the films studied for samples 1, 2 and 3. The spectrum for sample 4 is similar to those presented, but is not shown to avoid the Figure cluttering. The position of the main diffraction maximum (DM) in the spectra does not correspond to the lines for Co and Pt and known Co-Pt compounds. The analytical software interprets the indicated peaks as solid solution CoPt_x with a composition dependent on the thickness ratio. Note, that this spectrum is typical for most studied samples, the only difference being the precise position of the main diffraction maximum. A monotonic increase in the angular position of the DM with decrease in Co relative thickness can be seen. Thus, in the formed sample Co and Pt layers are significantly mixed with each other, and the film itself is mainly a solid solution with an average composition CoPt_x. The obtained result is consistent with previously obtained data [13].

When diodes are introduced into external magnetic field in the geometry shown in Figure 1, *b*, the electroluminescent radiation becomes partially circularly polarized. The dependencies of the circular polarization degree of EL on the magnetic field for all studied samples are shown in

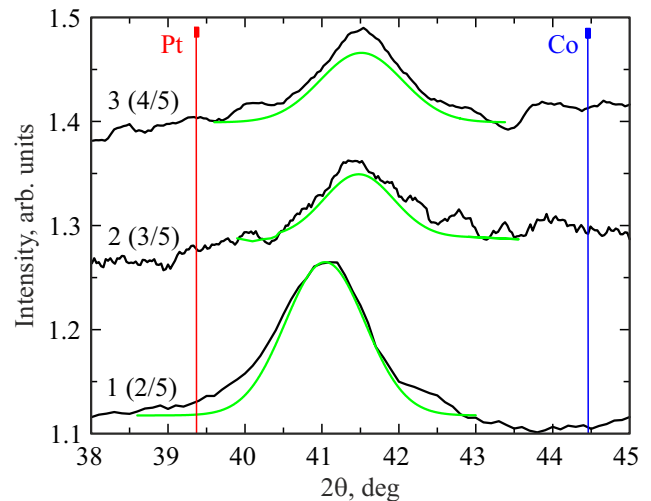


Figure 2. X-ray diffraction spectra measured in the area corresponding to the peak from the formed film. The numbers correspond to the samples labeling. Vertical lines indicate the angle values corresponding to the diffraction of Co and Pt.

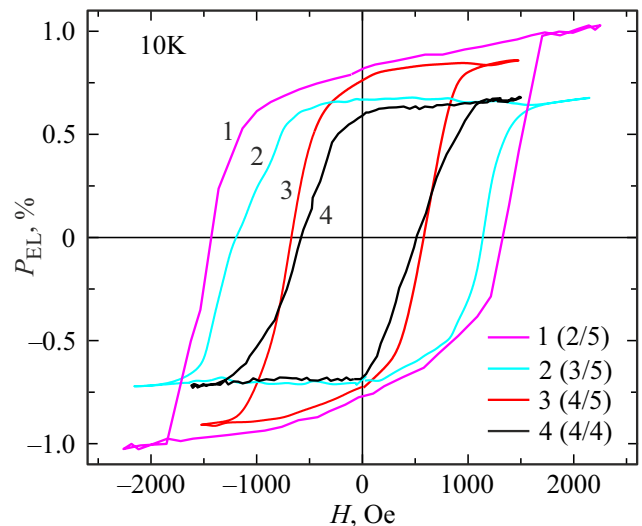


Figure 3. Magnetic field dependences of the circular polarization degree of electroluminescence, measured at temperature of 10 K and diode current of 10 mA (current density 50 mA/mm²). The sample number for each curve is indicated on the graph (composition values are given in parentheses).

Figure 3. All dependences P_{EL} describe a hysteresis loop reaching saturation in fields up to 550–1300 Oe. The type of dependences $P_{EL}(H)$ is determined by the magnetic field dependences of the magnetization of the studied layers CoPt_x, which clearly demonstrates the similarity of the hysteresis loops of circular polarization of EL ($P_{EL}(H)$) and polarization of light $P_T(H)$ passed through the magnetic layer (dependences $P_T(H)$ are presented in Figure 4). For both cases the magnitude of the coercive field (H_c), as well as of the saturation magnetic field (H_s) depend on the selected composition. The minimum values H_c and

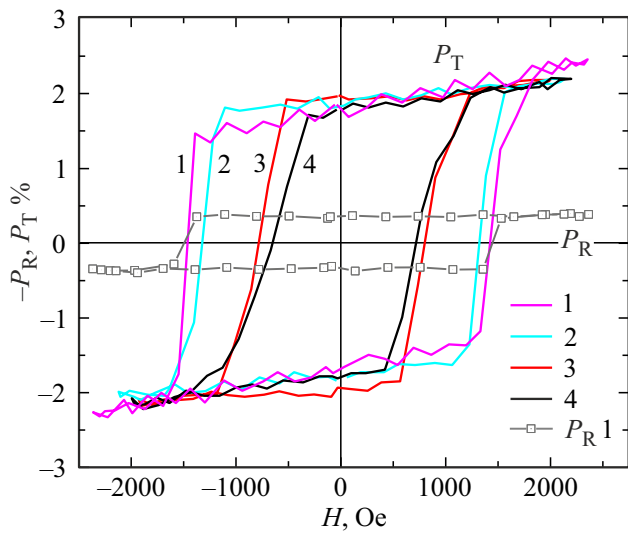


Figure 4. Magnetic field dependences of the circular polarization degree of transmission (P_T — lines) and reflection (P_R — dots), measured at temperature of 10 K. Sample number for each curve is given in graph.

H_s were obtained for the sample with the largest Co layer thickness ($a = 4$), while Pt content varying has little effect on these parameters. As Co thickness decreases, the values H_c and H_s monotonically increase to 1300 and 1700 Oe, respectively.

Thus, varying the thickness of Co layers in multilayer structure makes it possible to effectively control the remagnetization parameters and set the magnetic fields for switching circular polarization. The degree of EL polarization at magnetization saturation (P_{ELS}) non-monotonically depends on Co thickness and varies from sample to sample in the range of 0.6–1%. In contrast to P_{ELS} the polarization of light when radiation passes through the film P_{TS} varies slightly from sample to sample (Figure 4) and amounts to $\sim 2\%$ for all samples.

Summary data on the circular polarization degree measured in various geometries for the studied samples are presented in the Table.

Values of the circular polarization degree of electroluminescence, as well as the circular polarization degree of transmitted and reflected light, measured in magnetic field corresponding to magnetization saturation (P_{ELS} , P_{TS} and P_{RS} respectively) for the studied samples

No	Composition (a/b)	P_{ELS} , %	P_{TS} , %	P_{RS} , %	H_c , Oe	H_s , Oe
1	2/5	0.99	2.00	-0.29	1350	1700
2	3/5	0.67	2.00	-0.35	1150	1650
3	4/5	0.84	2.00	-0.30	570	1150
4	4/4	0.64	2.00	-0.44	500	1100

Let us proceed to discussion of the experimental results obtained. The change in the parameters of the hysteresis loop with varying Co content in the film CoPt_x is associated with change in the magnetic anisotropy constant [14,15], which value depends on the film composition. Note that previously perpendicular magnetic anisotropy was recorded for compositions in the range ($a = 1-4 \text{ \AA}$, $b = 4-5 \text{ \AA}$), when the composition deviates from the specified range, the films are formed with easy magnetic axis in the plane [13–15]. Thus, the film composition significantly affects the magnetic anisotropy. Variation in the circular polarization degree of electroluminescence may be due to the following factors [1,5,16].

1. Equilibrium spin polarization caused by Zeeman splitting of levels in quantum well. According to the paper [5], the value of the equilibrium spin polarization linearly depends on the magnetic field, and in field of 2000 Oe the value of the equilibrium spin polarization does not exceed 0.2%. Due to the specific nature of the dependence of the equilibrium polarization, as well as due to its smallness this factor will be excluded from consideration.

2. Injection of spin-polarized charge carriers from magnetized ferromagnetic contact into semiconductor and spin relaxation of injected carriers at the ferromagnet/semiconductor heterointerface and in the bulk of the semiconductor. The circular polarization degree of recombination radiation in quantum well as a result of spin injection is determined from the relation

$$P_{EL} = P_{fm} \exp(-x/l) / [1 + (\tau_s/\tau_R)], \quad (2)$$

where P_{fm} — spin polarization degree of carriers in ferromagnet [16], l — length of spin diffusion in semiconductor, τ_s — spin relaxation time, τ_R — life time relative to radiative recombination. Expression (2) is valid in the case of relatively small external magnetic field, and under the condition of high contact resistance at the ferromagnet/semiconductor interface, which is quite consistent with the $\text{CoPt}/\text{Al}_2\text{O}_3/\text{GaAs}$ system under consideration. The characteristic length of spin diffusion for holes in GaAs is $\sim 100 \text{ nm}$ [17]. The spin relaxation time of holes in similar $\text{InGaAs}/\text{GaAs}$ quantum well system was previously estimated as 6.5 ps at temperature of 1.4 K [18]. Also, expression (2) does not take into account spin relaxation at the ferromagnetic/semiconductor interface; this process will ensure a decrease in the P_{fm} factor.

3. Precession of injected spin-polarized carriers as they are transferred from the injector to the active region. This factor affects both the absolute value and the sign of the circular polarization degree; spin precession in spin light-emitting diodes was studied in detail in the papers [4,5];

4. Additional factors influencing the value of P_{EL} are the polarization of EL radiation upon reflection of unpolarized light from magnetic layer and upon light passing through magnetized ferromagnetic layer (P_R and P_T respectively). In the measurement geometry used (Figure 1, *b*) the detector registers only that part of the radiation that is

output from the substrate side, so the light polarization during transmission does not affect the measured value P_{EL} . The polarization of the radiation reflected from the ferromagnetic layer can contribute to P_{EL} , since part of the EL radiation is reflected from CoPt contact and output from the substrate side in the geometry of Figure 1, *b*. To estimate the polarization degree of the reflected radiation, appropriate measurements were performed. In Figure 4, the dots show the magnetic field dependence $P_R(H)$ for the sample 1, measured in the geometry shown in Figure 1, *c*. Similar to other characteristics, the type of dependence $P_R(H)$ is determined by the magnetization of the ferromagnetic contact, and the polarization value does not exceed 0.4%. The Table shows the values of P_{RS} at magnetization saturation. Due to the geometry of the experiment, the polarization of the reflected light has the opposite sign and, therefore, reduces the final value P_{EL} . The quantitative contribution of this effect to the value P_{EL} cannot be assessed due to the complexity of the intensity estimation of the light component reflected from the contact CoPt_x (it is necessary to take into account the transmittance, the possibility of multiple reflection, and the directional pattern of EL radiation). At the same time note that samples with the highest value P_{EL} are characterized by the smallest values P_R , which indirectly confirms the influence of the reflected wave on the measured value of the circular polarization degree, but does not explain the recorded differences in P_{EL} for different samples.

Let us consider the influence of the second and third of the listed factors on circularly polarized luminescence. The coincidence of the values of the circular polarization degree of light transmitted through CoPt_x indicates the similarity of the magnetic structure, since the degree of interaction of magnetic moments with the transmitted radiation is the same for all samples. Therefore, we can conclude that the value of the initial polarization P_{fm} is close for all compositions of magnetic films. Spin relaxation during transport of charge carriers in the spin injection mode does not differ, since the films are formed on the basis of the same semiconductor structure. Spin relaxation at the interface also cannot differ significantly due to the similarity of the properties of heterointerfaces for all films: in all cases, a layer of Al₂O₃ 1 nm thick was deposited on the semiconductor surface, and the first metal layer on the surface of Al₂O₃ was always platinum.

According to papers [4,5] the value P_{EL} is significantly influenced by the precession of spin-polarized carriers injected into the semiconductor from CoPt_x layer. It was previously shown that the cause of precession is the magnetic field of nonuniformly magnetized contact. The presence of inhomogeneities of structure and composition in the film creates an additional built-in magnetic field in the near-surface region, which has components in the direction perpendicular to the film normal. Due to the precession of carriers in this built-in magnetic field, as they are transferred to the active region, the average value of the spin polarization and, as a consequence, the circular

polarization degree of EL change. The magnitude of the built-in magnetic field and, accordingly, the final recorded value P_{EL} depend on the density of magnetic inhomogeneities. Presumably, exactly this parameter changes when the thickness of the layers of multilayer structure is varied, which determines the modulation of the value P_{EL} from sample to sample. Note that the modulation of the polarization degree is relatively small: the minimum value obtained for the sample 2 was 0.6%, the maximum for the sample 4 — close to 1%. The obtained values significantly exceed the level of experimental error in P_{EL} measurements, which is $\sim 0.05\%$.

4. Conclusion

Thus, the paper demonstrated the possibility to control the parameters of the magnetic field dependence of the circular polarization degree of electroluminescence in spin LEDs with multilayer magnetic contact CoPt_x. Control of the coercive field and saturation magnetic field within 500–1500 Oe is carried out by varying the thickness of Co layer (*a*). The paper did not reveal the influence of thickness of Pt (*b*) on the magnetic characteristics; nevertheless, Pt thickness affects the maximum value of the circular polarization degree of electroluminescence. It is assumed that the mechanism that controls the parameters of the hysteresis loop is change in the magnetic anisotropy constant; the mechanism responsible for the change P_{EL} is spin precession in the built-in magnetic field of the inhomogeneous magnetic film.

From a practical point of view, the obtained values of the circular polarization degree of EL significantly exceed the measurement error, which makes the formed diodes promising in optical information coding systems [1]. The ability to control the coercive field allows you to vary the required value of the magnetic field of switching of the spin light-emitting diode between two states of saturated magnetization.

Funding

This paper was supported by the Russian Science Foundation, project No. 21-79-20186.

Conflict of interest

The authors declare that they have no conflict of interest.

References

- [1] M. Holub, P. Bhattacharya. *J. Phys. D* **40**, R179 (2007).
- [2] *Opticheskaya orientatsiya / Pod. red. B.P. Zakharchenya, F. Mayer Nauka, L., (1989). (in Russian).*
- [3] A.G. Aronov, G.E. Pikus. *FTP* **10**, 6, 1177 (1976). (in Russian).

- [4] M.V. Dorokhin, M.V. Ved, P.B. Dyomina, A.V. Zdoroveishchev, A.V. Kudrin, A.V. Rykov, Yu.M. Kuznetsov. FTT **59**, *11*, 2135 (2017). (in Russian).
- [5] M.V. Dorokhin, P.B. Demina, A.V. Zdoroveishchev, S.V. Zaitsev, A.V. Kudrin. ZhTF **92**, *5*, 724 (2022). (in Russian).
- [6] S. Goel, N.H.D. Khang, Y. Osada, L.D. Anh, P.N. Hai, M. Tanaka. Sci. Rep. **13**, 2181 (2023).
- [7] G. Salis, R. Wang, X. Jiang, R.M. Shelby, S.S.P. Parkin, S.R. Bank, J.S. Harris. Appl. Phys. Lett. **87**, 262503 (2005).
- [8] Zhenhao Sun, Ning Tang, Shixiong Zhang, Shuaiyu Chen, Xingchen Liu, Bo Shen. Adv. Phys. X **8**, *1*, 2158757 (2023).
- [9] M. Mustaqeem, P.T. Chou, S. Kamal, N. Ahmad, J.-Y. Lin, Y.-J. Lu, X.-H. Lee, K.-H. Lin, K.-L. Lu, Y.-F. Chen. Adv. Func. Mater **33**, *19*, 213587 (2023).
- [10] S.H. Liang, T.T. Zhang, P. Barate, J. Frougier, M. Vidal, P. Renucci, B. Xu, H. Jaffres, J.-M. George, X. Devaux, M. Hehn, X. Marie, S. Mangin, H.X. Yang, A. Hallal, M. Chshiev, T. Amand, H.F. Liu, D.P. Liu, X.F. Han, Z.G. Wang, Y. Lu. Phys. Rev. B **90**, 085310 (2014).
- [11] S. Mooser, J.F.K. Cooper, K.K. Banger, J. Wunderlich, H. Siringhaus. Phys. Rev. B **85**, 235202 (2012).
- [12] M.V. Dorokhin, A.V. Zdoroveyshchev, M.P. Temiryazeva, A.G. Temiryazev, P.B. Demina, O.V. Vikhrova, A.V. Kudrin, I.L. Kalentyeva, M.V. Ved, A.N. Orlova, V.N. Trushin, A.V. Sadovnikov, D.A. Tatarskiy. J. Alloys Compd. **926**, 166956 (2022).
- [13] M.V. Dorokhin, P.B. Demina, A.V. Zdoroveishchev, D.A. Zdoroveishchev, A.G. Temiryazev, M.P. Temiryazeva, I.L. Kalentyeva, V.N. Trushin. FTT **65**, *6*, 989 (2023). (in Russian).
- [14] I.H. Cha, T. Kim, Y.J. Kim, G.W. Kim, Y.K. Kim. J. Alloys Compd. **823**, 153744 (2020).
- [15] J.C.A. Huang, T.H. W, A.C. Hsu, L.C. Wu, Y.M. Hu. J. Magn. Magn. Mater. **193**, *1–3*, 166 (1999).
- [16] Concepts in spin electronics / Ed. S. Maekawa. Oxford University Press, N. Y. (2006).
- [17] M.V. Dorokhin. Dok. dis. Natsional'nyj issledovatel'skij Nizhegorodskij gos. un-t im. N.I. Lobachevskogo, Nizhnij Novgorod (2016). (in Russian).
- [18] T. Amand, B.Dareys, B.Baylac, X. Marie, J. Barrau, M. Brousseau. Phys. Rev. B **50**, *16*, 11624 (1994).

Translated by I.Mazurov